Environmental Science & Technology

Article

pubs.acs.org/est

Measurements of Methane Emissions from Natural Gas Gathering Facilities and Processing Plants: Measurement Results

Austin L. Mitchell,[†] Daniel S. Tkacik,[†] Joseph R. Roscioli,[‖] Scott C. Herndon,[‖] Tara I. Yacovitch,[‖] David M. Martinez,[‡] Timothy L. Vaughn,[‡] Laurie L. Williams,[⊥] Melissa R. Sullivan,[†] Cody Floerchinger,[‖] Mark Omara,[†] R. Subramanian,[†] Daniel Zimmerle,[§] Anthony J. Marchese,[‡] and Allen L. Robinson*[,†,#]

[†]Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, United States

[‡]Department of Mechanical Engineering, Colorado State University, Fort Collins, Colorado 80523, United States

[§]The Energy Institute, Colorado State University, Fort Collins, Colorado 80523, United States

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

[⊥]Fort Lewis College, Durango, Colorado 81301, United States

Supporting Information

ABSTRACT: Facility-level methane emissions were measured at 114 gathering facilities and 16 processing plants in the United States natural gas system. At gathering facilities, the measured methane emission rates ranged from 0.7 to 700 kg per hour (kg/h) (0.6 to 600 standard cubic feet per minute (scfm)). Normalized emissions (as a % of total methane throughput) were less than 1% for 85 gathering facilities and 19 had normalized emissions less than 0.1%. The range of methane emissions rates for processing plants was 3 to 600 kg/h (3 to 524 scfm), corresponding to normalized methane emissions rates <1% in all cases. The distributions of methane emissions, particularly for gathering facilities, are skewed. For example, 30% of gathering facilities contribute 80% of the total emissions. Normalized emissions rates are negatively correlated with facility throughput. The variation in methane emissions also appears driven by differences between inlet and outlet pressure, as well as venting and leaking equipment. Substantial venting from liquids storage tanks was observed at 20% of gathering facilities. Emissions rates at these facilities were, on average, around four times the rates observed at similar facilities without substantial venting.

INTRODUCTION

Methane is the primary component of natural gas; it is also a potent greenhouse gas (GHG). The Environmental Protection Agency (EPA) estimates that the natural gas system contributes 23% of U.S. anthropogenic methane emissions.[1] However, there are discrepancies between recent studies and EPA GHG inventories in some natural gas producing areas.[2−4] The EPA GHG inventories largely rely on data collected in the early 1990s[5] and may not reflect recent changes in technology, operations, and regulation. New measurements are needed to characterize methane emissions from the natural gas system.

This study investigates the methane emissions from natural gas gathering and processing (G&P) facilities, which, collectively, gather natural gas from production wells, remove impurities, and deliver it to inter- and intrastate pipeline networks. We define gathering and processing as the equipment and pipeline network between the sales points at well pads and downstream delivery points. This includes gathering pipelines, and the equipment at gathering facilities: compressors (driven by electric motors ("motors") and/or natural gas-fired internal combustion engines ("engines") or turbines), dehydration systems to remove water, and treatment systems to remove hydrogen sulfide and/or carbon dioxide. Processing plants often house this equipment on a larger scale, acting as central nodes in a system of smaller gathering facilities. Processing plants also separate natural gas liquids (NGLs) (such as ethane, propane, butane, and heavier hydrocarbons) from methane. For this study, processing plants are defined as the facilities that meet the 40 CFR Part 60 Subpart KKK definition of "natural gas processing plant" based on the presence of NGL extraction. Facilities that only fractionate NGLs were not included in this study.

This paper presents facility-level measurements of methane emission rates at 130 G&P facilities (114 gathering and 16 processing). A mobile laboratory was used to perform downwind tracer flux measurements, which is an established technique to estimate the total emissions of methane (or other

Received: October 28, 2014
Revised: January 23, 2015
Accepted: January 28, 2015
Published: February 10, 2015

