

Methane Emissions from Natural Gas Compressor Stations in the Transmission and Storage Sector: Measurements and Comparisons with the EPA Greenhouse Gas Reporting Program Protocol

R. Subramanian,[†] Laurie L. Williams,[‡] Timothy L. Vaughn,[§] Daniel Zimmerle,[§] Joseph R. Roscioli,[⊥] Scott C. Herndon,[⊥] Tara I. Yacovitch,[⊥] Cody Floerchinger,[⊥] Daniel S. Tkacik,[†] Austin L. Mitchell,[†] Melissa R. Sullivan,[†] Timothy R. Dallmann,[†] and Allen L. Robinson^{*,†}

[†]Center for Atmospheric Particle Studies (CAPS) and the Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, United States

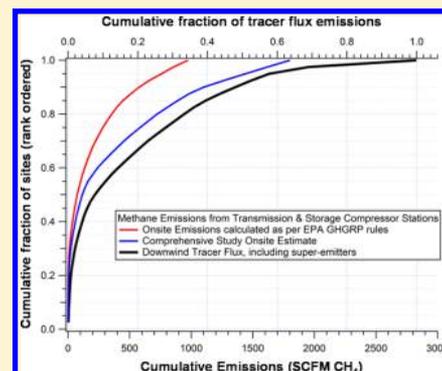
[‡]Department of Physics and Engineering, Fort Lewis College, Durango, Colorado 81301, United States

[§]Energy Institute, Colorado State University, Fort Collins, Colorado 80524, United States

[⊥]Aerodyne Research Inc., Billerica, Massachusetts 01821, United States

S Supporting Information

ABSTRACT: Equipment- and site-level methane emissions from 45 compressor stations in the transmission and storage (T&S) sector of the US natural gas system were measured, including 25 sites required to report under the EPA greenhouse gas reporting program (GHGRP). Direct measurements of fugitive and vented sources were combined with AP-42-based exhaust emission factors (for operating reciprocating engines and turbines) to produce a study onsite estimate. Site-level methane emissions were also concurrently measured with downwind-tracer-flux techniques. At most sites, these two independent estimates agreed within experimental uncertainty. Site-level methane emissions varied from 2–880 SCFM. Compressor vents, leaky isolation valves, reciprocating engine exhaust, and equipment leaks were major sources, and substantial emissions were observed at both operating and standby compressor stations. The site-level methane emission rates were highly skewed; the highest emitting 10% of sites (including two superemitters) contributed 50% of the aggregate methane emissions, while the lowest emitting 50% of sites contributed less than 10% of the aggregate emissions. Excluding the two superemitters, study-average methane emissions from compressor housings and noncompressor sources are comparable to or lower than the corresponding effective emission factors used in the EPA greenhouse gas inventory. If the two superemitters are included in the analysis, then the average emission factors based on this study could exceed the EPA greenhouse gas inventory emission factors, which highlights the potentially important contribution of superemitters to national emissions. However, quantification of their influence requires knowledge of the magnitude and frequency of superemitters across the entire T&S sector. Only 38% of the methane emissions measured by the comprehensive onsite measurements were reportable under the new EPA GHGRP because of a combination of inaccurate emission factors for leakers and exhaust methane, and various exclusions. The bias is even larger if one accounts for the superemitters, which were not captured by the onsite measurements. The magnitude of the bias varied from site to site by site type and operating state. Therefore, while the GHGRP is a valuable new source of emissions information, care must be taken when incorporating these data into emission inventories. The value of the GHGRP can be increased by requiring more direct measurements of emissions (as opposed to using counts and emission factors), eliminating exclusions such as rod-packing vents on pressurized reciprocating compressors in standby mode under Subpart-W, and using more appropriate emission factors for exhaust methane from reciprocating engines under Subpart-C.



INTRODUCTION

The growing production and utilization of natural gas in the United States (US) has raised questions about methane emissions from the natural gas system.^{1,2} Methane, a greenhouse gas (GHG), is the primary component of natural gas. The US Environmental Protection Agency (EPA) estimates that the natural gas system contributed about 23% of annual US methane emissions in 2012 by relying heavily on data collected

in the 1990s.³ Recent studies have suggested that the EPA and other emissions inventories may underestimate US national methane emissions from all sources by 25–75%.^{4–6}

Received: December 11, 2014

Accepted: January 21, 2015

Published: February 10, 2015

The 2012 EPA Greenhouse Gas Inventory (GHGI) attributes one-third of methane emissions from the US natural gas system to the transmission and storage (T&S) sector.³ This sector comprises about 2000 compressor stations distributed along approximately 300,000 miles of pressurized pipelines, underground storage facilities, and associated equipment.³ The EPA GHGI for the T&S sector is largely based on emission factors from a 1996 GRI/EPA study.⁷ During the past two decades, there have been substantial changes in the natural gas system, which warrants a re-examination of methane sources and emission rates, e.g. Allen et al.⁸ Two more recent studies reported emissions data from 13 T&S compressor stations,^{9,10} two fewer than the GRI/EPA study. The lack of methane emissions data was identified as a critical issue for assessing the environmental impacts of natural gas use.¹¹

Starting in 2011, T&S facilities that emit more than 25,000 t of carbon dioxide equivalent (MT-CO₂e) report their emissions under the EPA Greenhouse Gas Reporting Program (GHGRP; 40 CFR §98, Subpart C and Subpart W) using a combination of onsite emission measurements and emission factors. In 2012, 495 transmission and underground storage facilities reported methane emissions under this program, about one-quarter of the total facilities in the T&S sector.³ The EPA GHGI does not yet use the GHGRP emissions or activity data, but the EPA GHGRP represents a potentially important new data source for the next generation of natural gas system emissions inventories. However, the use of leak counts and emission factors for certain equipment types, the exclusion of emissions from sources in certain operating modes, and the use of a single emission factor for all unburned methane emissions in engine exhaust regardless of prime mover type (turbine or reciprocating engine) raise questions over the GHGRP's completeness as a basis for emissions inventories.

This paper presents methane emissions measurements made at 45 compressor stations in the T&S sector, more than the three previous studies combined.^{7,9,10} At each site, two independent yet complementary measurement methodologies were employed—comprehensive onsite emissions measurements and downwind dual-tracer flux. Results from the two independent methodologies are compared to investigate uncertainties in site-level emission estimates, EPA GHGI, and EPA GHGRP. Zimmerle et al.¹² use these results, industry-reported activity and emissions data from the GHGRP, and other data to estimate the 2012 national methane emissions for the T&S sector.

METHODOLOGY

Measurements were performed to characterize the methane emissions from 45 compressor stations in the T&S sector during the summer and fall of 2013. The measurements were made in collaboration with six major natural gas pipeline operators. Two independent yet complementary measurement methodologies were employed. Onsite emissions measurements provide a “bottom-up” estimate of site-level methane emissions by direct measurement of individual emission sources, which are then combined to provide a site-level estimate. Tracer flux techniques provide a “top-down” estimate of site-level emissions derived from measured methane and tracer gas concentrations in downwind plumes. The two approaches were deployed on the same day(s) to facilitate direct comparison of the two independent estimates of the site-level methane emissions, to evaluate site-level closure between

the top-down and bottom-up approaches, and to increase confidence in the results.

