Vaughn, TL, et al. 2017 Comparing facility-level methane emission rate estimates at natural gas gathering and boosting stations. *Elem Sci Anth,* 5: 71. DOI: https://doi.org/10.1525/elementa.257

RESEARCH ARTICLE

Comparing facility-level methane emission rate estimates at natural gas gathering and boosting stations

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Coordinated dual-tracer, aircraft-based, and direct component-level measurements were made at midstream natural gas gathering and boosting stations in the Fayetteville shale (Arkansas, USA). On-site component-level measurements were combined with engineering estimates to generate comprehensive facility-level methane emission rate estimates ("study on-site estimates (SOE)") comparable to tracer and aircraft measurements. Combustion slip (unburned fuel entrained in compressor engine exhaust), which was calculated based on 111 recent measurements of representative compressor engines, accounts for an estimated 75% of cumulative SOEs at gathering stations included in comparisons. Measured methane emissions from regenerator vents on glycol dehydrator units were substantially larger than predicted by modelling software; the contribution of dehydrator regenerator vents to the cumulative SOE would increase from 1% to 10% if based on direct measurements. Concurrent measurements at 14 normally-operating facilities show relative agreement between tracer and SOE, but indicate that tracer measurements estimate lower emissions (regression of tracer to SOE = 0.91 (95% CI = 0.83–0.99), R^2 = 0.89). Tracer and SOE 95% confidence intervals overlap at 11/14 facilities. Contemporaneous measurements at six facilities suggest that aircraft measurements estimate higher emissions than SOE. Aircraft and study on-site estimate 95% confidence intervals overlap at 3/6 facilities. The average facility level emission rate (FLER) estimated by tracer measurements in this study is 17–73% higher than a prior national study by Marchese et al.

Keywords: methane emissions; gathering; boosting; natural gas; climate change; Fayetteville

Introduction

Efforts to understand methane (CH_4) emissions over the entire US natural gas supply chain are motivated by increased natural gas production and usage. Natural gas produces less CO_2 when combusted than coal or petroleum on a per unit energy basis, and is often suggested as a bridge fuel to a lower-carbon energy sector. However, total greenhouse gas impacts from natural gas use are highly dependent on the emission rate of un-combusted natural gas (Alvarez et al., 2012) because methane (CH_4), the primary component of natural gas, has a global warming potential 30 times higher than CO_2 on a 100 year time horizon (including oxidation to CO_2, but excluding climate-carbon feedbacks) (Myhre et al., 2013).

Gathering systems use pipelines to collect gas from upstream producing wells and deliver it to gathering and boosting stations (hereafter "gathering stations"). Gathering stations include natural gas compressor equipment that boosts the pressure of the produced gas from well pressure to the required downstream pressure. Compressors are typically driven by reciprocating engines fueled by a fraction of the gas passing through the station. Gathering stations may also be equipped with a range of supporting equipment, including dehydrators to remove water, treating equipment to remove undesirable gases, fuel conditioning systems, piping and control lines, metering, and other associated support equipment. Stations discharge gas to downstream pipeline networks that feed processing plants, transmission systems,

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distribution systems, or other gathering stations. S1 provides an equipment overview of a typical Fayetteville gathering station measured in this study. Emissions from gathering pipelines outside of the gathering station boundary are not considered in this study, but are addressed in a companion study performed during the same field campaign by Zimmerle et al. (2017).

Recent studies have used a variety of measurement methods to quantify CH_4 emission rates from natural gas systems (Brandt et al., 2016; Littlefield et al., 2017). Top-down methods that rely on atmospheric CH_4 mole fraction measurements alone may have difficulty attributing emissions to distinct sources (Pétron et al., 2014; Peischl et al., 2015), e.g., from biogenic or thermogenic sources at the regional scale, or from individual point sources at the facility scale (Brantley et al., 2014). Tracer-release measurements help address the latter issue by providing confirmation that CH_4 emissions are co-located with tracer gas release points; however, tracer measurements may not capture all emissions from a facility under certain conditions (Roscioli et al., 2015). A bottom-up estimate based on direct measurements of all components at a facility is possible in theory, but is challenging in practice due to the large number of potential sources, and difficulty of measuring or accurately estimating every source. Certain direct measurements may not be possible due to personnel safety or accessibility issues. A variety of factors may contribute to differences between top-down and bottom-up estimates of CH_4 emissions. Recurring themes in recent discussions and studies (Brandt et al., 2016; Littlefield et al., 2017) include temporal variability, unrepresentative emission factors or activity data, and skewed emission rate distributions.

The work presented here is part of a large, multifaceted field campaign designed to estimate methane emissions from all segments of the natural gas supply chain within the study area (production, gathering and boosting, transmission, and distribution systems). Multiple contemporaneous measurement methods were used to develop estimates of methane emissions at the device, facility and regional-scale to help reconcile top-down and bottom-up estimates. Here, we compare facility-level CH_4 emission rate (FLER) estimates developed from on-site, dual-tracer and aircraft methods made either concurrently (same day, same time) or contemporaneously (both made during this field campaign) at gathering stations. Estimates for unmeasured sources were based on other measurements made in this study, or prior measurements specific to equipment encountered where possible. Facilities with emission sources well outside the measurement capability of a method were excluded from comparisons.