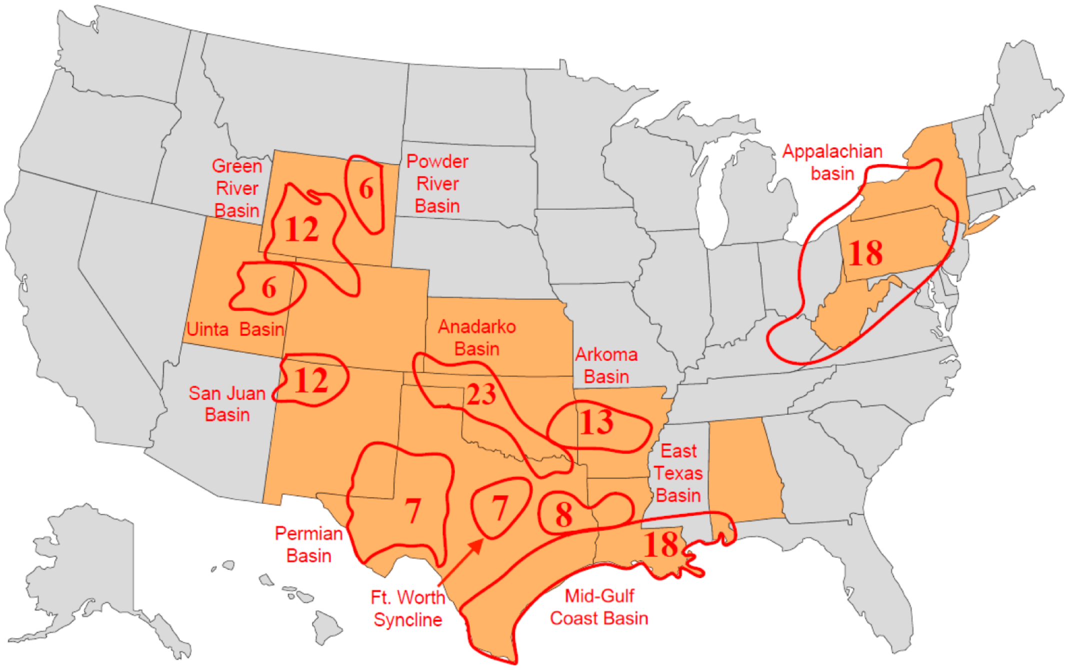

Figure 1. Locations of G&P facilities measured in this study. Numbers indicate the number of facilities sampled within outlined (red) oil and gas basins as defined by the American Association of Petroleum Geologists.[26] G&P facilities were sampled in each of the orange-colored states (www.map-generator.org).

species) from complex facilities with multiple emission points.[6−8] Onsite observations with an infrared camera were used to identify emission sources. Companion papers describe the downwind tracer flux methodology and its application to this study[9] and scaling of these data to estimate emissions for all U.S. G&P facilities.[10]

2. METHODS

Downwind tracer flux measurements were conducted at 136 G&P facilities between October 2013 and April 2014. Measurements were successful at 130 G&P facilities. These facilities are located in top gas producing states (including all of the top ten) and basins (Figure 1). The sampled facilities were operated by five natural gas midstream "partner" companies, which provided access and facility data to the study team. The project only investigated emissions from G&P facilities, not the gathering pipeline network that connects these facilities.

2.1. Selection of G&P Facilities for Measurement. The sampling strategy sought to obtain methane emissions data from a wide range of G&P facilities, which vary by age (reflecting technological and regulatory changes), inlet gas composition/pressure, regulatory environment, and regional/local and/or corporate design and operations. In most states, the basic census data needed to design a sampling program that systematically captures variation among G&P facilities are lacking. Therefore, the study team sought to replicate the diversity among G&P facilities owned by the five partner companies. They operate more than 700 gathering facilities and 27 processing plants in all regions of the country—roughly one-quarter and 5%, respectively, of all gathering and processing facilities nationally (Supporting Information (SI) A). All of these facilities were considered by the study team in developing the project sampling plan. This plan sought to (1) sample at the maximum number of processing plants, (2) maximize the number of successful measurements in the top gas producing regions and across all major gas types (e.g., shale, coal bed methane, etc.), and (3) sample a distribution of facility types representative of the partner company assets.

Details of the facility selection procedures are in SI A. Briefly, the study team was provided a list of all partner company G&P assets (>700 facilities). The study team performed desktop screening of the facilities using aerial imagery (Google Earth) to remove facilities with limited road access on their periphery (desired range of 0.5−2 km) or facilities that were located near obvious potential sources of methane interference. The list of prescreened facilities was then shared with partner companies, with a request for additional feedback regarding potential issues with road access or nearby nonpartner facilities, as needed. Over the course of the field campaign, fewer than ten facilities were removed from the prescreened lists by request of the partner companies. Reasons for removal included issues such as commissioning, decommissioning, transactions (sale of asset), construction, or litigation. Final selection for most gathering facilities was made by the study team from the prescreened list the evening before or the day of sampling, considering road access, wind forecasts, and logistics. Sampling at processing plants was scheduled in advance to align with operator and operations schedules.

The tracer teams were usually escorted to the target facilities by a company representative who was instructed to continue normal operations of the facility. At 14 facilities the study team onsite observer noted that the company personnel adjusted processes or fixed equipment during the sampling, presumably these changes were performed as part of normal operations (SI E). At only one facility (#44) did a tracer team note changes in facility operation that were likely not normal practice as the changes were undone after measurements had been completed. Company representatives were notified. All indications are that this was an isolated occurrence.

Partner company gathering assets were classified based on three equipment-specific functions in gathering systems

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219−3227

Table 1. Summary of Sampled Facilities and Partner Assets (total and %) by Facility Type[a]

	facility type											
	C		C/D		C/D/T		D		D/T		P	
sampled	34	26%	66	51%	8	6%	5	4%	1	1%	16	12%
partner assets	267	36%	379	51%	16	2%	23	3%	28	4%	27	4%

[a]C, compression only; C/D, compression and dehydration; C/D/T, compression, dehydration, and treatment; D, dehydration only; D/T, dehydration and treatment; and P, processing.

(compression ("C"), dehydration ("D"), and treatment ("T")) into five facility types (C, C/D, C/D/T, D, and D/T). Table 1 summarizes facility types sampled by the tracer teams compared to all partner company G&P assets as of August 2013. The distribution of facility types sampled in this study closely resembles the partner assets. The study facilities are also similarly proportioned to lower-48 U.S. gas production:[11] Fifty-two percent of the study facilities handled shale gas, 32% conventional, 8% coal-bed methane, 6% tight sands, and 2% offshore.

Detailed information on all 130 G&P facilities is included in SI B, Tables S1−S3. Sampling was conducted at 34 C facilities, which had reciprocating and/or centrifugal compressors. Engines were the primary driver at 31 of the C facilities. Scrubbers, strainers, filter separators, and/or coalescing filters were usually installed upstream of compressors. Produced water and condensate removed from the gas stream were stored in tanks onsite. Air and/or gas pneumatic devices (e.g., liquid level controllers) were also present at all G&P facilities.