Study Sites. Methane emissions were measured at 36 compressor stations in the transmission system and nine compressor stations associated with underground storage facilities operated by six partner companies. The stations were located in 16 states across the South, Mid-Atlantic, Northeast, Midwest, and the Mountain West. The stations were equipped with two types of compressors: reciprocating compressors driven by natural gas-fired reciprocating engines, and centrifugal compressors driven by gas-fired turbines (though a few are driven by electric motors). Nineteen stations were equipped with only reciprocating compressors; the average number of compressors at these sites was 4.8 with an average capacity of 2335 hp per compressor. Twenty-one stations were equipped with only centrifugal compressors; the average number of compressors at these sites was 2.4 with an average capacity of 11,741 hp per compressor. Five sites were “mixed” sites with both compressor types. At least one compressor was operating at 20 sites. Twenty-five sites reported to the EPA GHGRP in 2012. Additional site details are summarized in Tables S1 and S2 in the Supporting Information.

Site selection was performed by the study team from lists of sites provided by the six partner companies. Site selection was not random but instead was based on a number of factors: geographic location, technology, partner company greenhouse survey team schedules, and site suitability for tracer flux measurements. We sought to achieve broad geographic coverage, while we also acknowledged the travel schedule of the field teams. Sites were selected to span a range of technologies (centrifugal versus reciprocating compressors; wet versus dry seal centrifugal compressors), operating states (operating, standby depressurized, and standby pressurized), and GHG reporting status (reporter if required to report under GHGRP, otherwise nonreporter). The majority of the measurements were performed in partnership with partner company GHG onsite survey teams, which required schedule coordination. Finally, local road access and meteorological conditions were evaluated to determine the suitability of each site for tracer flux measurements.

The overall goal of the site selection process was to develop a site list that was broadly representative of the partner company fleets, which comprise ~56% of the interstate transmission facilities reported to the US Federal Energy Regulatory Commission (FERC) under Form 2.¹² Comparisons indicate that this objective was largely achieved. For example, the study sites had 187 compressors, of which 69% were reciprocating compressors; this is slightly lower than the census of reciprocating compressors at all partner-company stations (75% of 3052 compressors). Analysis of the GHGRP or equivalent data for 2012 indicates that the methane emissions from the study sites are representative of the broader company fleets; details are provided in the Supporting Information. Briefly, GHGRP-equivalent annual methane emissions data were available for 2012 from 343 Partner sites. This includes 29 study sites. A two-sample Kolmogorov–Smirnov (K–S) goodness-of-fit hypothesis test indicates that these two sets of 2012 methane emissions data were drawn from the same underlying distribution at 95% confidence (Figure S1, Supporting Information.)

Onsite Measurements. The onsite emissions measurements involved comprehensive leak detection followed by

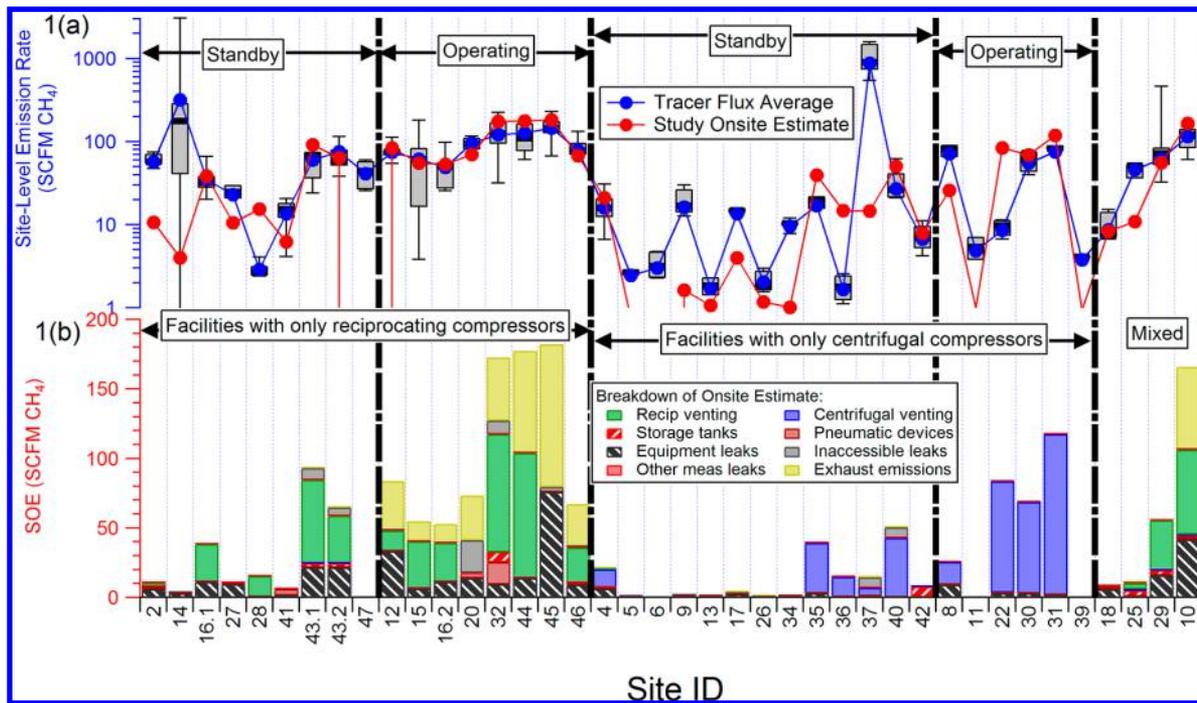


Figure 1. (a) Tracer flux-based site-level methane emission rates for the 40 sites for which both tracer flux and onsite emissions data are available in the same operating state. The boxes represent the 25th and 75th percentiles of the tracer flux data at a site; whiskers represent the 95th and 5th percentiles. The blue line represents the average tracer flux emissions rate, while the red line is the SOE. (b) The SOE broken down by source categories (Table S3, Supporting Information). Sites 18, 25, and 29 were on standby and had both reciprocating and centrifugal compressors (“mixed” sites). Mixed site 10 had one turbine and two reciprocating compressors on standby and four operating reciprocating compressors.

direct measurement of all safely accessible sources of nonengine exhaust methane emissions. Equipment leaks from valves, flanges, connectors, open ended lines, etc. were identified using infrared imaging. The methane emissions from all identified leaks and vented sources (reciprocating compressor rod-packing, blowdown vents, centrifugal wet seals, etc.) were then directly quantified using Bacharach Hi-Flow samplers, anemometers, turbine flow meters, or calibrated bags; in one case, a rotameter was used. Additional details of the onsite emissions measurement protocol are described in the Supporting Information.

