

# Assessment of Methane Emissions from Oil and Gas Production Pads using Mobile Measurements

Halley L. Brantley,<sup>†,‡</sup> Eben D. Thoma,<sup>\*,†</sup> William C. Squier,<sup>†</sup> Birnur B. Guven,<sup>‡</sup> and David Lyon<sup>§</sup>

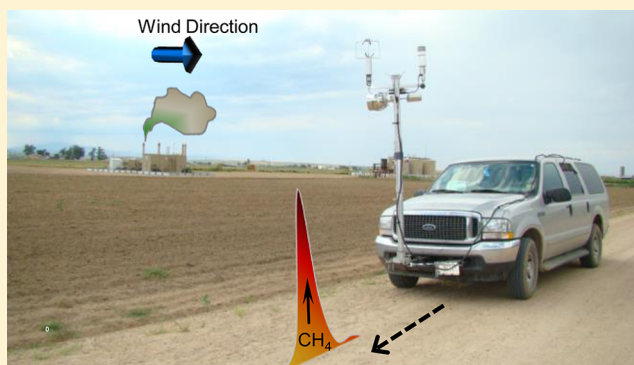
<sup>†</sup>Office of Research and Development, U.S. Environmental Protection Agency, 109 T.W. Alexander Drive, Research Triangle Park, North Carolina 27711, United States

<sup>‡</sup>Houston Advanced Research Center, The Woodlands, Texas 77381, United States

<sup>§</sup>Environmental Defense Fund, 301 Congress Ave., Suite 1300, Austin, Texas 78701, United States

## S Supporting Information

**ABSTRACT:** A new mobile methane emissions inspection approach, Other Test Method (OTM) 33A, was used to quantify short-term emission rates from 210 oil and gas production pads during eight two-week field studies in Texas, Colorado, and Wyoming from 2010 to 2013. Emission rates were log-normally distributed with geometric means and 95% confidence intervals (CIs) of 0.33 (0.23, 0.48), 0.14 (0.11, 0.19), and 0.59 (0.47, 0.74) g/s in the Barnett, Denver-Julesburg, and Pinedale basins, respectively. This study focused on sites with emission rates above 0.01 g/s and included short-term (i.e., condensate tank flashing) and maintenance-related emissions. The results fell within the upper ranges of the distributions observed in recent onsite direct measurement studies. Considering data across all basins, a multivariate linear regression was used to assess the relationship of methane emissions to well age, gas production, and hydrocarbon liquids (oil or condensate) production. Methane emissions were positively correlated with gas production, but only approximately 10% of the variation in emission rates was explained by variation in production levels. The weak correlation between emission and production rates may indicate that maintenance-related stochastic variables and design of production and control equipment are factors determining emissions.



## INTRODUCTION

Environmentally responsible development of oil and gas assets requires an understanding of atmospheric emissions of methane ( $\text{CH}_4$ ) and other organic pollutants as well as their potential impact on local and regional air quality and greenhouse gas budgets. Emissions are associated with many different processes in upstream (well development and production) and midstream (transportation and storage) oil and gas activities.<sup>1,2</sup> Although differing in profile, emissions occur in all phases of well construction, drilling, and completion, and continue as part of the ongoing production processes.<sup>3</sup> Oil and gas production pads (pads) typically consist of well heads, separation units, and storage tanks. Emissions from pads can be difficult to measure and model due to temporal variability and the large number of potential sources.<sup>4,5</sup> Pad emission profiles depend on a variety of factors including the geological formation, equipment design and maintenance state, and on operational procedures. For example, depending on engineering and control strategies, atmospheric-pressure condensate storage tanks are a significant potential source of emissions and can be challenging to measure.<sup>6,7</sup> Pad emissions can also vary over time as wells age and production levels and pressures change. Improving our understanding of emissions from production sites requires a

combination of approaches, including estimating emissions using engineering calculations for inventories,<sup>2,8,9</sup> direct measurements for refinement of emission and activity factors,<sup>10</sup> and new inspection techniques to inform departures from routine operations and support compliance activities.<sup>11</sup>

Direct (onsite) measurements can provide information on component-level emissions, but are resource intensive, requiring site access and special safety considerations. Furthermore, the high site-to-site variability decreases the probability of obtaining a representative sample from a small number of sites. To complement direct measurement approaches, a number of research groups are investigating the use of mobile inspection techniques to locate and assess emissions from off-site observing locations.<sup>4,12–14</sup> These emerging approaches vary with respect to execution requirements and emission estimation techniques; however, their mobile nature facilitates identification of unknown emission sources (e.g., pipeline leaks) and anomalous operating

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conditions (e.g., malfunctions). Unlike direct measurements, mobile approaches typically cannot isolate specific emitting components and are generally less precise than direct measures but are comparatively easier to implement, enabling emission assessments to be made at a greater number of locations on a more routine basis.