Sampling was conducted at 66 C/D facilities, which had dehydration equipment installed downstream of compressor(s) to remove vapor-phase water from the pressurized gas stream, reducing problems with hydrate formation and corrosion.[12] These were typically glycol dehydration systems which operate by contacting the gas stream (via absorbers) with a hydroscopic solvent (usually triethylene glycol), which is regenerated (by heating) and reused.[13]

Sampling was performed at five D only facilities, which represent 4% of partner gathering assets. These removed vapor-phase water from gas produced at nearby wells that had sufficient pressure to move downstream without compression.

Treatment (T) equipment was installed at gathering facilities that handle a raw gas stream rich in acidic gases, carbon dioxide (CO_2) and/or hydrogen sulfide (H_2S). Acid gas removal is based on absorption with one or more amine variants, separation, and regeneration of the amine solution. Treatment equipment were utilized at 6% of partner facilities. Eight C/D/T facilities and one D/T facility were sampled, representing 6% and 1% of study facilities, respectively.

Processing plants (P) are central nodes in most producing areas. They are generally much larger than the typical gathering facilities and are staffed by operators 24 h a day. Compression, dehydration, and treatment equipment are typical for processing plants. These plants also include equipment to separate natural gas liquids from the methane gas stream, such as cryogenic turboexpander and refrigeration skids. These plants will sometimes use fractionation processes to further separate high-value natural gas liquids into separate components of ethane, butane, propane, and hydrocarbons larger than pentane. The study team ruled out conducting measurements at 11 (out of 27) processing plants mainly due to the presence of interfering natural gas facilities (usually other processing plant(s)) owned by a different company.

Figure S1 (SI C) compares facility throughput, inlet and outlet pressure, and total horsepower for C, C/D, C/D/T, and P facilities. The C facilities included in this study had generally lower throughput, inlet and outlet pressures, and horsepower then the other types of facilities with compressors. Methane content ranged from 59% to 98% (mol/mol) across all facilities, but was not related to facility type. Gas composition data are summarized in SI C, Figure S2.

Despite efforts to prescreen facilities, the study team conducted measurements at 29 facilities that were either collocated (e.g., on the same pad or side-by-side) with nonpartner equipment or adjacent to other natural gas facilities. Nonpartner equipment was generally associated with gas production (e.g., wellheads, produced water and/or condensate tanks, flares, etc.). Both infrared camera surveys and proximal mobile lab measurements (immediately downwind of equipment) were performed to assess potential contribution of nonpartner equipment. At most facilities with nonpartner equipment, the methane contribution from such equipment appeared to be negligible in comparison to partner-operated equipment. Therefore, methane emissions estimates have not been modified to account for potential emissions from collocated nonpartner equipment. Facilities with collocated equipment and information on this equipment are summarized in SI D, Table S4.

2.2. Tracer Flux Measurements and Facility Survey. Roscoli et al.[9] describes the tracer flux methodology used by this study in detail. Briefly, at each facility, two tracer gases (nitrous oxide and acetylene; both chosen based on their availability and ability to be measured at high time-resolution) were released at known flow rates from points configured to bracket the target facility's emissions relative to prevailing wind direction (SI G, Figure S3). Concentrations of methane, ethane, acetylene, nitrous oxide, carbon monoxide, and carbon dioxide were measured using a mobile laboratory equipped with high time resolution instrumentation (1-Hz or faster, Aerodyne QC-TILDAS and/or Picarro cavity ring-down spectroscopy).[9] Measurements were made immediately upwind of each target facility to check for background contamination. Methane plumes were measured approximately 0.5−3 km downwind from the target facility. Multiple plume profiles, each about 30−120 s long, were obtained for each facility by driving the mobile laboratory at a constant speed on a road roughly perpendicular to the wind direction. When the winds were highly variable, plumes were sometimes measured while the mobile lab was stationary. Downwind measurements in the mobile lab were performed over 1−5 h period at most facilities.

While tracer flux measurements were being performed, a dedicated member of the study team served as an "onsite observer," who documented the initial operating state of the facility and any changes or repairs that were made (SI E, Table S5). The onsite observer also performed a comprehensive survey of the facility, including recording an equipment census (operational state and characteristics such as horsepower), and

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219−3227

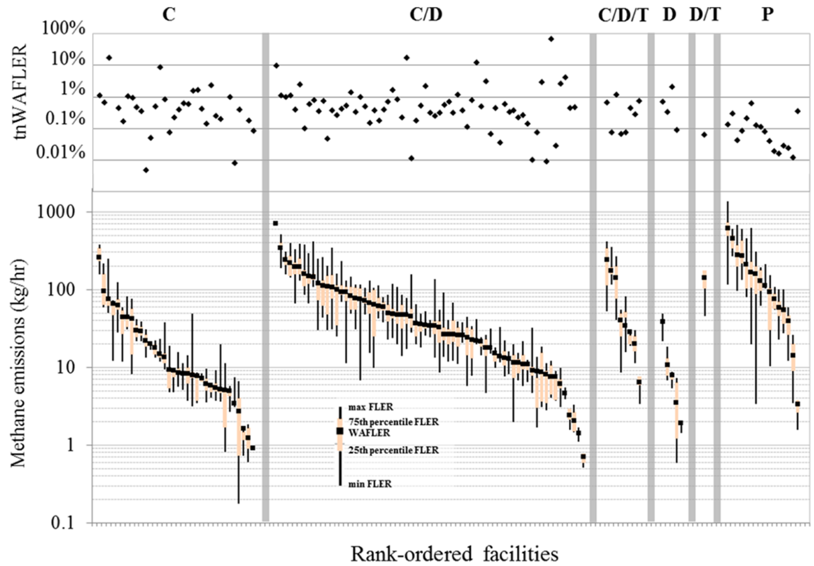

Figure 2. Tracer flux-based facility-level methane emission rates (FLER) for 130 G&P facilities. Facilities are sorted by type (C, D, C/D, etc.) and then rank ordered by WAFLER (square) within each facility type. Emissions are presented in log scale. The vertical boxes represent the 25th and 75th percentiles of FLER data at a facility; whiskers represent the min and max. Throughput-normalized methane emissions rates (tnWAFLER) for 124 facilities (as % CH_4 throughput on the day of sampling) are presented in the top panel in log scale.

readings of facility throughput, inlet and outlet gas pressure (SI B, Tables S1−S3). At 108 of the gathering facilities, the onsite observer also performed an infrared camera survey of all equipment to identify and document potential sources noncombustion methane emissions (SI F, Table S6). Gas composition data (% methane, ethane, etc.) were obtained from company records (SI B, Table S1).