A total of 1398 individual onsite methane emissions measurements were collected in this study (Table S5, Supporting Information). For each site, all measured onsite data were combined to create a bottom-up estimate of the site-level methane emissions, called the study onsite estimate (SOE), which is as comprehensive as possible to maximize comparability with the tracer flux measurements of site-level methane emissions. The SOE uses emission estimates in only two situations: (1) inaccessible sources (identified leaks or vents that could not be safely accessed) were estimated using “study factors”; (2) unburned methane in engine/turbine exhaust, not measured in this study, was estimated using AP-42 emission factors (see Supporting Information for details). Study factors are the average of all study measurements for the same component or leak type (Table S10, Supporting Information).

At 34 stations, the onsite methane emissions measurements were made by either the partner company or a partner-supported third-party contractor as part of the company’s ongoing, EPA-mandated GHGRP reporting activities. At 11 stations, the onsite emissions measurements were made by a project-supported contractor (URS Corporation). No significant differences were seen between the partner- and study-

funded onsite contractor data sets when compared against the corresponding downwind tracer flux measurements. At 13 of these stations (Table S2, Supporting Information), the partner contractor only performed a less comprehensive GHGRP-compliant onsite survey, not the more complete study onsite protocol; however, this does not appear to have made a significant difference except at one superemitter site discussed below. Differences between the two protocols are discussed in the Supporting Information.

Downwind Tracer Flux. Downwind tracer flux (or just “tracer flux”) is a technique developed to quantify the aggregate, site-level emissions of methane (or other species) from large, often complex sites with multiple leak points.^{13–15} The dual tracer flux method used here is described in detail by Roscioli et al.¹⁶ Briefly, two tracer gases (nitrous oxide and acetylene) were released at known emission rates from the target site. Each tracer was released from a single point onsite. The release points for the two tracers were separated from each other by up to 750 feet orthogonal to the wind direction so as to bracket the expected sources. The concentrations of the two tracers, methane, ethane, and other species were measured 0.5–3 km downwind of the site using high time-resolution instrumentation (1-Hz or faster, Aerodyne QC-TILDAS and/or Picarro cavity ring-down spectroscopy) deployed on a mobile sampling laboratory. Plume profiles were obtained for each facility by driving the mobile laboratory at a constant speed on a road roughly perpendicular to the wind direction. This process was repeated to obtain multiple plume transects downwind of each site; an average of eight plumes per site met study quality criteria.

The facility-level methane emission rate was calculated for each downwind plume based on the known tracer release rate and the background-corrected methane-to-tracer concentration

ratios.¹⁶ Modeling of pollutant dispersion is not required—this complexity is empirically captured by the tracers. Roscioli et al.¹⁶ describes the different types of plumes and data analysis procedures. Method assumptions and uncertainties are discussed in the Supporting Information. Each plume represents the site-wide emissions in the 1–3 min duration of each transect. While the study plan called for dual-tracer measurements at every site, instrument detection limits and malfunctions on certain days resulted in both dual-tracer and traditional single-tracer plume measurements. Multiple downwind plumes were obtained at each station providing multiple measures of the site-level emissions. The average methane emission rate for each site was calculated by averaging results from each plume weighted by plume-type-specific uncertainty ($1/\sigma^2$; see Supporting Information). The uncertainty of the site average emission rate is calculated as the unbiased sample variance of the mean (using Bessel's correction).

RESULTS

The site-level emissions data are summarized in Figure 1, panel a. The tracer flux measurements indicate that the methane emissions varied by almost three orders of magnitude across the 45 study sites. The methane emission rate from the lowest emitting site was 1.7 ± 0.2 SCFM versus 880 ± 120 SCFM for the highest emitting site (SCFM = standard cubic feet per minute at 1 atm and 15.6 °C; 1 SCFM = 19.2 g CH₄/min). The box-and-whisker plots show the distribution of emissions measured for each plume transect. At most sites, the plume-to-plume differences were small (coefficient of variation less than 20% at 35 sites, Table S9, Supporting Information), which indicates that emissions were relatively invariant in time. Therefore, the variation in emissions at a given site was much lower than variation across sites.

The ethane/methane ratio has been shown to effectively distinguish thermogenic (natural gas) methane from biogenic methane.¹⁷ Figure 2 shows excellent agreement between the ethane/methane ratio measured in the downwind plumes and gas composition data provided by the partner companies (slope 0.94 ± 0.02 , $R^2 = 0.97$, excluding one outlier). This demonstrates that the background-corrected methane measured in each plume is associated with the target facility and not some other source(s). The one outlier in this comparison (site 24) had a significant off-site source of natural gas methane that interfered with the downwind tracer flux measurements; that site is excluded from the subsequent analysis.

Although the tracer flux technique characterizes site-level emissions, it does not identify the specific sources that drive the emissions. The onsite data can address this shortcoming. However, a potential concern with onsite surveys is systematic bias due to inaccessible sources and leaks not detected, especially at large and complex compressor stations with hundreds of valves, vents, and other potential emission points. Therefore, before the onsite data are examined, we compare the SOE calculated from the onsite measurements to the tracer flux data.

Comparison of Tracer Flux and SOE. Figure 3 compares the SOE and tracer flux estimates of the site-level methane emissions at the 40 sites where valid tracer flux and onsite data were collected at the same site in the same operating state on the same day(s). The sites are grouped by compressor type and operational state; if at least one compressor was running, the site is classified as “operating.” The inset of Figure 3 shows the comparison for all 40 sites with simultaneous tracer flux and

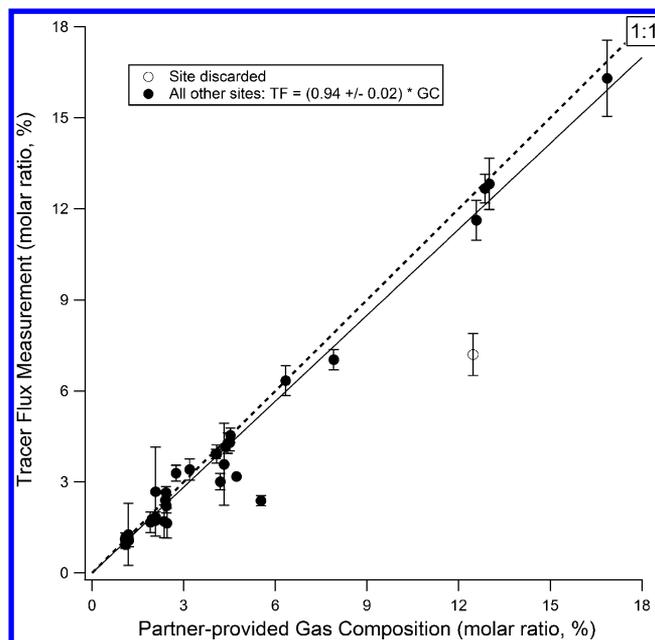


Figure 2. Comparison of ethane/methane molar ratios from partner-company-provided gas composition data with the downwind tracer flux measurements. Disregarding the one site with a known off-site interfering methane source, the two sets of data are strongly correlated ($R^2 = 0.97$).