Methods

Measurements for this study were collected during a four-week field campaign conducted in September–October 2015 in an eastern portion of the Fayetteville shale play (the "study area" S2). Fayetteville gas contains little hydrogen sulfide or other trace gas contaminants and few hydrocarbons heavier than ethane. Consequently, the gas is considered "sweet and dry" and requires only dehydration and acid gas removal prior to sale. There are no processing plants or storage facilities within the study area; gas discharged from gathering stations is routed directly to transmission or distribution systems. Ninety-nine (79%) of the 125 gathering stations in the study area are owned and managed by companies who provided site access and supported or participated in measurement activities. These companies ("study partners") also provided activity data, compressor engine exhaust stack test data, and insight into their operations that was critical to accurate modeling and interpretation of facility-scale emission estimates. S2 provides a complete description of the study organization and role of study partners. In short, the research team retained independent control of the study while benefiting from the working knowledge of owners and operators.

Field campaign

Three independent measurement teams using dual-tracer, aircraft, and on-site measurement methods measured 36 unique gathering stations during the field campaign (29% of the total gathering stations in the study area). Personnel from Colorado State University (CSU) coordinated measurement teams ("tracer", "aircraft", and "on-site"), ensured adherence to measurement protocol, and recorded facility operating conditions to help interpret measurement results. Tracer measurements were made in weeks 1–3 of the field campaign and on-site measurements were made in weeks 2–4. Thus, paired measurements (tracer and on-site, or aircraft and on-site) may be either concurrent or contemporaneous.

Each evening CSU personnel used surface wind forecasts to identify three to six gathering stations with suitable downwind road access for dual-tracer measurement the following day. Identified facilities were shared with study partners and measurement teams that evening or the following morning to streamline logistics. During the day, CSU and tracer personnel chose measurement locations from the list of identified facilities based on the observed local wind conditions, without input from study partners. Once present in the study area (week 2), the on-site team accompanied the tracer team to make concurrent measurements. After the tracer team departed the study area (week 4), the on-site team made contemporaneous measurements at facilities previously measured by tracer. The aircraft team, whose primary objective was to develop a regional mass balance of the study area (Schwietzke et al., 2017), measured gathering stations opportunistically throughout the field campaign. On flight days, CSU personnel coordinated aircraft measurements by providing the current location of ground-based measurement teams to the aircraft team via text message.

Measurements

Dual-tracer release

Tracer release is an established technique (Lamb et al., 1995; Allen et al., 2013; Mitchell et al., 2015; Subramanian et al., 2015) that estimates an emission rate by releasing a tracer gas at a known rate near an emission source and comparing mixing ratios of the tracer gas and the target analyte (CH_4 in this case) as measured in a downwind transect of the emission plume. The unknown emission rate of the target analyte is deduced from the known tracer release rate and the measured mixing ratio enhancements, that is, $\frac{Q_{CH_4}}{Q_{tracer}} = \frac{\Delta_{CH_4}}{\Delta_{tracer}}$. In this study, dual-tracer gases were released to provide an internal standard and empirical measure of tracer co-dispersion, along with an emission rate uncertainty for each measured plume, as in Roscioli et al. (2015). FLER estimates for each gathering station were based on two to fourteen individual dual-tracer plume transects made ~200–1200 m downwind of the target facility. During each transect, measurements of tracer gases (N_2O, C_2H_2) and CH_4 were made with Tunable Infrared Laser Direct Absorption Spectroscopy trace-gas monitors with sub-ppb precision. For a detailed discussion of the methods and results of the tracer-based FLER estimates used in this study, and the associated uncertainty, see Yacovitch et al. (2017). All tracer measurements ("tracer") were performed by Aerodyne Research Incorporated.

Aircraft facility measurements

A new aircraft mass balance method (Conley et al., 2017) was used to estimate CH_4 emissions from individual gathering stations. In this method, the aircraft circumscribed an imagined cylinder around the target facility and its emission plume while making in-situ measurements of CH_4 (Picarro 2301f), wind speed, pressure, and temperature. During measurements, the aircraft circled the target facility on a 900–1500 m fixed radius course, beginning 75–175 m above ground level. The aircraft continued upward, flying loops (10–33 in this study) in 100 m elevation increments until enhancements of CH_4 were no longer observed, which occurred at a height of 280–580 m in this study. This height defines the top of the imagined cylinder, while the bottom is defined by the ground; CH_4 flux through these surfaces can be neglected. Briefly, CH_4 flux through the wall of the imagined cylinder was estimated by first dividing the imagined cylinder into six altitude bins. Then a (discretized) path integral for each loop was computed from measured CH_4 concentration, air density, and wind speed normal to the flight path. Path integrals within each bin were averaged and multiplied by the bin height, resulting in a mass emission rate for each bin. Mass emission rates from each bin were then summed resulting in FLER estimate for the target facility. Aircraft measurements ("aircraft") and resulting emission estimates for this study were made by Scientific Aviation Incorporated, and a detailed discussion of the methods, results and uncertainty are presented in Conley et al. (2017). Note that this method targets individual facilities and differs from "area mass balance methods" utilized to quantify emissions from multiple facilities over larger spatial scales (Karion et al., 2013; Pétron et al., 2014; Schwietzke et al., 2017).

Study on-site estimate (SOE)

The study on-site estimate (SOE) is a comprehensive statistical estimate of CH_4 emissions from a gathering station comparable to tracer and aircraft FLER estimates. SOEs were developed from on-site direct measurements (ODMs) and engineering estimates in a Monte Carlo model (See S4). Engineering estimates were made for compressor engine crankcase vents, and glycol dehydrator ("dehydrator") regenerator vents. Emissions from unburned methane entrained in compressor engine exhaust (combustion slip) were estimated based on 111 measurements of representative engines made by measurement contractors for study partners in the year prior to the study (January to September, 2014). The SOE in this study does not include emissions from malfunctions, maintenance, or other intended or unintended operating conditions for which on-site teams had no means to measure or accurately estimate emissions ("immeasurable sources"). The presence of immeasurable sources prevented complete on-site measurements, and therefore tracer to SOE comparisons, at three facilities but did not affect aircraft to SOE comparisons. Each occurrence is described in Results and Discussion, and S5.