This paper describes a novel mobile inspection approach, EPA Other Test Method (OTM) 33A,<sup>17</sup> and its use to generate CH<sub>4</sub> emission rate data from oil and gas production sites in the Denver-Julesburg (DJ) Basin, the Barnett Shale, Pinedale, and Eagle Ford from 2010 to 2013. OTM 33A uses a combination of mobile sampling to identify sources and stationary measurements to quantify emissions. In addition to the analysis of repeated measurements at nine sites, the emission estimates from the OTM 33A field studies were compared with recent on-site studies led by the Eastern Research Group (ERG)<sup>15</sup> and Allen et al.<sup>16</sup> The ERG study,<sup>15</sup> conducted for the City of Fort Worth, TX, used both direct measurement and source estimation methods to characterize CH<sub>4</sub> and volatile organic compound emissions at 388 production sites containing wells, produced water storage tanks, separators, and compressors. Component-level source identification in the ERG study<sup>15</sup> was accomplished by infrared camera observations and direct source measurements were conducted using Hi Flow samplers (Bacharach Inc., New Kensington, PA), toxic vapor analyzers, and evacuated canisters. The measurements were used by the City of Fort Worth to evaluate the adequacy of setback provisions for pads and compressor stations. The results of the ERG study<sup>15</sup> indicated that compressors, leaking tank thief hatches, and pneumatic valve controllers are the most frequently encountered and significant emissions sources of CH<sub>4</sub>. Using similar on-site measurement techniques, Allen et al.<sup>16</sup> measured CH<sub>4</sub> emissions from 150 production sites in four regions of the United States to evaluate engineering estimates of CH<sub>4</sub> emissions from natural gas production that are used in national inventories. Their results indicated that emissions from pneumatics and equipment leaks were higher than estimated in the EPA greenhouse gas (GHG) emissions inventory.<sup>16</sup>

## MATERIALS AND METHODS

OTM 33A<sup>17</sup> is a mobile inspection approach used to locate sources and determine real-time emission rates with screening-level accuracy ( $\pm 60\%$ ), without the need for site access or location-specific modeling. The technique is applicable to select oil and gas sources such as roadway proximate pads located in relatively open areas. In addition to downwind vehicle access and favorable plume transport conditions required for all mobile assessment methods, the emission characterization portion of OTM 33A relies on relatively consistent meteorological conditions, obstruction-free line of sight observation, and a knowledge of the distance to the source.<sup>17</sup>

**Sampling Platform Design and Protocol.** The OTM 33A equipment configuration, further described in OTM33A Appendix A,<sup>17</sup> used either a G1301-fc cavity ring-down spectrometer (Picarro, Inc., Santa Clara, CA) or a GG-24-r off-axis integrated cavity output spectrometer (Los Gatos Research Inc., Mountain View, CA) as CH<sub>4</sub> concentration measurement instruments (CMIs). The mobile measurement platforms were sports utility vehicles containing the CMI, computer control system, and battery systems allowing engine-off instrument operation during stationary observations to prevent self-sampling of vehicle exhaust. The vehicles were fitted with rotatable front-mounted masts with a height of 2.7 m

allowing the CMI probe and meteorological instruments to be located away from the body of the vehicle. Primary wind field data were acquired using a model 81000 V Ultrasonic Anemometer (R.M. Young, Inc., Traverse City, MI). A collocated compact weather station (model AIO 102780, Climatronics Corp., Bohemia, NY) provided secondary wind data along with temperature, atmospheric pressure, and relative humidity measures. Location was recorded using a Hemisphere Crescent R100 Series GPS system (Hemisphere GPS, Calgary, AB Canada). A LabView (National Instruments, Inc., Austin TX) computer program time-aligned the data stream while allowing user control of the system.

The accuracy, linearity, and range of the CH<sub>4</sub> CMIs were confirmed in predeployment testing with in-field accuracy verified to be within  $\pm 5\%$  of actual using nominal 20 ppm CH<sub>4</sub> (air balance) gas standard challenges as per OTM 33 Section 9.4.<sup>17</sup> The CMI readings were not corrected for atmospheric water vapor (OTM 33A Appendix A)<sup>17</sup> which introduces an approximate 1.5% average negative bias to CH<sub>4</sub> emission determinations for the conditions encountered in this study.

For a typical pad assessment, emissions were located through downwind, drive-by inspection, keying on sharply elevated CH<sub>4</sub> spikes indicative of proximate source plumes. Maximizing real-time CH<sub>4</sub> concentrations measured by the CMI, the vehicle was positioned in the plume at a safe and appropriate downwind observing location with the probe facing the source, and the engine was turned off. Distance from the measurement vehicle to the emission source ranged from 10 to 200 m with an average distance of 57 m. Data were acquired for a 15 to 20 min time period with the vehicle remaining stationary. Auxiliary data from infrared cameras (FLIR Systems, Inc., Boston MA), when available, helped identify the source location, facilitating laser rangefinder measurements of the distance from the mobile platform to the source. Distances were later confirmed through Google Earth images coupled with wind-concentration rose data. The vehicle was positioned to minimize line-of-sight wind flow obstructions.