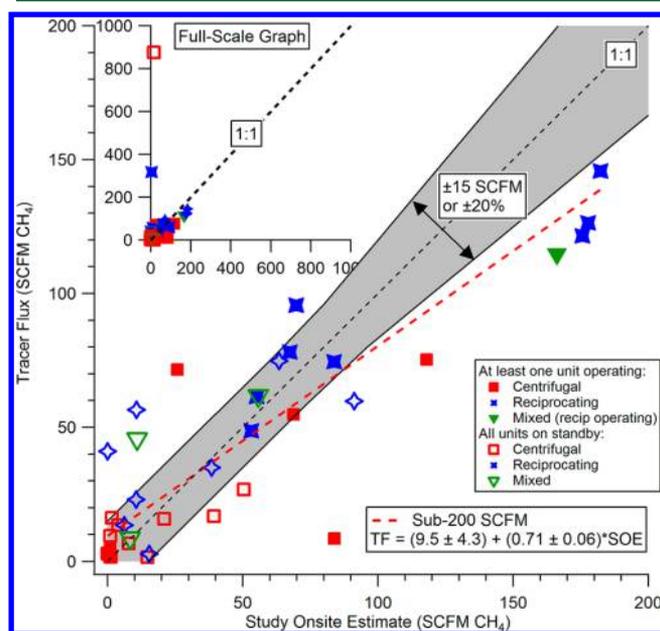


Figure 3. Comparison of the site-level tracer flux average and SOE methane emissions. The main panel shows 38 sites with emissions less than 200 SCFM, while the inset shows the full data set of 40 sites. Open symbols indicate sites where all compressors were in standby mode; solid symbols indicate sites where at least one compressor was running. Gray-shaded area indicates ± 15 SCFM or $\pm 20\%$ uncertainty bars (whichever is larger). Red dashed line is a linear fit (Deming regression) to sub-200 SCFM data.

onsite data, while the main panel shows data for the 38 sites that emitted less than 200 SCFM.

At most sub-200 SCFM sites, there is reasonable agreement between the two independent measures of site-level emissions. For example, Figure 3 shows that the onsite and tracer flux data

for the 38 sub-200 SCFM sites are scattered about the one-to-one line. At 24 of these sites, the two independent measurements agree to within ± 15 SCFM or $\pm 20\%$ (whichever is greater), and data for five other sites are within $\pm 40\%$. The average 95% confidence interval of the tracer flux data is $\pm 29\%$ (Table S9, Supporting Information). Therefore, the data from well over half the sites agree within this bound before even accounting for uncertainty in the SOE. Unfortunately, the dominant uncertainty of the SOE is likely nonparametric uncertainty of uncharacterized emissions (undetected or identified but inaccessible) rather than parametric uncertainty associated with individual measurements or instruments. This nonparametric uncertainty applies to every site (operating and standby) but is impossible to quantify. We expect that the uncertainty of the SOE at most sites to be comparable to that of the tracer flux data. Hence, the site-level emissions at essentially all the sub-200 SCFM sites likely agree within measurement uncertainty.

There appears to be a systematic trend between the two data sets for the sub-200 SCFM sites—a Deming regression yields an intercept of 9.5 SCFM and a slope of 0.71. Therefore, for this subset of sites, the tracer flux method systematically measured somewhat higher methane emission rates than the SOE at lower-emitting sites, while SOE was higher than tracer flux at higher emitting sites. No single common factor explains the discrepancies between the two data sets. Potential explanations include identified but inaccessible sources, estimated engine exhaust emissions, and potential biases in both tracer flux and onsite measurement techniques. For example, the four highest SOE sites had operating reciprocating engines (#10, #32, #44, and #45). At these sites, engine exhaust emissions estimated with AP-42 emission factors contributed 27–57% of SOE. The AP-42 emission factors may overestimate engine methane emissions at these sites; it is also possible that the downwind measurement did not fully capture lofted exhaust emissions.¹⁶ Emissions from identified but unmeasured leaks (when the emission point was not safely accessible) may be greater than the study factors used to estimate them. At some sites, the onsite team followed a GHGRP-compliant protocol (Table S2, Supporting Information) and did not measure all onsite leaks; the tracer flux data was higher than SOE at many of these sites. These and other potential factors are described in more detail in the Supporting Information.

The inset of Figure 3 indicates that the tracer flux estimate was almost two orders of magnitude larger than the SOE at two sites. At both of these sites, there were known issues with the onsite survey. Site 37 had one standby pressurized centrifugal compressor and two standby depressurized centrifugal compressors. The SOE at this site was 15 SCFM versus 880 ± 120 SCFM for the tracer flux. One of the station isolation valves at this site was frosted over and hissing, indicating a very significant leak. The onsite survey team followed a GHGRP-compliant survey and used an acoustic measurement device (VPAC) to characterize the leak at this valve, reporting a leak of only 3 SCFM. Although acoustic devices are approved by EPA to measure leaks across valves, they have been shown to substantially underestimate leak rates.¹⁰ This appears to have occurred at site 37. The other site with a very large discrepancy was site 14, a large storage facility. The SOE at this site was 4 SCFM versus 320 ± 160 SCFM for the tracer flux. This site was undergoing maintenance, during which there was significant venting of isolation valves on a pipeline from the storage field to the compressor station. However, this vent could not be

safely measured by the onsite team. Therefore, its emissions were estimated using the open ended line study factor, which likely substantially underestimated the actual venting. The partner company reported that the venting occurred for 15–20 h. The data for these two sites (37 and 14) highlight the challenges of safely performing onsite measurements of large emission sources.

DISCUSSION

Superemitters and Skewed Distributions. Figure 4, shows that the cumulative distributions of the site-level

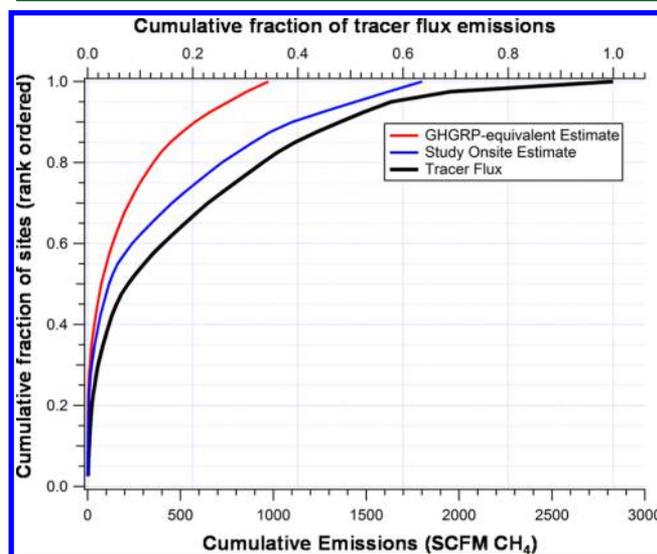


Figure 4. Cumulative distributions of site-level methane emissions from the 40 sites with both tracer flux and onsite emissions measurements obtained in the same operating state. Two different onsite estimates are shown: the SOE, which includes all measured emissions, and the GHGRP-equivalent estimates, which are based on EPA GHGRP protocols (see text for details).