2.3. Analysis of Tracer Flux Data. The tracer flux data were analyzed systematically to quantify the methane emission rate, ensure proper attribution of methane emissions to target facility, and validate codispersion of tracer(s) with methane. The methane facility-level emission rate (FLER) was calculated for each downwind plume based on tracer release rate and the background-corrected methane-to-tracer concentration ratios.[9] No modeling of pollutant dispersion is required—that complexity is empirically captured by the tracers. Therefore, the FLER is an estimate of the aggregate (or total) methane emissions from the target facility during a 30−120 s period (the typical duration of a plume transect). Roscioli et al.[9] describes the types of plumes measured, the acceptance criteria, and the process used to analyze each plume.

Multiple downwind plumes were obtained at each facility and the FLER was estimated for each plume. The weighted-average FLER (WAFLER) was calculated by weighting each FLER estimate by its estimated plume-specific uncertainty ($1/\sigma^2$), providing more weight to the plume types meeting the most stringent acceptance criteria.[9] The weighted sample variance of WAFLER was divided by a correction factor $[1 - ((\Sigma_{i=1}^{n} w_i^2)/(\Sigma_{i=1}^{n} w_i)^2)]$ to obtain the unbiased weighted sample variance, where w_i represents the weight of each plume. Complete results, including number of accepted plumes, distance downwind, 25th, 75th, minimum and maximum FLER, WAFLER, and the unbiased standard deviation of WAFLER are in SI H, Tables S7−S8. All emissions results are presented as grams CH_4 (19.26 g per standard cubic foot CH_4 at 15.6 °C and 1 atm[14]).

Sampling occurred at one gathering facility in two distinct operating states, before and after substantial venting from a pipe at the top of a tank storing liquids was stopped. The venting was stopped by closing a dump valve on the first stage scrubber that was stuck open. Two WAFLER estimates are reported for this facility (#'s 35 and 59), based on the before and after measurements, respectively.

The tracer flux method assumes that the tracer gases codisperse with the methane being emitted from the target facility. Since the tracer release points were set up to bracket the facility, the use of two tracers provides a stringent test on the horizontal dispersion of the emissions. 75% of accepted plumes were dual tracer, covering 112 facilities (83% of the gathering facilities and 100% of the processing plants). Co-dispersion of tracer gases with methane emissions from elevated vents, flares, and exhaust stacks in the vertical plane is more difficult to verify. Roscioli et al.[9] modeled tracer codispersion with the exhaust of natural gas-powered engines and turbines using Gaussian dispersion modeling with Briggs plume rise equations.[9,15] Exhaust is discharged at high temperatures (up to 540 °C) and high velocities (up to 45 m/s). A conservative, worst-case analysis suggests the potential bias in WAFLER from low recovery of methane emissions in engine or turbine exhaust is small (<30%) for almost four-fifths of the G&P facilities in this study (SI J). Processing plants (but typically not gathering facilities) can also have elevated flares, vents, or other emissions sources; the potential bias due to partial recovery of methane from these sources was not examined in this study.

3. RESULTS

The study team recorded more than 1400 plumes which passed the acceptance criteria[9] and therefore provided a valid measure of the facility-level emission rate (FLER) of methane. Roscioli et al.[9] provides plots of example plume profiles. Within a plume, methane concentrations are tens to thousands of ppbv above background. The average number of plumes per facility is 11 with a range of 2−42. Each plume provides a short snapshot of the facility-level emissions over the duration of the plume transect (30−120 s). The plume-to-plume differences in the

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219−3227

FLER values provide a measure of the variability of the methane emissions at a given facility.

Figure 2 presents box-and-whisker plots of the accepted plumes for all 130 facilities (131 WAFLERs). The facilities have been sorted by type (C, D, C/D, etc.) and then rank ordered by WAFLER within each facility type. Methane (CH_4) throughput (tonnes/h) was calculated from natural gas throughput (tonnes/h) recorded onsite and the inlet gas composition data provided by the companies. The CH_4 throughput-normalized WAFLER (tnWAFLER), ratio of WAFLER to the CH_4 throughput, is also plotted in Figure 2 for 124 facilities (125 tnWAFLER estimates; five facilities had zero throughput and no throughput data were recorded at one facility).

Across all 114 gathering facilities, WAFLER ranged from 0.7 to 700 kg/h, while tnWAFLER ranged from 0.004% to 70% for the subset of the facilities with nonzero/known CH_4 throughput (n = 108). The WAFLER estimates varied widely within and across facility types. However, in general, the plume-to-plume variability in FLER estimates at a given facility is relatively small; the median relative standard deviation or coefficient of variation (CV) is 0.36 and more than two-thirds of the facilities had CVs less than 0.5. This suggests that the emissions from most facilities were relatively stable over the several hour measurement period. The CV at 11 facilities is >0.8. Large CVs may be associated with intermittent sources (e.g., pressure relief valves) and/or changes in facility operation. For example, FLER estimates at Facility #102 (CV 0.62) were much higher after the onsite observer documented increased flame height and smoke production at the flare as well as significant flashing events at the condensate tank batteries. This facility was shut down for maintenance after measurements were completed. At five facilities, a brief methane release event was observed which was associated with specific operations such as a compressor blowdown or startup. All of these events that were measured are cataloged in SI Table S9. They only occurred for a small fraction of the overall measurement period and therefore were not included in the WAFLER calculations.