On-site Direct Measurements (ODMs) are the sum of all measurements made by high-flow samplers at a facility during the field campaign. ODMs refer to component or device-level measurements of flanges, unions, valve stem packing, rod packing vents, connectors, pressure regulators, tank vents, open-ended lines, pneumatic devices and controllers, and other sources expected to emit within the measurable leak rate of the high-flow sampler (0.05 SCFM-8 SCFM or equivalently 0.058-9.24 kg CH_4/h) (Bacharach, Inc., 2015). ODMs were made with the Bacharach Hi Flow® sampler and individual measurement uncertainties are assumed to correspond to the instrument accuracy (±10%) (Bacharach, Inc., 2015). Due to recent comments (Allen et al., 2015; Howard et al., 2015) about instrument accuracy and sensor transition failure, high-flow samplers were calibrated daily according to the manufacturers' recommendations (Bacharach, Inc., 2015). All ODMs made in this study are classified by major equipment category and component type in S3.

ODMs were made by AECOM, or a combined team including AECOM and study partner personnel; the same measurement protocol was followed by both teams (S2). On-site measurement teams used optical gas imaging (OGI) (FLIR® GF320, Opgal EyeCGas®), or a combination of OGI and laser methane detection (Heath Consultants RMLD-IS®) to locate emission sources.

Simulated Direct Measurements (SDMs) encompass the same source categories as ODMs and provide an estimated emission rate for sources where an ODM was attempted but out of range, or would have been attempted had the source been safely accessible. SDMs do not account for immeasurable sources. Emission sources observed but not measured due to inaccessibility or personnel safety concerns were documented as "observed not measured" and were accounted for in the Monte Carlo model by re-sampling from representative ODMs made in this study. Measurements with recorded values outside the measurable leak rate range of the high-flow sampler were also accounted for in the Monte Carlo model. A description of SDMs is provided in S4.

Simulated Combustion Slip accounts for the CH_4 component of un-burned fuel entrained in natural gas-fired compressor engine exhaust. Study partners provided compressor engine exhaust test data from 111 engines measured in the year prior to this study; combustion slip was not measured in this study. This sample represents an estimated one fourth of all gathering compressor engines within the study area. Tests were performed on engines located within the study area by measurement contractors using standard protocol (EPA Method 19 (US EPA, n.d.), EPA Method 320 (US EPA, n.d.)). Test data were provided for 24 Caterpillar® G3500 series engines and 87 Caterpillar® G3600 series engines. These engine series represent approximately 93% of all gathering compressor engines within the study area; all compressor engines at measured gathering stations belonged to one of these engine series. This ensured the applicability of exhaust test data and resulted in improved combustion slip estimates relative to compiled emission factors such as EPA AP-42 (US EPA, n.d.) (see S4). No uncertainty is provided for individual engine measurements; uncertainty is developed in the Monte Carlo model from the variation in measured combustion slip within an engine series. Compressor engine exhaust test data used in SOE development are provided in S3.

Simulated Crankcase Vents account for CH_4 vented from compressor engine crankcase vents because of imperfect piston ring sealing. Crankcase vents on compressor engines were not measured in this study, but were simulated based on a Caterpillar® crankcase ventilation application guide (Caterpillar, n.d.), which states that crankcase hydrocarbon emissions are normally 3% of total hydrocarbon exhaust emissions at engine mid-life, but could be as high as 20% due to engine wear. Crankcase emissions were calculated in the Monte Carlo model by multiplying combustion slip emissions by a factor drawn at random from a normal distribution (mean 3%, assumed standard deviation 2%). Johnson et al. (2015) measured crankcase vent methane emissions and combustion slip on Caterpillar® 3500 and 3600 series compressor engines and found crankcase vent emissions were 14.4% of combustion slip on average (range 7%–22%). Engine wear, which is a primary cause of increased crankcase vent emissions, cannot be readily deduced for engines at gathering stations in this study.

Simulated Dehydrator Regenerator Vents account for CH_4 emissions from dehydrator regenerator vents. All dehydrators at measured gathering stations employed flash tank vapory recovery systems, an emission control technique. Methane emissions from dehydrator regenerator vents were calculated in the Monte Carlo model using the emission factor for dehydrators with flash tank vapor recovery from a 1996 GRI study (Myers, 1996) (0.003 (–52%/+102%) kg/h CH_4 per MMscf per day of gas processed).

Alternative emission factors

Recent measurements of dehydrator regenerator (this study) and crankcase vents (Johnson et al., 2015) may indicate that these categories are conservatively estimated in SOEs. Direct measurements of regenerator vents on four dehydrators equipped with flash tanks and regenerator control devices were made in this study. Measurements were normalized by the gas throughput of each dehydrator, resulting in emission factors of 0.11, 0.28, 0.31, and 0.41 kg/h CH_4 per MMscf per day of gas processed (see S4). Emission factors based on these four measured units are one to two orders of magnitude greater than the GRI study emission factor for dehydrators with flash tanks and overlap or exceed those provided in the GRI study for dehydrators without flash tanks (0.14 (–50%/+101%) kg/h CH_4 per MMscf per day of gas processed). Flash tanks are an emission control technique that can reduce methane emissions by 90% (US EPA, 2006); the regenerator vent control devices on measured units do not affect methane emissions. GRI-GLYCalc nullifies flash tank emissions when the user indicates the presence of flash tanks on a simulated unit (see S4). Alternate method comparisons of tracer and aircraft to SOEs developed using these recent measurements of crankcase vents and dehydrator regenerator vents are provided in S6.