methane emissions measured by this study are highly skewed with a small number of sites contributing disproportionately to the total overall emissions. For example, the highest emitting 10% of the sites contribute over 50% of the aggregate tracer flux emissions across all sites; conversely, the lowest emitting 50% of the sites contributed less than 10% of the aggregate emissions. A large fraction of this is driven by the two superemitters (sites 14 and 37) that were captured by tracer flux but not by onsite measurements. However, the SOE distribution is only modestly less skewed than the tracer flux data, with just 15% of the tested sites responsible for 50% of the cumulative SOE. Not capturing the two superemitters results in the SOE underestimating the total emissions from all sites by 35% relative to the tracer flux technique. The component-level emissions data are similarly skewed.¹²

The skewness of T&S methane emissions, which is quantified here for the first time, appears similar to other natural gas sectors. For example, 10% of the gas wells contributed nearly 70% of the emissions from 250 Texas gas wells measured as part of the Fort Worth study.^{1,9} Highly skewed emissions dominated by a small number of sites have important implications for the construction of emission inventories.⁶

Major Onsite Sources of Fugitive and Vented Emissions. Although there are differences between the tracer

flux and SOE for the 38 nonsuperemitter sites, the difference in the cumulative emissions measured by the two techniques across this subset of sites is small (9%). This means that the comprehensive onsite measurements performed by this study did not systematically miss important emission sources for this subset of sites, and that the onsite measurements can be used to identify major methane sources at these sites.

Figure 1, panel b shows the SOE for each site broken down into major source categories (data are in Tables S9 and S10, Supporting Information). The relative importance of each source category varied from site to site, and no one category was associated with every high emitting site. Compressor venting was the most important source category and contributed almost 50% of the aggregate SOE across all sites. All but two of the sites with above-average emissions had substantial compressor vent emissions. For reciprocating-only stations on standby, comprehensive onsite measurements showed significant compressor rod-packing vent emissions when at least one compressor was in the standby pressurized mode; no compressor vent emissions were observed at sites where every reciprocating compressor was depressurized. Only 14 facilities had natural-gas-driven pneumatic devices; these were not a major source of methane. There was no correlation of methane emissions with site size (as measured by either total horsepower or number of compressors). Additional discussion of the onsite data is in the Supporting Information.

The onsite measurements indicate clear trends in the study-average emissions if one controls for compressor technology (reciprocating-only versus centrifugal-only stations) or operating state (standby versus operating). Figure 1, panel b shows that sites with at least one compressor operating generally emitted more methane than sites completely on standby. Figure 5 shows that these differences are due to emissions from compressor vents and components. However, the onsite data did not capture the superemitter emissions; both superemitter sites were on standby. There are not enough data from this study to conclude whether superemitters are more commonly found in standby or operating modes.

For sites on standby, Figure 5, panel a shows that there are greater per-compressor emissions from reciprocating compressors than from centrifugal compressors, with rod-packing vent emissions from standby pressurized reciprocating compressors the main contributor to this difference. For operating sites, the emissions from centrifugal-only and reciprocating-only sites were comparable on a per-compressor basis. However, centrifugal compressors had five times the horsepower rating of reciprocating compressors in this study; therefore, centrifugal compressors have much lower methane emissions than reciprocating compressors when normalized to horsepower rating or throughput capacity. Zimmerle et al.¹² show that over the past 20 years, centrifugal compressors have become much more common in the T&S sector; therefore, these differences in methane emissions based on compressor technology have important implications for the national inventory.

Operation of reciprocating engines emitted considerably more engine exhaust methane than centrifugal compressors (Figure 1b). The study-average AP-42-based exhaust emissions for reciprocating engines were 13 SCFM/compressor versus only 0.5 SCFM/compressor for turbines.

Emissions are often expressed as a loss rate relative to the methane throughput.^{2,4,6} However, 25 of the study sites were on standby, so they had zero throughput. Figure 2 indicates that substantial (>20 SCFM) methane emissions were

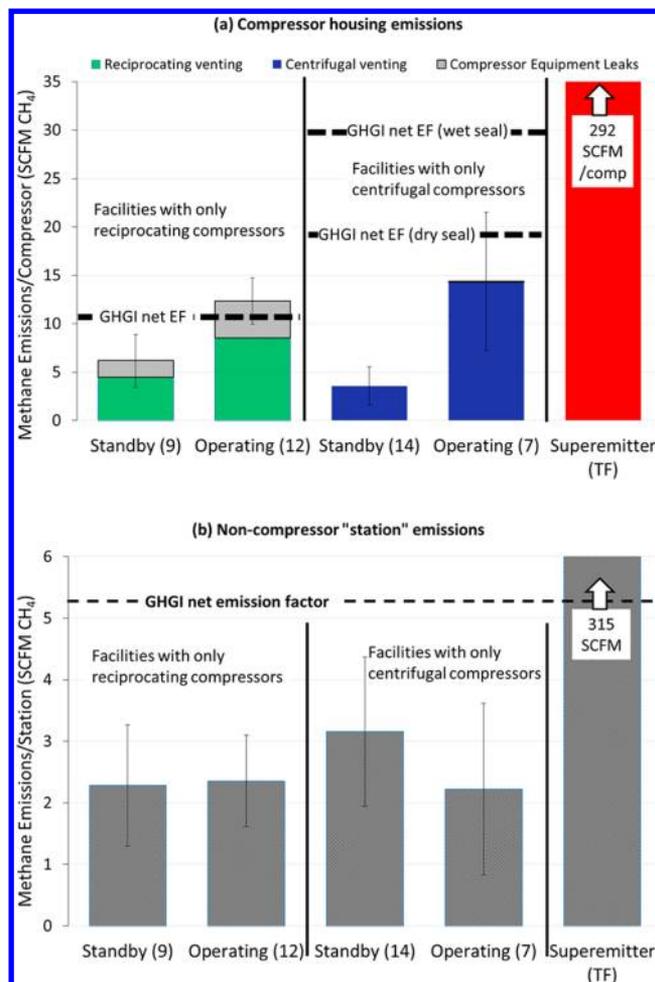


Figure 5. Study-average onsite methane emissions compared to GHGI net emission factors (EFs) for transmission compressor stations: (a) compressor housing, and (b) noncompressor sources. The left four bars in each panel are averages of onsite measurements (numbers in parentheses on the x-axis labels indicate the number of sites in each average), which did not capture the superemitter emissions. The right-most bars show tracer flux data for the two superemitter sites; these two bars extended well beyond the y-axis scales to the value listed on each panel. GHGI net emission factors for compressor stations at storage sites (not shown) are comparable or even higher than those used for transmission.¹² Both dry and wet seal centrifugal compressors were sampled in this study but are combined into one category for simplicity. GHGI net emission factors are a composite of all operating conditions, as discussed in the text. Error bars are one standard of error.

measured at about half of these sites; in fact, the two superemitter sites (site 37 and 14) were on standby. Figure 1, panel b indicates that emissions from standby sites are associated with leaky isolation valves, rod-packing vents on standby pressurized reciprocating compressors, and miscellaneous leaks from other pressurized equipment. T&S stations are often on standby in the summer when the demand for natural gas is lower. Station utilization has also been influenced by the development of new shale gas plays, such as the Marcellus, which have reduced the need to transport gas from the southeast/Texas to the northeast US.