WAFLER estimates for C/D and C/D/T are higher than those C and D facilities. For example, the median WAFLER for C and D facilities are 8.8 and 7.8 kg/h, respectively, while those of C/D and C/D/T facilities were 35 and 37 kg/h, respectively. However, across the gathering facility types there are no systematic trends in the tnWAFLER because C/D and C/D/T facilities also have higher throughputs. Median tnWAFLER for C, C/D, C/D/T, and D facilities are 0.46, 0.45, 0.36, and 0.51% of CH_4 throughput, respectively.

Figure 2 indicates that methane emissions are generally a small fraction of CH_4 throughput. tnWAFLER is less than 1% at 85 (out of 108) gathering facilities and less than 0.1% at 19 of them. tnWAFLER exceeded 5% at six gathering facilities (two C and four C/D). Five of these had very low CH_4 throughput (<0.5 tonnes/h). The exception is facility #35 (throughput: 7.7 tonnes/h), which was venting a substantial amount of gas when sampling started. At facility #94, which had the highest tnWAFLER (70%) and the lowest throughput (1.4×10^{-2} tonnes/h), a substantial leak from a pipe union was observed.

Median WAFLER and tnWAFLER of the 16 processing plants are 120 kg/h and 0.079%, respectively. With one exception, the processing plants were high CH_4 throughput facilities, averaging 250 tonnes/h on the day of measurements (range 1.6−780 tonnes/h; SI B, Table S2). WAFLER at processing plants ranged from 3 to 600 kg/h. tnWAFLER for processing plants were lower than for gathering facilities, ranging from 0.012% to 0.62% of CH_4 throughput.

Partial recovery of lofted emissions is a concern at processing plants, most of which had high flares as well as elevated exhaust stacks, vents, and other components that may emit methane. Using previously mentioned dispersion modeling, we estimate that, under conservative, worst case assumptions, only two (out of 16) processing plants may have >30% bias in WAFLER due to low recovery of uncombusted methane in engine and/or turbine exhaust (facility #'s 123 and 126, SI J). The potential bias due to low recovery of exhaust methane at processing plants is lower than gathering facilities because the majority of the compression power at processing plants in this study was provided by turbines or motors, which have much lower exhaust methane emissions than engines.[16,17] For example, all of the compressors at four other processing plants were driven by either turbines (facility #'s 118, 127, and 129) or motors (facility #125). Therefore, low recovery of exhaust methane for these plants is a small issue despite the fact that all of the tracer flux measurements were performed <0.5 km downwind from these plants, which was not optimal for codispersion of lofted methane and tracers. A larger concern may be methane emissions associated with high flares and elevated vents or leaks, but these have not been investigated by this study.

The cumulative methane emissions rate summed across all 114 gathering facilities in this study was 6300 kg/h with a cumulative tnWAFLER ($\sum$WAFLER/$\sum CH_4$ throughput) of 0.20%. Cumulative methane emissions from all 16 processing plants was 2,700 kg/h, with a cumulative tnWAFLER of 0.075%. Accounting for potential biases in WAFLER due to uncombusted methane emissions from gas-driven engines and turbines only has a small impact on cumulative emissions (SI J). For example, adding the conservative, worst-case estimate of unrecovered exhaust methane to WAFLER would lead to cumulative methane emissions from all gathering facilities to 7400 kg/h, versus 6300 kg/h previously, and increase the cumulative tnWAFLER from 0.2% to 0.24%. Unrecovered exhaust methane emissions from processing plants would increase from 2700 kg/h to 3100 kg/h, and cumulative tnWAFLER would increase from 0.075% to 0.085%. While recovery of methane emissions from high flares or elevated sources common at processing plants is uncertain, it seems unlikely that such emissions could be large enough to significantly alter normalized emissions at high-throughput processing plants.

Five gathering facilities (#4, 28, 32, 100, and 101) had zero throughput, but were still emitting methane (65, 5.1, 1.6, 1.4, and 0.7 kg/h, respectively). The highest WAFLER (65 kg/h) at a zero-throughput facility was attributed to atmospheric venting from two liquids storage tanks via infrared camera survey. Unlike the other zero-throughput facilities, facility #4 was flowing gas before measurements began. Emissions at the other zero throughput facilities were observed from various sources, including venting from a liquids storage tank and an engine fuel purge (facility #28) and a broken hose leading to an nonoperational pressurized (NOP) compressor (facility #32). The only sources of methane emissions observed at facility #101 with the infrared camera were collocated nonpartner equipment (a dehydration unit and a fuel scrubber).

4. DISCUSSION

4.1. Comparison to Existing G&P Emissions Data.

Relatively little data have been published on methane emissions

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219−3227

from G&P facilities. One of the GRI/EPA studies reported methane emissions at seven processing plants in the TX/LA and West regions.[8,18] More recent measurements, using comprehensive onsite measurements (including exhaust methane), have been made at nine processing plants.[19,20] The reported methane emissions rates ranged from 45 to 840 kg/h (this study: 3−600 kg/h CH_4). The cumulative throughput-normalized methane emissions rate was 0.16% versus 0.075% for the 16 plants reported here. Higher normalized emission rates were reported in the GRI/EPA study, which also used the tracer flux method. The cumulative methane emissions rate normalized by cumulative plant capacity (throughput data were not given) for the GRI/EPA data was 0.38%. This is likely a lower bound estimate because processing plants may operate below rated capacity.