Methane emissions from acid gas removal (AGR) units are not included in SOEs. Two gathering stations with AGR units were measured in the study but were excluded from method comparisons because of incomplete measurement and immeasurable sources, respectively (S5). No measurements of AGR unit reboiler vents were made in this study.

Method comparisons

Comparisons are made between tracer and SOE, or aircraft and SOE. Tracer and SOE are compared at facilities measured concurrently in the absence of immeasurable sources. Aircraft and SOE are compared at facilities measured contemporaneously assuming the absence of immeasurable sources. Method comparisons were performed using the approaches of Bland and Altman (1986), and Neri et al. (1989). The approach of Bland and Altman ("difference plot") is generally accepted as the appropriate technique for analyzing method comparison studies (Hollis, 1996). It indicates the presence or absence of bias between methods, and provides an estimate of expected agreement between methods based on the sample population. The approach of Neri et al. is a variance-weighted least-squares ("VWLS") regression that minimizes the orthogonal distance between measurement data points and the line of best-fit, considering the error in both *x* and *y* data (see S7). Additionally, a bootstrap (Draper and Smith, 1998) was performed to estimate a 95% confidence interval on the VWLS regression slope. Ten-thousand new input datasets were constructed from SOE distributions output by the Monte Carlo model and normal distributions created from tracer or aircraft measurements and associated uncertainty. VWLS fits were performed on each re-sampled dataset; the 2.5th and 97.5th percentile of re-sampled VWLS regression slopes provide a 95% confidence interval on the original regression slope.

Ordinary least-squares regression is inappropriate because the regression slope depends on the choice of independent variable and both measurements are error affected. The slopes of ordinary least-squares regressions from either choice of independent variable may not bound the slope of the orthogonal regression (York, 1966). Orthogonal regression is required to predict results from one measurement method when measurements were made with another measurement method and both measurement methods are error affected (Altman and Bland, 1983).

Results and discussion

Figure 1 summarizes facility-level CH_4 emission rates and associated uncertainty for each measurement method at 17 gathering stations included in method comparisons. Tracer and SOE 95% confidence intervals overlap at 11 out of 14 facilities, while aircraft and SOE confidence intervals overlap at three out of six facilities. However, aggregate comparisons between methods across multiple facilities provides a more robust evaluation of method agreement.

Simulated Combustion Slip was the largest source category and contributed 78% to the cumulative SOE for the 17 facilities included in method comparisons shown in **Figure 1**. *ODMs* contributed 15%, *SDMs* contributed 5%, *Simulated Crankcase Vents* contributed 2%, and *Simulated Dehydrator Regenerator Vents* contributed less than <1% to the cumulative SOE.

Recall that facilities with immeasurable sources were excluded from method comparisons. Therefore, cumulative SOE contributions are representative of "normally operating" gathering stations within the study area, and are not representative of all gathering stations within, or outside of, the study area. The relative contribution of source categories would change if immeasurable sources were included, and combustion slip would no longer be the largest contributing source. For example, emission rates from tanks at two gathering stations were much greater than the measurement capability of high-flow samplers used by on-site teams and posed safety issues for personnel using direct measurement methods. These facilities are not included in method comparisons; each occurrence is described in S5. An SOE was calculated for all sources except tanks at these facilities, and this result was compared to tracer and aircraft measurements to estimate the magnitude of tank emissions.

At one facility, the tracer FLER (182 kg/h) was four times greater than the SOE (42 kg/h) leading to estimated tank venting emissions of 140 kg/h. The other was measured by aircraft three times (October 2, 3, and 14, 2015 resulting in aircraft provided FLER estimates of 276 (± 99 kg/h), 676 (± 119 kg/h), and 739 (± 107 kg/h). Tracer and on-site teams visited this facility to corroborate the aircraft measurements and tracer estimated 606 (± 278 kg/h) venting from a produced water tank (see S5). Emissions from these two tanks alone are greater than the cumulative SOE for facilities included in method comparisons. Tank venting emissions of a similar magnitude were also observed in Mitchell et al. (2015). Similar to Subramanian et al. (2015), not capturing the emissions from these two tanks results in an SOE 36% lower than tracer for facilities with valid on-site and tracer measurements (see S3). At