Implications for the EPA GHG Inventory. The EPA GHG inventory for methane categorizes emissions from T&S compressor stations into compressor housing emissions,

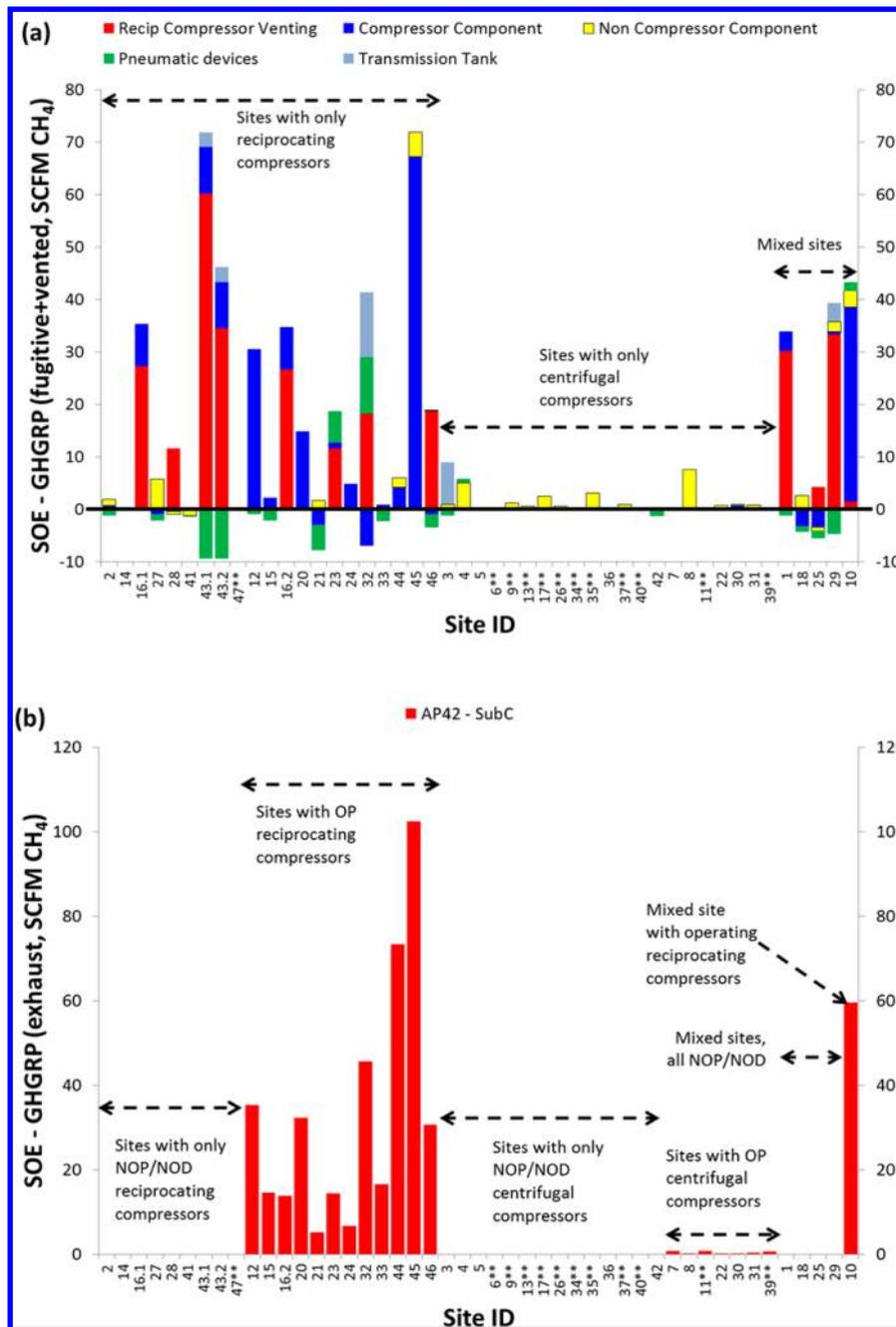


Figure 6. Differences by source category (Table S3, Supporting Information) between the SOE and the GHGRP-equivalent estimate for (a) fugitive and vented emissions and (b) turbine/engine exhaust emissions. Centrifugal compressor vents are minor (less than 0.02 SCFM per site) and therefore excluded for visual clarity in panel a. The SOE uses AP-42-based emission factors to estimate the exhaust emissions of methane, while the GHGRP exhaust emissions are based on Subpart-C. Site IDs with two asterisks indicate that the GHGRP-compliant onsite protocol was used at that site.

noncompressor “station” emissions, exhaust emissions, blow-downs, and emissions from pneumatic devices. The onsite data from this study can be used to evaluate two of these categories: compressor housing and station emissions. The EPA GHGI compressor housing emission factors are a composite of emissions from rod-packing vents (or wet seal or dry seal vents for centrifugal compressors), blowdown and isolation valves, and other compressor equipment (excluding exhaust emissions), weighted by time in operating and standby modes, by mostly using component counts and emissions measurements from the 1990s. Unfortunately, the weighting factors

used by the EPA GHGI are not well documented. This complicates making direct comparisons with the study data.

To make comparisons with our data, effective or net GHGI emission factors were derived from the 2012 GHGI by normalizing the reported net annual emissions (potential less EPA-estimated reductions) in each category by the corresponding GHGI activity. For example, in the transmission sector, GHGI estimates that 7235 reciprocating compressors emit 772.6 Gg/year of methane (activity count in Table A-129, net emissions in Table A-141 of ref 3). Therefore, the net GHGI emission factor is 107 Mg/year for each reciprocating compressor, or 10.6 SCFM (assuming 8760 h/year). The

same calculation was performed for centrifugal compressors. The EPA GHGI does not break out reductions into all categories, so reductions were proportionally distributed across the unspecified categories.¹²

To make more meaningful comparisons with the GHGI, we calculated average emissions from the study onsite data for four types of sites: reciprocating-only sites with all compressors on standby; reciprocating-only sites with one or more compressors operating; centrifugal-only sites with all compressors on standby; and centrifugal-only sites with one or more compressors operating. We also separated the study data into two categories that correspond to those used by the GHGI: compressor housing (compressor vents, valves, and other components, but not engine or turbine exhaust emissions) and station/noncompressor components (storage tanks and non-compressor housing equipment leaks). Before averaging, the compressor housing emissions from each site were normalized by the number of compressors at that site.

Figure 5, panel a compares the different study averages calculated from the onsite data for compressor components to the EPA GHGI net emission factors for reciprocating and centrifugal compressors from the transmission sector. Since the GHGI net emission factor is an annualized composite of operating and nonoperating compressors, it should be bounded by the study-average emission rates for operating and nonoperating compressors. This occurs for sites with only reciprocating compressors, which indicates that there is reasonable agreement between these two data sets for this type of compressor. However, the GHGI methane emission factors for both dry and wet seal centrifugal compressors are larger than the study-average results based on the onsite emissions measurements. The GHGI net emission factors for wet seal and dry seal centrifugal compressors are both larger than the study average of the onsite measurements at sites with operating compressors by a factor of two and 34%, respectively; the difference is even larger for study sites on standby.