Even fewer data are available for gathering facilities. The GRI/EPA study measured methane emissions from two gathering facilities of 86 and 120 kg/h (this study: <1 to 700 kg/h CH_4). No throughput or capacity data were reported for GRI/EPA facilities, precluding comparison of normalized emission rates. Measurements were performed at seven gathering facilities between 2004 and 2005, but only summary data were reported.[20] The raw data from previous studies are reproduced and additional comparisons are made in SI K.

All of the processing plants included in this study have reported methane emissions to the EPA greenhouse gas reporting program (GHGRP, 40 CFR 98, Subpart W and Subpart C). Valid comparisons of WAFLER to GHGRP are not possible due to a range of issues in how emissions are calculated under GHGRP. For example, GHGRP does not require reporting of all onsite methane emissions sources (including tanks, acid gas removal units, and gas-driven pneumatics); it excludes other sources in certain operating modes, such as rod-packing vents from NOP reciprocating compressors; and it includes blowdown emissions.[21] Furthermore, combustion emissions reported under Subpart-C may not provide a realistic characterization of uncombusted methane from operating engines.[22]

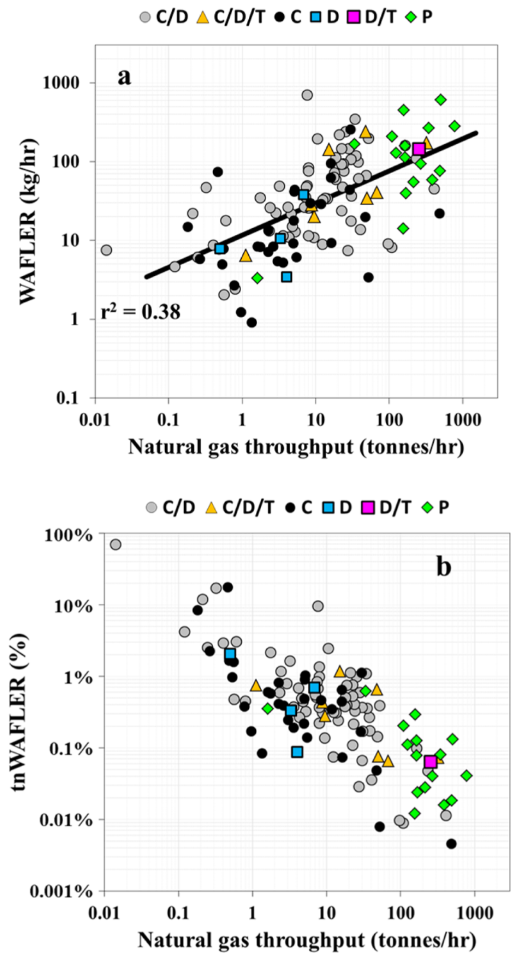

Figure 3. (a) WAFLER and (b) tnWAFLER (% CH_4 throughput) versus reported natural gas throughput on the day sampling was conducted in log−log scale. The line in (a) is a linear regression of the data.

4.2. Sources of Methane Emissions. Figure 3 indicates that there is some relationship between WAFLER and tnWAFLER and facility natural gas throughput. The absolute methane emissions are generally higher at facilities with larger throughput, but the normalized methane emissions rates (tnWAFLER) generally decrease with increasing natural gas throughput. The opposite relationship of WAFLER and tnWAFLER with throughput (Figure 3) indicates that some of the methane emissions are likely independent of throughput. A leak, for example, may emit the same amount of methane whether the facility is at full or partial capacity.

One-third of the variance (r^2 = 0.38) in WAFLER is explained by linear regression with throughput, Figure 3(a). Although throughput explains some of the trend in the emissions data, WAFLERs vary by about an order of magnitude at facilities handling similar volumes of gas. This underscores that there are many factors influencing emissions. Numerous noncombustion emissions sources were documented by the onsite observer via infrared camera survey, which was conducted at 108 gathering facilities (infrared cameras were not used at processing plants, which are subject to regular leak detection and repair (LDAR) for regulatory compliance). Observable emissions sources (leaking and/or venting) were noted at 71 out of 108 gathering facilities (SI F, Table S6). This included venting (flashing and off-gassing) from liquids storage tanks at 48 facilities, leaking or venting from compression equipment at 42 facilities, and gas pneumatics at 28 facilities.

On the basis of the infrared camera videos and other onsite information, a subset of the observed noncombustion emission sources have been classified as substantial (SI M, Tables S17 and S18). These specific sources appeared to be emitting substantially more methane than other sources of the same type (e.g., valves and vents). Since the infrared camera only provides a qualitative indication of the magnitude of the emissions, classification of a source as substantial also involved discussion with company representatives and, if possible, proximal methane measurements using the mobile laboratory. The onsite observer and/or company representative was often able to identify the substantial emission sources without the aid of an infrared camera.

Venting from liquids storage tanks was the most common noncombustion source classified by the onsite observer. It is normal for some flashing or off-gassing from stored liquids to occur. Tank venting, however, may not have always been caused by an issue with the liquids storage tanks themselves, but to a problem elsewhere on the facility (e.g., the stuck dump valve mentioned earlier) that was connected to the tanks. Of the 48 gathering facilities with observable venting from liquids storage tanks, substantial venting from tanks was observed at 23 (SI M). Abnormal process conditions were identified at six of

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219−3227

the 23 facilities. Through the strategic placement of the tracer release locations, the methane emissions rates from liquids storage tanks were estimated at five facilities with substantial venting. These tank-related methane emissions ranged from around 10 to 650 kg/h (SI Table S17). At three gathering facilities (#38, 61, and 76) a company representative made adjustments that had a noticeable impact on tank venting (SI Table S5). Other substantial sources included leaking valves and pipe unions, venting from the dehydrator, and compressor crank case/rod packing vents (SI Table S18). Some of the issues observed in the field (e.g., stuck valves or open thief hatches) that were causing substantial methane venting were addressed by the company representative without interrupting operations. At a few facilities, emissions sources had already been tagged (usually a ribbon) by prior inspections but the issue had not yet been addressed.