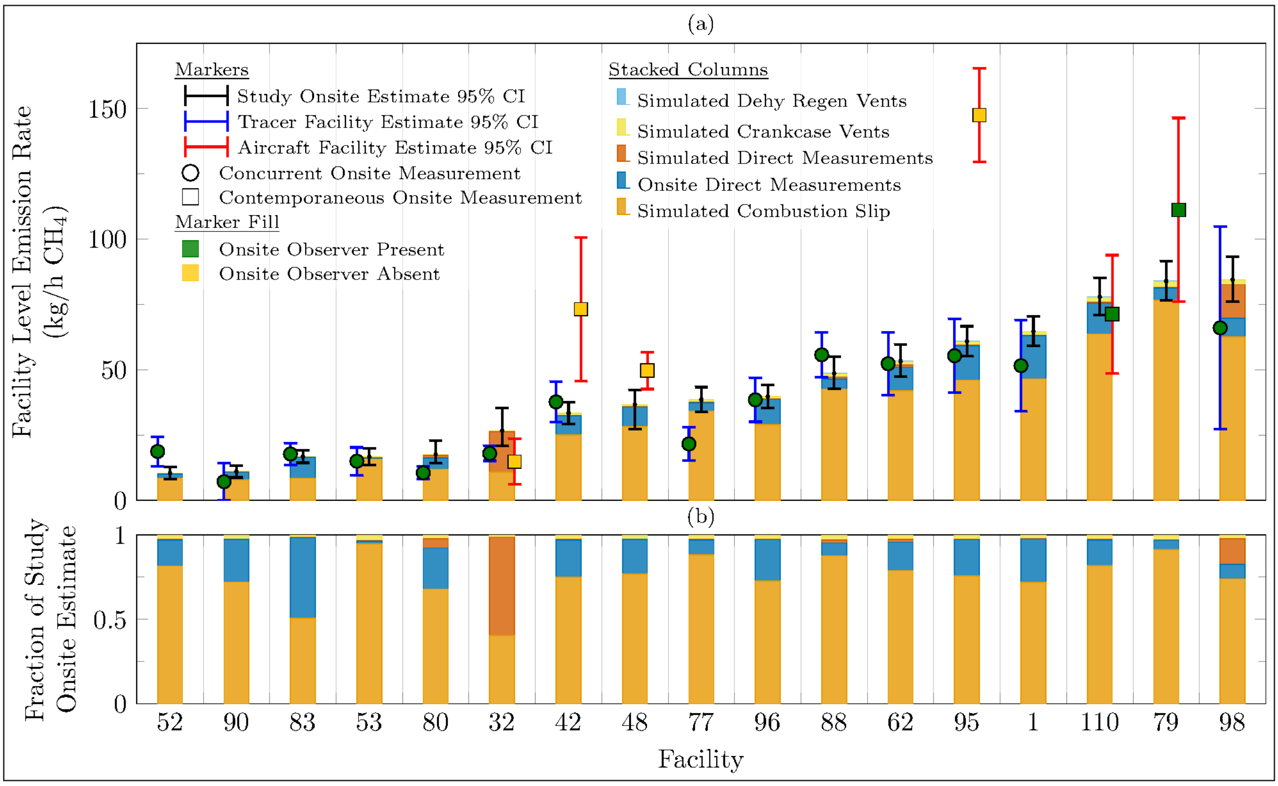

Figure 1: Facility-level CH_4 emission rate summary at all facilities included in method comparisons. Study on-site estimates (SOE) are the sum of on-site direct measurements plus engineering estimates for unmeasured sources (stacked columns, black error bars). Tracer (left mark, blue error bars) and aircraft (right mark, red error bars) are overlaid at facilities where these measurements were compared to SOEs. Marker shape and fill indicate same/different day and the presence/absence of on-site observers, which influence the comparability of measurements. Bottom panel illustrates the fraction of the SOE contributed by each component; combustion slip contributes more than half of emissions at 16 of 17 facilities and accounts for three quarters of cumulative SOE emissions for these 17 facilities. Conversely, on-site direct measurements (ODMs) contribute less than one fourth of emissions at 15 of 17 facilities and accounts for 15% of the cumulative SOE for these 17 facilities. DOI: https://doi.org/10.1525/elementa.257.f1

another gathering station, a compressor engine was shut down and several attempts were required to restart it. The tracer team saw increased and highly variable emissions from the facility during the restart; on-site teams had no means to quantify these emissions.

Dual-tracer release measurements of gathering stations were made previously in a national study by Mitchell et al. (2015); however, no on-site or aircraft measurements were made. These measurements were used by Marchese et al. (2015) to develop a national estimate of CH_4 emissions from gathering stations, which indicates an average emission rate of 53,066 scf/day (43 kg/h) per station. The influence of tank venting on the average FLER for 31 gathering stations measured by tracer in this study was evaluated in S8. Excluding tank venting emissions, the average FLER is 50.4 kg/h or 17% greater than the national average. Including tank venting emissions, the average FLER is 74.5 kg/h or 73% greater than the national average.

Tracer facility estimate and study on-site estimate comparison

When compared in aggregate by difference plot and variance-weighted least-squares regressions, tracer predicts lower FLER than SOE for 14 concurrently-measured gathering stations at the 95% confidence level (see **Figure 2**). In **Figure 2a** the difference of tracer and SOE is plotted against the uncertainty weighted mean of tracer and SOE. The mean of differences (termed "bias") is –4.9 kg/h indicating that tracer predicts lower FLER than SOE. A paired t-test is used to determine if the bias is significant. The shaded area in **Figure 2a** highlights the 95% confidence interval on bias. The confidence interval does not include $x = 0$, which indicates that the bias is statically significant at the 95% confidence level.

In **Figure 2b** a VWLS regression (dashed line) is performed on tracer and SOE. The slope of the regression (tracer = 0.91·SOE, $R^2 = 0.89$) is less than unity, indicating that tracer predicts lower FLER than SOE. The 95% confidence interval (shaded region) on the regression slope (tracer = 0.83·SOE to tracer = 0.99·SOE) does not include the line of equality ($y = x$), indicating that tracer predicts lower FLER than SOE at the 95% confidence level.