Figure 5, panel b shows that study-average noncompressor methane emissions based on the onsite data are comparable across all four categories of stations, independent of compressor type as might be expected. However, the study-average data are about half the EPA GHGI noncompressor emission factor for transmission stations and one-sixth the GHGI net emission factor for storage facilities. The differences between the GHGI and study onsite data shown in Figure 5 are probably, at least partially due to technology improvements and other factors that have occurred over the past two decades.¹²

Although the comparisons with onsite data shown in Figure 5 suggest that the EPA GHGI may overestimate emissions from certain source categories, these comparisons exclude the contribution of the superemitter emissions, which were only measured by the tracer flux method. The superemitter emissions are shown using separate bars in Figure 5. Figure 5, panel a shows tracer flux data from site 37, since the high emissions at this site were likely from the compressor housing; Figure 5, panel b shows tracer flux data from site 14, where the superemitter venting was yard piping ("station" emissions). Average emissions based on this study would likely exceed the EPA GHGI net emission factors if these superemitter emissions were included in the analysis. For example, that could happen if superemitter emissions similar to those shown in Figure 5 occurred at a few percent of sites nationally. However, we lack the data to determine whether the magnitude and frequency of

the superemitters encountered in this study are representative of the entire T&S sector.

Implications for the EPA GHG Reporting Program. In 2011, the EPA started requiring compressor stations in the T&S sector that emit more than 25,000 MT-CO₂e to report their methane emissions under the EPA GHGRP, which is by far the largest effort to date to collect methane emissions data from the T&S sector. These publically available data will likely be a critical input for the next generation of methane emission inventories. The comprehensive onsite measurements in this study can be used to construct a GHGRP-equivalent estimate for the methane emissions from each study site to evaluate the GHGRP protocols.

The GHGRP-equivalent estimate uses the Subpart-W methodology for onsite fugitives and venting and Subpart-C emission factors for engine/turbine exhaust emissions. Briefly, Subpart-W estimates emissions from leaking components are based on leaker counts and emission factors; it excludes emissions from sources in certain operational modes, specifically rod-packing vent emissions from standby pressurized reciprocating compressors. Subpart-C uses a single emission factor for all engines/turbines irrespective of the prime mover type, unlike AP-42, which has specific emission factors depending on engine type (rich burn or lean burn, two-stroke or four-stroke reciprocating engines, and turbines). Methodological differences between the GHGRP-equivalent estimate of site-level emissions and SOE are summarized in Table S3 of the Supporting Information.

Figure 6 shows the source-category-by-source-category differences between the SOE and the GHGRP-equivalent estimate across all study sites. The aggregate SOE for the 45 sites was 2097 SCFM versus only 1148 SCFM for the aggregate GHGRP-equivalent estimate. Since only 25 sites exceed the annual 25,000 MT-CO₂e GHGRP reporting threshold (which is largely determined by fuel combustion CO₂, not methane), the aggregate GHGRP-equivalent methane emissions drop further to 790 SCFM. Therefore, only 38% of the aggregate SOE would be reported under the current EPA GHGRP rules. In addition, the onsite measurements did not capture the two superemitters, so the bias is even larger relative to the total emissions. Only 27% of the aggregate tracer flux methane emissions would be reported under the current GHGRP due to a combination of the GHGRP rules, the GHGRP reporting threshold, and gaps (superemitters) in the onsite measurements.

The 45% difference between the aggregate GHGRP-equivalent estimate and the aggregate SOE for all study sites was almost evenly divided between fugitive/vented emissions and exhaust emissions. Figure 6, panel b indicates that there are very large differences between the AP-42 and Subpart-C methane emission factors for reciprocating engines (0.8 SCFM for Subpart-C versus 451 SCFM using AP-42 emission factors). Therefore, Subpart-C significantly underestimates the unburned methane in reciprocating engine exhaust emissions. Reciprocating engines, in particular two-cycle engines, can emit significant methane due to cyclic combustion, piston ring blow-by, etc.,¹⁸ while modern turbine combustors operate with 99% or higher combustion efficiency.¹⁹

Figure 6, panel a shows large differences between the GHGRP-equivalent estimate and the SOE for fugitive/vented emissions. A major cause of this difference is that the EPA GHGRP does not (as of 2014) require reporting of rod-packing vent emissions from standby pressurized reciprocating

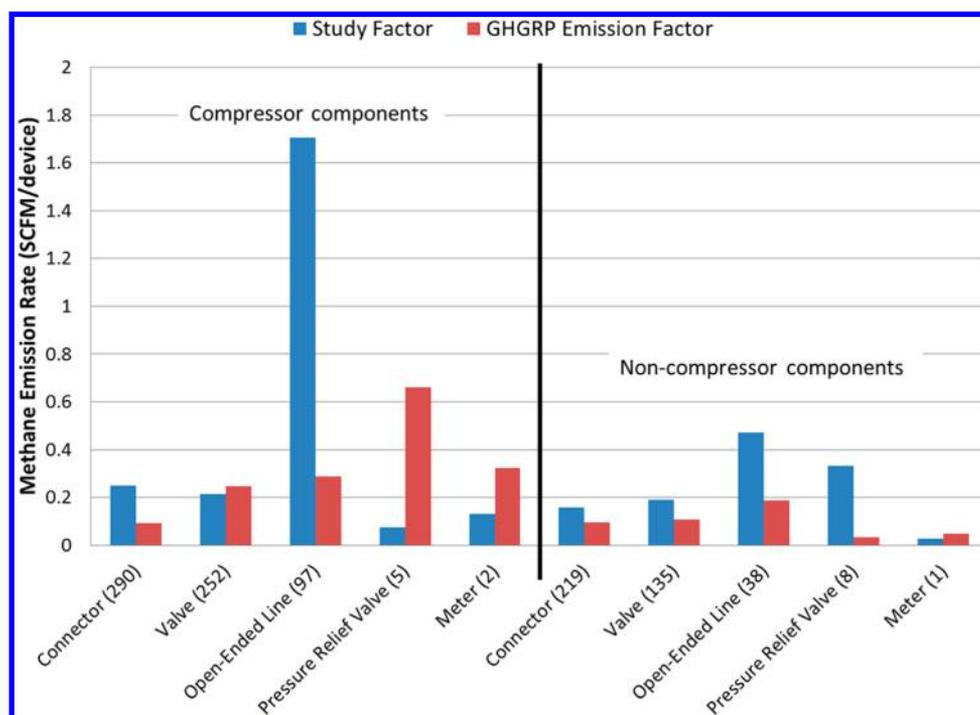


Figure 7. Comparison of the study-average emissions factors (study factors) with the EPA GHGRP emission factors for compressor and noncompressor component sources. Numbers in parentheses on the *x*-axis labels indicate number of study measurements in each category.

compressors. These vents contributed 273 SCFM (13%) to the aggregate SOE and therefore represent an important gap in the GHGRP protocol. Outdated GHGRP emission factors for component leaks account for most of the remaining gap between the measured fugitive/vented emissions and the emissions reportable under GHGRP. For example, Figure 7 indicates that the study-average emission factor for compressor connectors is three times the GHGRP emission factor; the study-average emission factor for open-ended lines is six times larger than the GHGRP emission factor. The study average measurements of leaks from noncompressor connectors and open-ended lines are also larger than the corresponding GHGRP emission factors.