Emission rate estimates were calculated using a point source Gaussian (PSG) approach with a custom MATLAB (MathWorks, Natick, MA) analysis program (OTM 33A Appendix F1).<sup>17</sup> This approach relies on variations in wind direction to move the plume around the observation location in three dimensions; further assumptions include a point source and Gaussian plume dispersion. The analysis software time-aligned the measurements to correct for sampling line delay, rotated the 3-D sonic anemometer data to polar coordinates centered on the predominant wind direction, and binned the CH<sub>4</sub> concentrations by wind direction data in ten degree increments. The results were fitted with a Gaussian function to determine the average peak CH<sub>4</sub> concentration in the plume. Background concentrations were determined by the program during time periods with no plume-probe overlap (OTM 33A Section 8.7).<sup>17</sup> The program calculated the representative atmospheric stability indicator (ASI) from an average of the turbulence intensity (TI), measured by the 3D-sonic anemometer and the standard deviation in 2-D wind direction ( $\sigma\theta$ ), acquired by the compact meteorological station. By defining a seven unit ASI scale with steps of equal increments ( $TI = 0.025$ ,  $\sigma\theta = 4.0^\circ$ ), an ASI value for each measurement was assigned which ranged from 1 ( $TI > 0.205$ ,  $\sigma\theta > 27.5^\circ$ ) to 7 ( $TI < 0.08$ ,  $\sigma\theta < 7.5^\circ$ ), roughly corresponding to the Pasquill stability classes A through D.<sup>18</sup> For the PSG emission estimate, the values of horizontal ( $\sigma_y$ ) and vertical ( $\sigma_z$ ) dispersion are determined

from an interpolated version of point source dispersion tables using the measured source distance and the ASI (OTM 33A Section 12, Appendix F1).<sup>17</sup> The PSG emission estimate ( $q$ ) is a simple 2-D Gaussian integration (no reflection term) multiplied by mean wind speed ( $u$ ) and the peak concentration ( $c$ ) determined by the Gaussian fit: ( $q = 2\pi \cdot \sigma_y \cdot \sigma_z \cdot u \cdot c$ ).<sup>17</sup>

**Method Validation Using Controlled Release Experiments.** A set of 107 controlled CH<sub>4</sub> release experiments were conducted to investigate data quality indicators and the expected accuracy range for the PSG approach in relatively obstruction-free, open areas as encountered in this study (OTM 33A Section 9).<sup>17</sup> The experiments used single point releases from slightly dispersed, mass flow-controlled cylinders of 99.9% CH<sub>4</sub>, performed at a variety of site locations, observation distances, and under a range of atmospheric conditions. Release rates ranged from 0.19 g/s to 1.2 g/s with 60% at approximately 0.6 g/s. Based on these experiments, a primary set of three data quality indicators was identified: (1) fitted peak CH<sub>4</sub> concentration centered within  $\pm 30$  degrees of the source direction; (2) an average in-plume concentration greater than 0.1 ppm; and (3) a Gaussian fit with an  $R^2 > 0.80$ . The plume centering indicator helps ensure the identity of the upwind source and can protect against off-axis interfering sources and poor plume advection conditions. The concentration limit helps protect against insufficient plume transport and the  $R^2$  indicator helps identify interfering sources and obstructed wind flow conditions (non-Gaussian transport).

The percent error ([estimated emission rate-release rate]/[release rate]) of the controlled release experiments that met the data quality criteria ranged from -60% to 52% with 72% of the measurements within  $\pm 30\%$ . Without application of the data quality indicators, the set of release experiments produced accuracy values ranging from -87% to 184% of actual. The 184% overestimate was believed to be due to pooling and release under partially stagnant conditions and a trial wind variance indicator was developed for this case (not observed in field trials). Factors affecting accuracy can include insufficient plume advection and nonrepresentative concentration profiles caused by near-field obstructions or poor plume-probe overlap. Potential data quality indicators such as wind speed and plume concentration statistics are being investigated as part of OTM 33A method development.<sup>17</sup> For the current analysis, only measurements that met the three primary criteria were included (representing 77% of the controlled release measurements and 71% of the field measurements).

**Description of Field Studies and Production Data.** OTM 33A was used in eight two-week field campaigns in four oil and gas production basins: Colorado DJ Basin, July 2010 and 2011; Texas Barnett shale, September 2010 and 2011; Texas Eagle Ford Shale, September 2011; and Wyoming Pinedale, which includes the Pinedale Anticline and Jonah fields, June 2011, July 2012, and June 2013. Data sets for each individual basin were combined as the methods of data collection were similar, although there were some software and hardware improvements in later studies. All measurements were collected in the daytime on days with no significant precipitation.