Multiple linear least-squares regressions were performed to quantify the contribution of different factors to the WAFLER at gathering facilities (SI M). Only gathering facilities with positive throughput were included in the regression. Variables describing engine and turbine horsepower, facility throughput, and pressure were significant in all models tested. The independent variables in the final regression model were natural log of throughput, delta pressure, and dummy variables for facilities with turbine-driven compressors (n = 7) and facilities classified with substantial venting from liquids storage tanks (n = 22; one facility with substantial tank venting had zero throughput). Horsepower was not included in the final model because it was highly correlated with throughput, which made the individual contribution of these variables difficult to resolve. The adjusted r^2 of this model is 0.67 and the Root Mean Square Error is 0.754 (diagnostic plots in SI M, Figures S8−S11).

For the 22 gathering facilities flagged by the onsite observer for substantial methane emissions from liquids storage tanks, the average WAFLER is 300% higher than nonflagged facilities. In other words, facilities classified as having substantial methane emissions from liquids storage tanks (~20% of gathering facilities) have around four times the methane emissions than similar, nonflagged facilities.

The regression model also indicates that the WAFLER at facilities with turbine-powered compressors was, on average, 75% lower than at facilities with only engine-powered compressors. The cumulative tnWAFLER among the seven facilities with turbines is <0.01% of CH_4 throughput. It is not known if the lower emissions at these facilities were due solely to the use of turbines, which have much less exhaust methane than engines, or if it is indicator for some other characteristics of facilities equipped with turbines. Lower methane emissions were also observed from turbine equipped facilities in the transmission and storage sector.[22]

Finally, the regression model indicates that the WAFLER increased with the pressure difference across the facility. The magnitude of some fugitive leaks scale with pressure. The number of stages of compression also increases with delta pressure.

4.3. Distribution of Methane Emissions. Figure 4 presents cumulative distributions of WAFLER (as a percentage of the total methane emissions) separately for the gathering (C, C/D, C/D/T, D, D/T) facilities and processing plants. Distributions are presented on two bases: facility number (Figure 4(a)) and cumulative CH_4 throughput (Figure 4(b)).

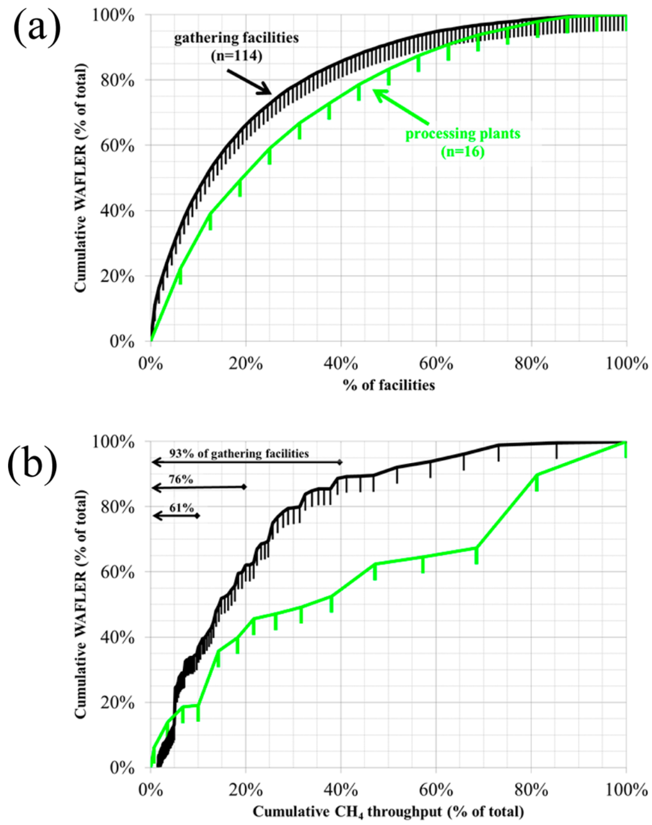

Figure 4. Skew in the methane emissions data illustrated by cumulative distributions of WAFLER (as a % of total methane emissions) of gathering facilities (black) and processing plants (green) sorted in (a) by WAFLER in descending order and (b) by CH_4 throughput (as a % of total) in ascending order. Each tick represents a single facility, and, in (b), the space between ticks represents % of total throughput accounted for by each facility. In (a), more than 80% of emissions come from about 30% of gathering facilities. In (b), the percentage of gathering facilities that contribute 10, 20, and 40% of throughput are indicated by arrows (top left). For example, 76% of the gathering facilities contribute only 20% of the throughput, but account for more than 60% of the emissions. (Included are three C facilities and two C/D facilities that had zero throughput but were still emitting methane.)

The cumulative distributions of WAFLER are skewed, similar to methane emissions data for production wells[23,24] and transmission and storage facilities.[22] In Figure 4(a), the skew is demonstrated by the fact that a minority of facilities contribute to the majority of the emissions. For example, less than 30% of gathering facilities (37 total) are responsible for almost 80% of the total methane emissions. One facility (facility #35) accounts for 10% of total gathering methane emissions. The five zero throughput facilities contributed 1% of the total gathering methane emissions.