A fundamental assumption of the tracer method is that tracer gases released at a facility undergo the same dispersion as the target analyte emitted from the facility (CH_4 in this case). Buoyant combustion plumes may violate this co-dispersion assumption and result in a low-biased FLER. Roscioli et al. (2015) estimated worst-case recovery of combustion slip for several scenarios using Gaussian dispersion modeling with Briggs plume rise equations. They found that tracer may not recover up to 50% of the combustion slip plume when downwind measurements are made at distances of less than 1000 m. Recovery improves with increasing downwind distance. Yacovitch et al. (2017) found no evidence of plume rise at gathering stations measured in this study by comparing plume emissions to downwind measurement distance. However, this finding is not absolutely conclusive because downwind measurement distance varied little since it was dictated by the presence of roads (see Figure S14 in Yacovitch et al. (2017)). Conversely, while combustion slip estimates

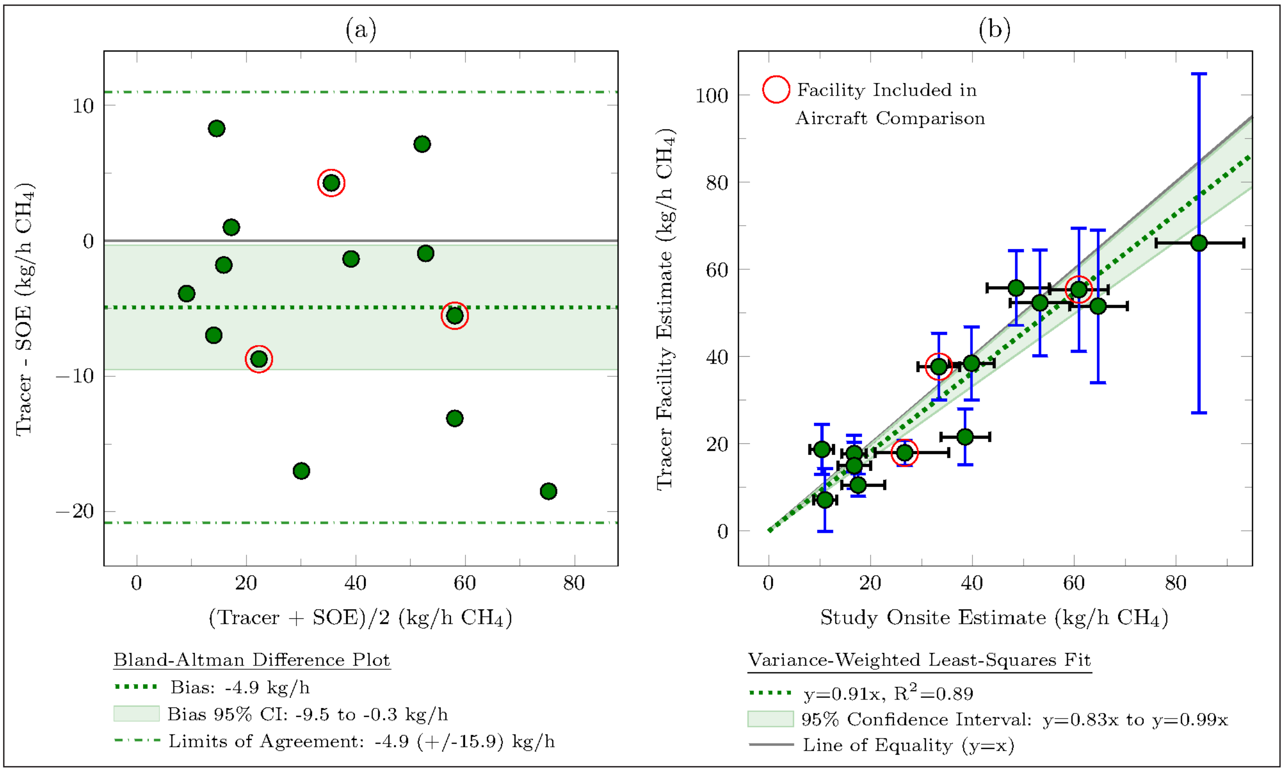

Figure 2: Tracer predicts lower facility-level CH_4 emission rates than study on-site estimates at the 95% confidence level using **(a)** Difference plots and **(b)** Variance-weighted least-squares regressions. DOI: https://doi.org/10.1525/elementa.257.f2

were developed from a robust dataset, contemporaneous combustion slip measurements were not performed and the possibility of a high bias in modeled combustion slip cannot be eliminated completely. Assessing plume recovery by releasing tracer gases directly into compressor engine exhaust stacks in conjunction with tracers placed on the ground in typical locations, as done in Lamb et al. (1995), may be a worthwhile addition to future tracer measurements at facilities with CH_4 emissions entrained in elevated, buoyant plumes. S6 discusses an alternate method comparison where SOEs were developed based on recent direct measurements of crankcase vents (Johnson et al., 2015) and dehydrator regenerator vents (this study). The results of this comparison also indicate that tracer predicts lower FLER than SOE (regression of tracer to SOE = 0.76 (95% CI = 0.69 to 0.83), R^2 = 0.92).

Aircraft facility estimate and study on-site estimate comparison

Aircraft and SOE are compared at 6 facilities measured contemporaneously by difference plot and VWLS regression. Confidence in this comparison is reduced relative to the tracer and SOE comparison, because measurements were made between 1 and 22 days apart. Additionally, the absence of immeasurable sources could not be confirmed when on-site observers were absent during aircraft measurements. Two facilities were measured with on-site observers present during both measurements, and four facilities were measured without observers present during aircraft measurements (S3).

Aircraft predicts higher FLER than SOE when compared by difference plot and VWLS regression, as shown in **Figure 3**. When compared by difference plot, aircraft is biased high relative to SOE (32.4 kg/h). However, the bias is not statistically significant because the 95% confidence interval includes $x = 0$; however, the bias is significant at the 90% confidence level.

In **Figure 3b** a VWLS regression (dashed line) is performed on aircraft and SOE. The slope of the regression (aircraft = 1.49·SOE, R^2 = 0.53) is greater than unity, indicating that aircraft predicts higher FLER than SOE. The 95% confidence interval (shaded region) on the regression slope (aircraft = 1.32·SOE to aircraft = 1.67·SOE) does not include the line of equality ($y = x$), indicating that aircraft predicts higher FLER than SOE at the 95% confidence level.