Oil and gas production information for the counties sampled was obtained from DI Desktop (Drillinginfo, Austin, TX). Included in the data set were well type, operator, first production date, spatial coordinates of the well, and annual and monthly hydrocarbon liquids, gas, and water production levels. OTM 33A measurements were spatially matched with

production data using aerial imagery (Google Earth<sup>19</sup> and ArcGIS<sup>20</sup> base maps). When coordinates did not align with aerial imagery, additional data sets provided by the State of TX<sup>21</sup> and State of CO<sup>22</sup> were used to cross-reference location information. Monthly production values were available for 81% of the measurements. When monthly production was not available, annual values were converted to monthly estimates. The matched data set was analyzed using R<sup>23</sup> and ArcGIS 10.<sup>20</sup>

Both emissions estimates and production values were log-normally distributed and for this reason, data in figures are shown on a log scale. The mean and 95% CI of the log-transformed data were calculated using a nonparametric bootstrap<sup>24,25</sup> and then transformed back into the original scale. The nonparametric bootstrap involved resampling with replacement 1000 times, the mean of each of the samples was taken and the 95% CIs were calculated from the resulting normally distributed means. The nonparametric bootstrap was chosen because it does not assume the underlying data comes from a normal distribution. To compare OTM 33A emissions estimates with the direct measurement studies conducted by ERG<sup>15</sup> and Allen et al.,<sup>16</sup> direct measurements were converted from CH<sub>4</sub> scfm into g/s using a molar volume of 40.87 mol m<sup>-3</sup> and summed by site. Measurements from the ERG study<sup>15</sup> were matched with the corresponding monthly production values from DI Desktop (Drillinginfo, Austin, TX) based on the recorded Entity ID. Production values for the sites measured by Allen et al.<sup>16</sup> were reported by the well operators to the study team.

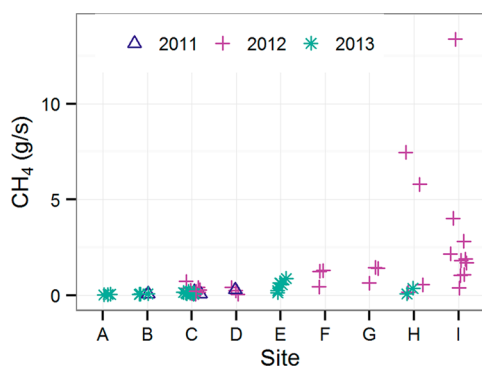
## ■ RESULTS AND DISCUSSION

**Description of Sites with Repeat Measurements.** The OTM 33A mobile inspection approach was used to identify and assess CH<sub>4</sub> emissions from roadway proximate well pads with an average in-plume concentration enhancement over background >0.1 ppm. No attempt was made to measure or statistically account for well pads with apparently low (and thus difficult to measure) emissions. In many cases, infrared camera videos (examples in Supporting Information (SI) Supplemental B) acquired from off-site observing locations, simultaneously with the CH<sub>4</sub> measurements, helped to identify specific emission sources. Storage tank-related emissions were frequently observed. The emission rates and video examples presented here may not be representative of current conditions due to engineering advancements, changes in work practices, and the implementation of new state regulations.

To improve understanding of both technique and source variability, repeat measurements (three or more) were made at nine sites in the Pinedale Basin, with the number of measurements per site ranging from 3 to 21 (SI Table S1). The consistent winds and lack of obstructions in the Pinedale Basin create favorable conditions for OTM 33A. Measurements were made in different years at four of these sites (Figure 1), and the time between measurements ranged from <1 day to 732 days (SI Table S1). For sites A–G, the 95% CI for the geometric mean was less than 1 g/s while at sites H and I, large variations in emissions were observed, resulting in a CI  $\geq 2$  g/s (SI Table S1).

The results indicate that while relatively low emissions (<2 g/s) frequently persist over time, the larger emissions observed using OTM 33A are likely episodic in nature. One source of persistent low-level emissions observed with the infrared camera is believed to be a vented produced water tank at Site C (SI Video S1). Previous studies have shown that flashing





**Figure 1.** CH<sub>4</sub> emission rates (g/s) measured at repeated sites in Pinedale, WY by year.

from a condensate tank after a separator dump can result in episodic large emissions.<sup>6</sup> CH<sub>4</sub> emissions greater than 2 g/s were observed at 13% of the 210 unique sites measured. The variability of emission rates at sites H and I indicates that these larger emissions may be episodic events that cannot be used to infer annual emission rates without a greater understanding of their frequency and duration (Figure 1).