The EPA GHGI does not yet utilize the information in the GHGRP database. This database provides valuable new information on the T&S sector in terms of compressor types, component counts, and other activity factors that are relevant to an emissions inventory. However, before using the GHGRP for inventory calculations, one must account for all the biases and uncertainties in the emissions data shown in Figures 4 and 6; this is done by Zimmerle et al.¹² The value of the GHGRP data for emissions inventory development would be improved by requiring more direct measurements of emissions (as opposed to using counts and emission factors), avoiding the use of acoustic devices, eliminating exclusions such as rod-packing vents on standby pressurized reciprocating compressors, and using more appropriate emission factors for exhaust methane from reciprocating engines.

■ ASSOCIATED CONTENT

⑤ Supporting Information

Description of the 45 study sites; comparison of the annual GHGRP emissions reported for the study sites with the corresponding data for the Partner fleet of compressor stations; methodological details for the onsite emissions measurements,

the EPA GHGRP, and tracer flux; and site-level onsite and tracer data presented in this manuscript. This material is available free of charge via the Internet at <http://pubs.acs.org>.

■ AUTHOR INFORMATION

Corresponding Author

*Phone: 412-268-3657; fax: 412-268-3348; e-mail: alr@andrew.cmu.edu.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank our sponsors for financial support, technical advice, and access to sites for sampling: Dominion, Dow Chemical, Enable Gas Transmission, LLC (formerly CenterPoint Energy Gas Transmission Company, LLC), Environmental Defense Fund (EDF), Interstate Natural Gas Association of America (INGAA), Kinder Morgan, Columbia Pipeline Group (formerly NiSource), TransCanada, and The Williams Companies, Inc. Funding for EDF's methane research series, including this work, was provided by Fiona and Stan Druckenmiller, Heising-Simons Foundation, Bill and Susan Oberndorf, Betsy and Sam Reeves, Robertson Foundation, Alfred P. Sloan Foundation, TomKat Charitable Trust, and the Walton Family Foundation. We also thank the project science advisory panel for the valuable feedback: J. Kuo, D. Picard, R. Talbot, and M. Whelan. The views and opinions expressed are those of the authors and do not necessarily reflect those of the sponsors or the project science advisory panel.

■ REFERENCES

- (1) Alvarez, R. A.; Pacala, S. W.; Winebrake, J. J.; Chameides, W. L.; Hamburg, S. P. Greater focus needed on methane leakage from natural gas infrastructure. *Proc. Natl. Acad. Sci. U. S. A.* **2012**, *109* (17), 6435–6440 DOI: 10.1073/pnas.1202407109.

(2) Howarth, R. W.; Santoro, R.; Ingraffea, A. Methane and the greenhouse gas footprint of natural gas from shale formations. *Clim. Change* **2011**, *106* (4), 679–690.

(3) EPA. *Inventory of US Greenhouse Gas Emissions and Sinks: 1990–2012*; US Environmental Protection Agency: Washington, DC, 2014. <http://epa.gov/climatechange/ghgemissions/usinventoryreport.html>.

(4) Petron, G.; Frost, G.; et al. Hydrocarbon emissions characterization in the Colorado front range: A pilot study. *J. Geophys. Res.: Atmos.* **2012**, *117*, D04304 DOI: 10.1029/2011JD016360.

(5) Miller, S. M.; Wofsy, S. C.; et al. Anthropogenic emissions of methane in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110* (50), 20018–20022 DOI: 10.1073/pnas.1314392110.

(6) Brandt, A. R.; Heath, G. A.; et al. Methane leaks from North American natural gas systems. *Science* **2014**, *343*, 733–735.

(7) Harrison, M. R.; Shires, T. M.; Wessels, J. K.; Cowgill, R. M. Methane emissions from the natural gas industry. *GRI-94/0257 and EPA-600/R-96–080, Final Report*; Gas Research Institute and Environmental Protection Agency: Washington, DC, 1996; Vol. 1–15.

(8) Allen, D. T.; Torres, V. M.; et al. Measurements of methane emissions at natural gas production sites in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110* (44), 17768–17773 DOI: 10.1073/pnas.1304880110.

(9) ERG *City of Fort Worth: Natural Gas Air Quality Study (Final Report)*; Eastern Research Group, Inc.: Morrisville, NC, 2011.

(10) Harrison, M. R.; Galloway, K. E.; Hendler, A.; Shires, T. M.; Allen, D. T.; Foss, M.; Thomas, J.; Spinhirne, J. Cooperative Agreement No. XA-83376101. *Natural Gas Industry Methane Emission Factor Improvement Study: Draft Final Report*; US EPA: Washington, DC, 2011.

(11) Moore, C. W.; Zielinska, B.; Petron, G.; Jackson, R. B. Air impacts of increased natural gas acquisition, processing, and use: A critical review. *Environ. Sci. Technol.* **2014**, *48* (15), 8349–8359 DOI: 10.1021/es4053472.

(12) Zimmerle, D.; Williams, L.; Vaughn, T.; Subramanian, R.; Duggan, J.; Wilson, B.; Opsomer, J.; Robinson, A. L. Methane emissions from the natural gas transmission and storage system in the United States. *Environ. Sci. Technol.*, **2015**, in preparation.

(13) Lamb, B.; Shorter, J. H.; et al. Development of atmospheric tracer methods to measure methane emissions from natural gas facilities and urban areas. *Environ. Sci. Technol.* **1995**, *29*, 1468–1479.

(14) Shorter, J. H.; McManus, J. B.; Kolb, C. E.; Allwine, E. J.; Siverson, R.; Lamb, B. K.; Mosher, B. W.; Harriss, R. C.; Howard, T.; Lott, R. A. Collection of leakage statistics in the natural gas system by tracer methods. *Environ. Sci. Technol.* **1997**, *31*, 2012–2019.

(15) Czepiel, P. M.; Shorter, J. H.; Mosher, B.; Allwine, E. J.; McManus, J. B.; Harriss, R. C.; Kolb, C. E.; Lamb, B. K. The influence of atmospheric pressure on landfill methane emissions. *Waste Manage.* **2003**, *23*, 593–598.

(16) Roscioli, J. R.; Yacovitch, T. I.; et al. Measurements of methane emissions from natural gas gathering stations and processing plants—Part 1: Measurement methods. *Atmos. Meas. Technol. Discuss.* **2014**, *7*, 12357–12406 DOI: 10.5194/amtd-7-12357-2014.

(17) Yacovitch, T. I.; Herndon, S. C.; et al. Demonstration of an ethane spectrometer for methane source identification. *Environ. Sci. Technol.* **2014**, *48* (14), 8028–8034 DOI: 10.1021/es501475q.

(18) Heywood, J. B. *Internal Combustion Engine Fundamentals*; McGraw-Hill: Burr Ridge, IL, 1988.

(19) Lefebvre, A. H. *Gas Turbine Combustion*; CRC Press: Boca Raton, FL, 1998).