The observed bias in the aircraft and SOE method comparisons may be partly explained by the inability of aircraft to partition emissions from facilities within (or very near) the flight boundary. The loops flown by the aircraft covered significant area beyond the target gathering station and often included other facilities due to the geographic density of facilities in an active gas field. Post-campaign analysis of activity data indicated that emissions from nearby wells were included in aircraft measurements at least twice during the field campaign (S5). These field observations suggest that aircraft facility measurements should be utilized with caution when emissions from nearby facilities may confound results. S3 lists additional facilities within the aircraft sampling

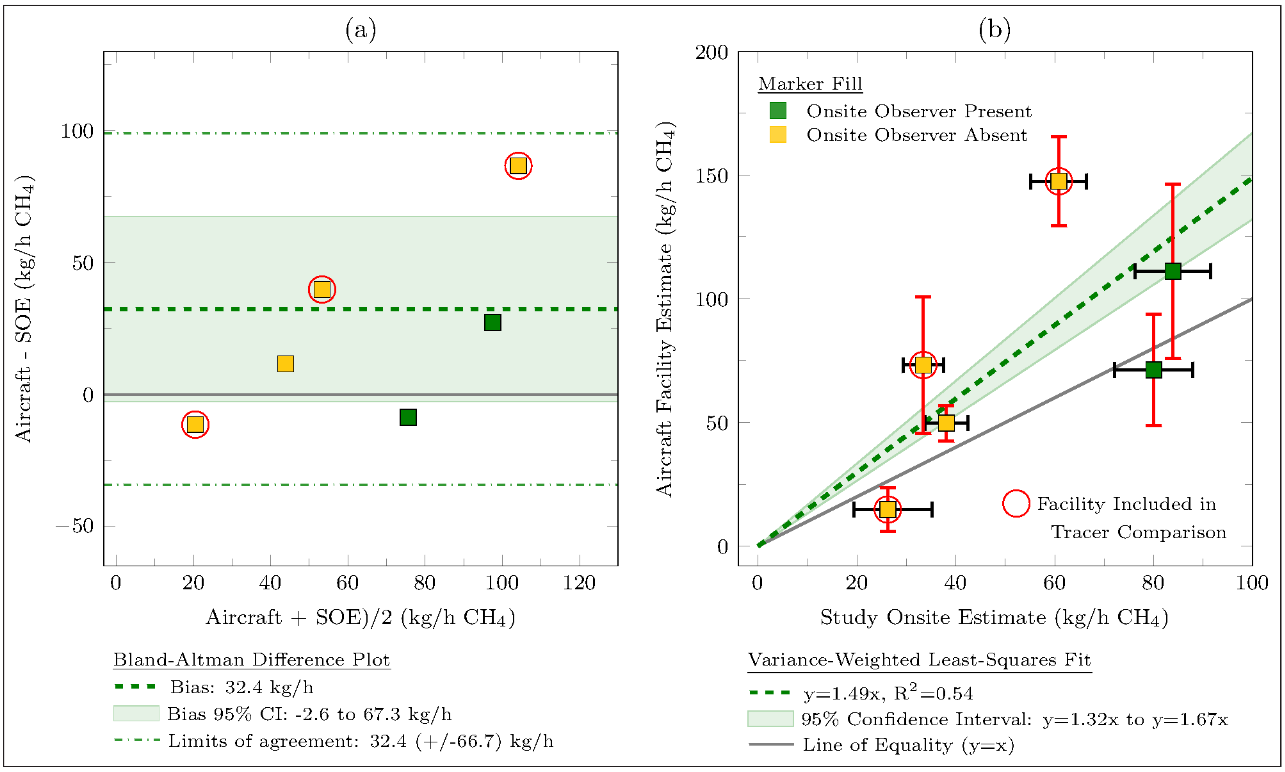

Figure 3: (a) Aircraft predicts higher facility-level CH_4 emission rates than SOE, but this result is not significant at the 95% confidence level. **(b)** Variance-weighted least-squares regression shows that aircraft predict higher facility-level CH_4 emission rates than study on-site estimates at the 95% confidence level. DOI: https://doi.org/10.1525/elementa.257.f3

footprint boundary at gathering stations measured in this study. Aircraft facility measurements may not suffer this limitation when measuring emissions from facilities without interfering sources, for example, well-isolated transmission and storage facilities, power plants, and landfills.

Additionally, the lowest flight altitude is limited by safety and regulations. Calculation of emission fluxes therefore must extrapolate emission rates from the lowest altitude loop to the ground level. This is generally the largest source of method uncertainty (Conley et al., 2017). Equipping the aircraft with tracer gas measurement capabilities and using tracer release gases could help to isolate emissions originating from the target facility from other nearby facilities, and may help quantify the effects of extrapolating calculated fluxes to ground level by evaluating tracer recovery rates under real field conditions.

S6 discusses an alternate method comparison where SOEs were developed based on recent field campaigns where crankcase vents (Johnson et al., 2015) and dehydrator regenerator vents (this study) were measured directly. The results of this comparison also indicate that aircraft predicts higher FLER than SOE (regression of aircraft to SOE = 1.22 (95% CI = 1.08 to 1.38), $R^2 = 0.54$).