Site I was measured on four separate days in 2012. On each of the days, the emissions appeared to originate from the same tank. Infrared videos indicate that all of the emissions >3.0 g/s occurred during the time period that a thief hatch on a condensate tank was open (SI Video S4, Video S5, and Video S6). On the last day the site was measured, the thief hatch was closed and the measured emissions seemed to originate from a pressure relief device and were <3.0 g/s (SI Video S7).

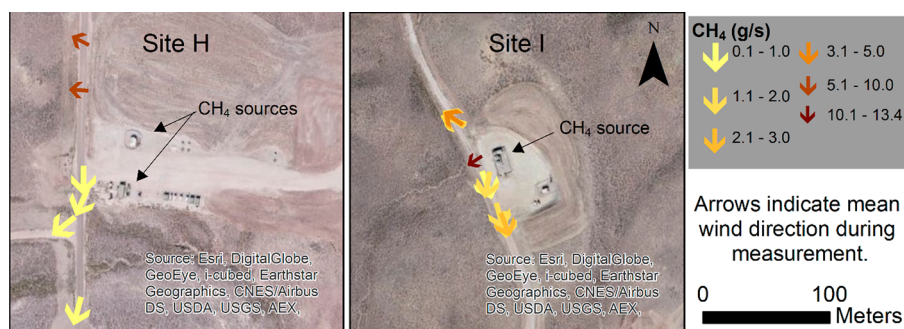
Another potential cause of variation in emissions levels is the variability in plume capture. Depending on meteorological conditions, the plume measured can include all of the sources on the pad or only some of the sources (Figure 2). Measurements were made at Site H on 3 days in 2012 and 1 day in 2013 (four and two independent emission measurements, respectively). The higher emissions observed were only present on one of the days in 2012 and originated from the tank on the north side of the pad (SI Video S2), whereas the smaller emissions seemed to originate from the southern edge of the pad (SI Video S3).

**Comparisons of CH<sub>4</sub> Emissions by Basin and with Direct Measurement Studies.** A total of 318 OTM 33A measurements that met the data quality criteria were collected. Of these measurements, 31 were excluded from the analysis because the measured emissions either did not originate from routine pad operations (e.g., evidence of active pad

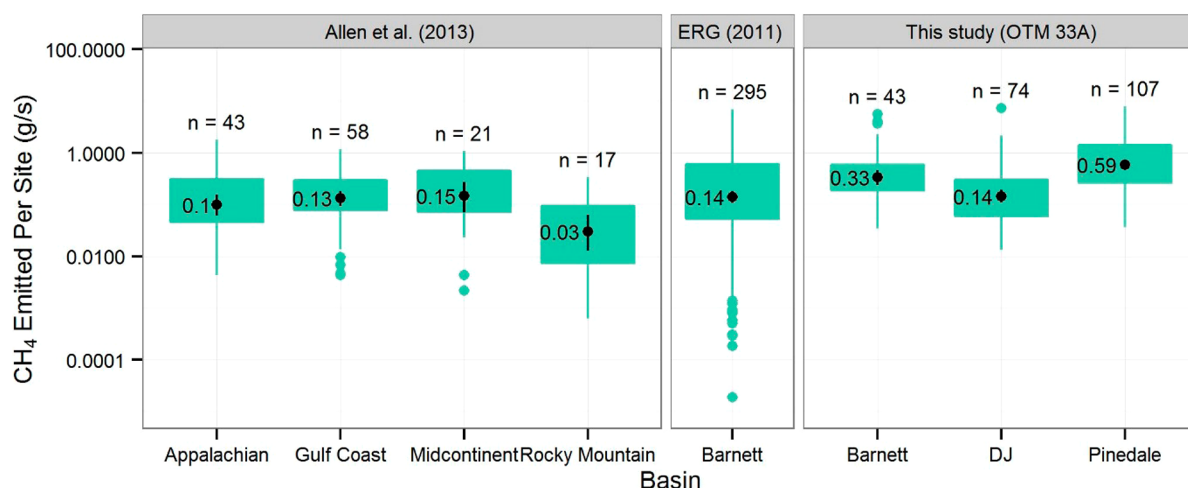
maintenance, pipeline leaks, gas processing plants, etc.) or no current production data were available, resulting in a total of 210 unique sites. The sites were classified into gas or oil pads based on the TX Railroad Commission definition of a gas well<sup>26</sup> (>100 Mscf of gas per barrel of hydrocarbon liquids). Gas pads constituted 93%, 2%, 75%, and 84% of the sites measured in the Barnett, DJ, Eagle Ford, and Pinedale basins, respectively. Methane emissions were averaged by site and month, resulting in a total of 228 combinations of emission and production values. Due to the small sample size in the Eagle Ford ( $n = 4$ ), these measurements were excluded from the basin comparison (Figure 3). CH<sub>4</sub> emissions were log-normally distributed with geometric means and 95% confidence intervals (CIs) of 0.33 (0.23, 0.48), 0.14 (0.11, 0.19), and 0.59 (0.47, 0.74) g/s in the Barnett, Denver-Julesburg, and Pinedale basins, respectively. Emissions by basin were compared using a Kruskal–Wallis one-way analysis of variance test and pairwise Wilcoxon rank-sum tests and were found to be significantly different ( $p < 0.05$ ). The differences in emissions between basins are likely a result of a combination of factors, including but not limited to variations in gas and oil production, emissions control devices, and natural gas and oil composition.

The OTM 33A measurements were compared with the results of the direct measurement studies of routine pad operations conducted by ERG<sup>15</sup> and Allen et al.<sup>16</sup> (Figure 3). The studies encompass a range of pads that vary with respect to oil and gas composition, production levels, amount and type of production equipment, age, and emission control measures, resulting in a broad distribution of emissions. The mean of the CH<sub>4</sub> emissions measured using OTM 33A in the Barnett Shale, 0.33 (0.23, 0.48) g/s, is more than twice the mean of the emissions measured by ERG<sup>14</sup> 0.14 (0.11, 0.18) g/s. Nevertheless, the interquartile range of the OTM 33A measurements in the Barnett falls within the interquartile range of the ERG emissions estimates despite the differences in the measurement methods and the bias toward higher-emitting sites in the OTM 33A measurements.

Both onsite and remote measurement techniques can provide important information on emissions. Whereas direct measurements can accurately quantify component-level emissions, they are less amenable to locating and assessing malfunction-related or large short-term emissions such as condensate tank flashing. The measurements by Allen et al.<sup>16</sup> were limited primarily to equipment leaks, pneumatic controllers, and chemical injection pumps. Condensate tank emissions were measured at some sites but rarely could all of the emission points be accessed. In the ERG study,<sup>15</sup> due to



**Figure 2.** Map of repeated measurements at sites H and I. The directions of the colored arrows indicate mean wind directions and the locations indicate the locations of the mobile platform during the measurement.



**Figure 3.** Comparison of measured  $\text{CH}_4$  emissions per pad (g/s) from Allen et al.,<sup>16</sup> ERG,<sup>15</sup> and OTM 33A by basin. Boxes represent the 1st and 3rd quartiles of the data, while whiskers extend to the largest measurement that is within 1.5 times the interquartile range (IQR). Means and 95% CIs are shown in black and were calculated using a nonparametric bootstrap.

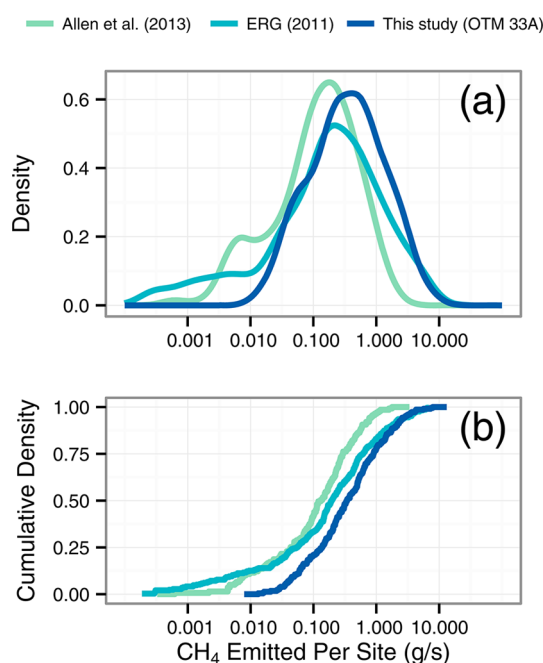
lack of condensate production, flash emissions were not represented. Although both studies measured fugitive component leaks, neither identified or measured potentially larger maintenance-related emissions (e.g., open thief hatch or failed pressure relief valve). In contrast, OTM 33A measurements generally represent an integrated plume including all potential sources on a pad. Supporting infrared camera footage from the OTM 33A studies indicated that emissions often originate from condensate storage tanks which have previously been shown to comprise a significant source<sup>6,5</sup> (SI Supplemental B). OTM 33A is also more likely to capture malfunction-related  $\text{CH}_4$  releases than direct measurement methods because of its mobile and off-site measurement capabilities.

However, the remote nature of the OTM 33A method and its application in these studies to only sites with downwind average in-plume concentrations greater than 0.1 ppm result in an effective lower sampling limit of approximately 0.010 g/s, compared with <0.001 g/s limits for the on-site measurement techniques (Figure 4a). As a result, the OTM 33A measurements only represent the upper end of the distribution in this comparison (Figure 4b).