Conclusions

This study provides the first contemporaneous, and in many cases, concurrent comparisons of facility-level CH_4 emissions from gathering stations utilizing both direct and atmospheric (downwind) measurement methods. SOEs were developed in a Monte Carlo model from on-site direct measurements and engineering estimates. Combustion slip contributed 78% to the cumulative SOE for the 17 facilities included in method comparisons and operating under normal conditions and was modeled using exhaust test data from 111 engines measured in the study area in the year prior to this study (January to September, 2014). Two engine series were tested, and all engines at measured gathering stations belonged to one of them. The quality and specificity of this test data improved the accuracy of study on-site estimates by providing more accurate quantification of combustion slip than would have been possible using EPA emission factors (See S4). The clarity of method comparisons was improved by on-site observers who documented maintenance, episodic, and malfunction events during measurements. This information was used along with study partner operational data to identify facilities for exclusion from method comparisons where measurement methods were affected unequally and bias would result.

This unique combination of circumstances greatly reduced the uncertainty for pair-wise comparison of the two primary methods for estimating CH_4 emissions from larger natural gas facilities – bottom-up estimates based on detailed, device-level, on-site measurements and top-down estimates based on downwind measurements of tracer and target-gas mixing ratio. The reduced uncertainties, in turn, provided a high confidence indication that while tracer and SOE show good correlation (r = 0.91), tracer methods may under-estimate emissions from this type of facility. As suggested by previous studies, one hypothesis for FLER underestimation by tracer relative to SOE is that CH_4 entrained in buoyant compressor engine exhaust plumes released from stacks above building height may not be fully recovered by tracer when releasing tracer gases at ground level only. Future tracer studies at gathering stations should test for co-dispersion by releasing tracer gases from compressor engine exhaust stacks.

Additional advancements are likely needed for aircraft-based measurements of individual facility emission plumes to be successful in areas where it is difficult to isolate the target facility from nearby sources. Advancements could include tracer gases released at the target facility to distinguish target facility emissions from nearby sources, and improved methods for estimating mass flux below the lowest altitude flown by the aircraft. Study methods and results also highlighted additional areas of interest to future field campaigns. At two gathering stations, produced water tanks were observed emitting at rates several times that of all other sources at the facility combined (S5). Knowledge of the cause, frequency and duration of these types of emission sources would provide a better understanding of their contribution to overall methane emissions from gathering stations. Direct measurements of regenerator vents on four dehydrators equipped with flash tanks and regenerator control devices were made in this study. All four showed emission rates greater than predicted by modeling software for dehydrators both with and without flash tanks, indicating the need for further empirical characterization of this source and validation of software used to predict methane emissions.

Data Accessibility Statement

Relevant data are included online in Supplemental Materials.

Supplemental Files

The supplemental files for this article can be found as follows:

- **S1.** Example Fayetteville Gathering Station with Compression and Dehydration (C/D Gathering Station). DOI: https://doi.org/10.1525/elementa.257.s1
- **S2.** Field Measurements and Protocol. DOI: https://doi.org/10.1525/elementa.257.s1
- **S3.** Data Tables: On-site measurements, Study On-site Estimate (SOEs), Tracer and Aircraft measurements. DOI: https://doi.org/10.1525/elementa.257.s1
- **S4.** Modeling Emissions: Study On-site Estimate (SOE) Component Categories. DOI: https://doi.org/10.1525/elementa.257.s1
- **S5.** Paired Measurements Excluded from Comparisons. DOI: https://doi.org/10.1525/elementa.257.s1
- **S6.** Alternate Method Comparisons Using SOEs Developed from Measured Dehydrator Regenerator Vents. DOI: https://doi.org/10.1525/elementa.257.s1
- **S7.** Variance-weighted least-squares regression. DOI: https://doi.org/10.1525/elementa.257.s1

- **S8.** Comparison to GHGI. DOI: https://doi.org/10.1525/elementa.257.s1
- **S9.** Simulated Direct Measurements (SDMs). DOI: https://doi.org/10.1525/elementa.257.s1

Acknowledgements
The authors thank Dag Nummedal, Laurie Williams, Cody Pickering, and Jean Opsomer for their contributions and acknowledge the helpful feedback of the technical review committee.

Funding information
Funding for this work was provided by RPSEA/NETL contract no 12122-95/DE-AC26-07NT42677 to the Colorado School of Mines. Cost share for this project was provided by Colorado Energy Research Collaboratory, the National Oceanic and Atmospheric Administration, Southwestern Energy, XTO Energy, a subsidiary of ExxonMobil, Chevron, Statoil and the American Gas Association, many of whom also provided operational data and/or site access. Additional data and/or site access was also provided by CenterPoint, Enable Midstream Partners, Kinder Morgan, and BHP Billiton.

Competing interests
The authors have no competing interests to declare.

Author contributions
- Contributed to conception and design: TLV, CSB, GH, GP, DZ
- Contributed to acquisition of data: TLV, CSB, TIY, JRR, SCH, SC, SS, GP
- Contributed to analysis and interpretation of data: TLV, CSB, TIY, JRR, SCH, SC, SS, GP, DZ
- Drafted and/or revised the article: TLV, CSB, GH, GP, DZ
- Approved the submitted version for publication: TLV, CSB, TIY, JRR, SCH, SC, SS, GAH, GP, DZ

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How to cite this article: Vaughn, TL, Bell, CS, Yacovitch, TI, Roscioli, JR, Herndon, SC, Conley, S, Schwietzke, S, Heath, GA, Pétron, G and Zimmerle, D 2017 Comparing facility-level methane emission rate estimates at natural gas gathering and boosting stations. *Elem Sci Anth,* 5: 71. DOI: https://doi.org/10.1525/elementa.257

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Knowledge Domain: Atmospheric Science

Part of an *Elementa* Special Forum: Oil and Natural Gas Development: Air Quality, Climate Science, and Policy

Submitted: 10 May 2017 **Accepted:** 29 August 2017 **Published:** 24 November 2017