**Comparison of Measurements with Production Values.**  $\text{CH}_4$  emissions from the direct measurement studies and OTM 33A were compared to monthly gas production using a linear regression on the log transformed data (Figure 5). Sites with gas production <1 Mscf/day or  $\text{CH}_4$  emissions <0.0005 g/s were excluded from the analysis (five sites in the ERG study<sup>15</sup>). Gas production values explained more of the variation in the OTM 33A measurements than the measurements from the on-site studies, although variation in gas production still accounted for only 8.3% of the total variation in emissions ( $R^2 = 0.083$ ) (Figure 5).

The OTM 33A  $\text{CH}_4$  emission estimates were also compared with hydrocarbon liquids and water production and the (arithmetic) mean age of active permitted wells on the site using Pearson correlation coefficients (Table 1) and a multivariate linear regression.

Approximately 23% and 15% of the pads measured using OTM 33A reported no hydrocarbon liquids or water production, respectively. To use these pads in the log-transformed model, pads with no reported oil or water production were assigned 0.01 bbl/day. Several values were

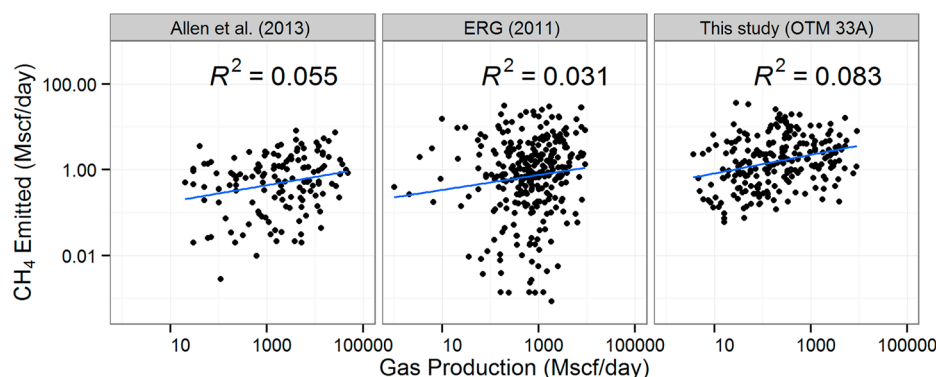


**Figure 4.** Density (a) and cumulative density (b) of measurements of  $\text{CH}_4$  emission rates (g/s) from this study (OTM 33A), Allen et al.,<sup>16</sup> and ERG.<sup>15</sup> Note the logarithmic x-axis.

tested and the choice of this value did not significantly affect the results. When considering the correlation between production and emissions individually,  $\text{CH}_4$  emissions were most strongly correlated with gas production ( $R = 0.29$ ).  $\text{CH}_4$  emissions were also positively correlated with water production, negatively correlated with mean age, and not correlated with hydrocarbon liquids production (Table 1).

A multivariate linear regression was conducted to determine the effect of gas and hydrocarbon liquids production and age of the well on  $\text{CH}_4$  emissions simultaneously. Water production was not included in the model because it was so highly correlated with gas production ( $R > 0.7$ ) that the effects could not be separated. The following model was used:

$$\log(\text{CH}_4) = \beta_1 \log(\text{gas}) + \beta_2 \log(\text{oil}) + \beta_3 \text{age} \quad (1)$$



**Figure 5.** CH<sub>4</sub> emissions (Mscf/day) versus reported monthly gas production (Mscf/day). Blue lines represent the linear regression lines.

**Table 1.** Pearson Correlation Coefficients (*R*) of Emissions and Production

	CH <sub>4</sub> emissions (Mscf/day)	gas production (Mscf/day)	hydrocarbon liquids production (bbl/day)	water production (bbl/day)
CH <sub>4</sub> emissions (Mscf/day)	1.00			
gas production (Mscf/day)	0.29	1.00		
hydrocarbon liquids production (bbl/day)	−0.01	0.44	1.00	
water production (bbl/day)	0.22	0.77	0.40	1.00
mean age (years)	−0.20	−0.59	−0.34	−0.57

where CH<sub>4</sub> represents measured emissions in g/s, gas is total reported production in Mscf/day, oil is total reported hydrocarbon liquids production in bbl/day, and age is the mean age of the wells in years. Age was not significantly correlated with CH<sub>4</sub> emissions, while gas production was significantly positively correlated, and oil production was significantly negatively correlated (SI Table S2). The negative correlation with oil production is consistent across the basins (SI Figure S1). This negative correlation with oil production is likely due to the lower fraction of CH<sub>4</sub> in wet gas compared to dry gas. Furthermore, emissions from condensate tanks, which are more prevalent in wet gas areas, typically contain a lower fraction of CH<sub>4</sub> and higher fraction of heavier hydrocarbons such as VOCs when compared with produced gas.<sup>6</sup> The inclusion of hydrocarbon liquids and age in the model did not explain much more of the variation in emissions resulting in an adjusted *R*<sup>2</sup> of only 0.096, in contrast to an *R*<sup>2</sup> of 0.083 when only gas production was included (Figure 5).

Other important sources of variation not accounted for in this analysis include emissions controls and equipment present on the pads. Further uncertainty is introduced by the production data: daily or hourly production levels may not be consistent with monthly production.

Although the OTM 33A CH<sub>4</sub> emissions data include episodic features (e.g., flash emissions), it is instructive to compare emission rates as a percent of production with the measurements by Allen et al.<sup>16</sup> and ERG.<sup>15</sup> The differences between the CH<sub>4</sub> emissions estimates of the three studies are amplified when emissions are considered as a percentage of total production rather than in mass emission rate (SI Figure S2). For the sites measured using OTM 33A, approximately 0.72

(0.44, 1.17)%, 1.36 (0.97, 1.95) %, and 0.58 (0.39, 0.86) % of production was emitted on average (with 95% CI) in the Barnett, DJ, and Pinedale basins, respectively, compared with 0.11 (0.09, 0.16)% of production measured by ERG<sup>15</sup> in the Barnett shale and 0.01 (0.01, 0.01) % and 0.09 (0.04, 0.20)% measured by Allen et al.<sup>16</sup> in the Appalachian and Rocky Mountain basins, respectively (SI Figure S2). As evidenced in the statistical analysis, differences in production rate explain only a fraction of the variation in emissions. The percentages from this study only represent emissions from routine well pad operations and thus cannot be directly compared to other estimates of total CH<sub>4</sub> emitted as a percent of production such as those by Brandt et al.<sup>5</sup> that include emissions from many other processes.

Mean gas production at the OTM 33A sites was significantly lower than mean gas production at the sites measured in the direct measurement studies (SI Figure S4). Gas production at the OTM 33A sites ranged from 3.7 (Mscf/day) to 9021 (Mscf/day) with 37% of the sites producing <100 Mscf/day. In contrast, Allen et al.<sup>16</sup> reported a gas production range of 20 to 47 690 (Mscf/day) with only 10% of the sites producing <100 Mscf/day and with approximately 20% of the measured sites producing >10,000 Mscf/day. The gas production values of the ERG<sup>15</sup> sites ranged from 0.06 to 9085 Mscf/day in the Barnett with 10% of the sites producing <100 Mscf/day (SI Figure S4). The OTM 33A results indicate that sites with very low gas and oil production can emit a much greater fraction of the gas produced than sites with higher production levels. Maintenance issues (e.g., fugitive leaks, open or leaking thief hatches, failed pressure relief devices, malfunctioning separator dump valves) could be more prevalent at smaller older production sites than at higher producing sites that are potentially better maintained and may have fundamentally different engineering designs (e.g., use of buffer tanks to suppress flash emissions). Furthermore, many of the fugitive processes can emit at levels that are not linearly associated with production rates as is evidenced by the lack of correlation between emissions and production and the finding by Allen et al.<sup>16</sup> that equipment leaks are underestimated by the 2011 EPA national inventory.

In summary, the OTM 33A mobile inspection method can be used to complement direct measurement techniques and expand our knowledge of the upper range of the distribution of CH<sub>4</sub> emissions. OTM 33A was successfully applied to quantify CH<sub>4</sub> emissions at 210 oil and gas well pads with an accuracy of ±60% determined by controlled release tests. Well pad emissions were log-normally distributed and differed significantly by basin with geometric means ranging from 0.14 g/s in



the Denver-Julesburg to 0.59 g/s in the Pinedale basin. Repeat measurements at 9 sites indicated consistent low emission rates at seven sites and highly variable emissions at two sites, one a documented malfunction. The production rates accounted for approximately 10% of the variation in sampled emission rates in a multivariate linear regression on age, hydrocarbon liquid and gas production. Normalizing emissions by gas production amplified the differences between the remote and onsite measurements. Compared to the direct measurements in the Barnett, the mean of the remote measurements was approximately twice as large in terms of mass emissions rate, but approximately seven times as large when considered as a percentage of production, indicating that sites with lower production levels can emit a much greater percentage of production. Infrared camera videos indicate that emission rates may be strongly affected by stochastic variables. In particular, equipment malfunctions or operator error may cause emission rates to increase substantially compared to routine operating conditions. Accurately estimating site emissions on a regional scale likely will require determining the average magnitude and frequency of these stochastic events.

## ■ ASSOCIATED CONTENT

### ● Supporting Information

Supplemental figures, tables, and IR videos are supplied. This material is available free of charge via the Internet at <http://pubs.acs.org>.

## ■ AUTHOR INFORMATION

### Corresponding Author

\*Phone: 1-919-541-7969; fax: 1-919-541-0359; e-mail: [thoma.eben@epa.gov](mailto:thoma.eben@epa.gov).

### Notes

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