Figure 4(b) shows that the emissions are also skewed when viewed from a CH_4 throughput basis. There are many low throughput facilities—61% of the facilities only contributed 10% of the total CH_4 throughput, but almost 40% of the total gathering methane emissions. At the other end of the distribution, nine facilities accounted for more than 60% of the total gathering throughput, but emissions from these nine facilities were only around 10% of the total gathering methane emissions. Without these nine facilities, the cumulative tnWAFLER of all study gathering facilities would increase

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219−3227

from 0.20% to 0.43%. Cumulative distributions for C and C/D gathering facilities are included in SI N.

Normalized emissions from processing plants were generally lower than gathering facilities (Figure 3(b)) and their cumulative distributions of WAFLER less skewed (Figure 4). One factor may be the LDAR programs employed at processing plants, which are intended to reduce emissions. It is also unlikely that a single, high-magnitude leak would go unnoticed at any processing plant because they are typically staffed by an operator at all times. For example, a 1% leak at the average processing plant (250 tonnes/h) would emit more than 2500 kg/h. A single emission source at even a fraction of this magnitude would likely be visible and/or audible, making detection without infrared camera survey possible. Of the 25 facilities with the lowest tnWAFLER, 19 were staffed by full-time operator(s) (8–24 h/day). All of the processing facilities, but only about 14% of gathering facilities, were staffed. The other facilities were checked by their operator at least a few times a week, most daily.

The skewed distributions shown in Figure 4 are important to consider when making national emissions estimates.[25] These data provide insight into the frequency of high emitters (the "fat tail"). However, the sensitivity of the normalized emissions rates presented here to the skewed distributions complicates comparisons to existing GHG inventories. Marchese et al.[10] use these emissions data to estimate the total methane emissions from natural gas gathering and processing in the U.S.

■ ASSOCIATED CONTENT

Supporting Information

A, Facility selection process; B, facility information collected by onsite observer; C, summary figures of onsite information; D, nonpartner equipment collocated with study facilities; E, documented changes to equipment and/or facility state(s); F, observations from infrared camera surveys at gathering facilities; G, brief description of tracer flux methodology; H, tracer flux measurements results; I, measurements of intermittent methane release events; J, downwind recovery of exhaust stack methane; K, comparison of facility-level emissions at processing plants; L, multiple linear least-squares regression variables and results; M, summary of high emitters; and N, cumulative distributions for C and C/D facilities. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

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

Present Address

[#]Department of Mechanical Engineering, Carnegie Mellon University, 5000 Forbes Avenue, Scaife Hall 402, Pittsburgh, Pennsylvania, 15213.

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Funding

Fiona and Stan Druckenmiller, Heising-Simons Foundation, Bill and Susan Oberndorf, Betsy and Sam Reeves, Robertson Foundation, Alfred P. Sloan Foundation, TomKat Charitable Trust, the Walton Family Foundation, Access Midstream, Anadarko Petroleum Corporation, Hess Corporation, Southwestern Energy, and Williams Corporation.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank the sponsors of this work for financial support, technical advice, and access to sites for sampling. The sponsors were Access Midstream, Anadarko Petroleum Corporation, Environmental Defense Fund (EDF), Hess Corporation, Southwestern Energy, and Williams Corporation. Funding for EDF's methane research series, including this work, is 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. DCP Midstream, while not a financial sponsor, provided site access for one of their processing plants selected at random. Subsequent to the field sampling campaign of this study, the Williams Companies purchased the controlling interest in Access Midstream Partners L.P. ("Access") by acquiring 100% of the general partnership interest of Access. A merger between Access and Williams Partners L.P. is planned to occur in the first quarter of 2015. We also thank the project science advisory panel for valuable feedback: David T. Allen, Garvin Heath, Michael Levi, and James McCarthy. 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. Thanks to Alex Davis (Carnegie Mellon University) for consultation on aspects of the data analysis.

■ ABBREVIATIONS

FLER	Facility-level emission rate (for the duration of a plume)
OP	Operational pressurized (compressor)
NOP	Nonoperational pressurized (compressor)
NOD	Nonoperation depressurized (compressor)
scfm	Standard cubic feet (of gas) per minute at 60 °F and 1 atm
WAFLER	Weighted-average facility-level emission rate (of methane)
tnWAFLER	Throughput-normalized weighted-average facility level emission rate (of methane)

■ REFERENCES

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

(2) Karion, A.; Sweeney, C.; Pétron, G.; Frost, G.; Michael Hardesty, R.; Kofler, J.; Miller, B. R.; Newberger, T.; Wolter, S.; Banta, R.; et al. Methane emissions estimate from airborne measurements over a western United States natural gas field. *Geophys. Res. Lett.* **2013**, *40* (16), 4393–4397.

(3) Pétron, G.; Frost, G.; Miller, B. R.; Hirsch, A. I.; Montzka, S. A.; Karion, A.; Trainer, M.; Sweeney, C.; Andrews, A. E.; Miller, L.; et al. Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study. *J. Geophys. Res.: Atmos.* **2012**, *117* (D4), D04304.

(4) Pétron, G.; Karion, A.; Sweeney, C.; Miller, B. R.; Montzka, S. A.; Frost, G. J.; Trainer, M.; Tans, P.; Andrews, A.; Kofler, J.; et al. A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin. *J. Geophys. Res.: Atmos.* **2014**, *119* (11), 2013JD021272.

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219–3227

(5) Harrison, M. R.; Shires, T. M.; Wessels, J. K.; Cowgill, R. M. *Methane Emissions from the Natural Gas Industry*; Gas Research Institute and Environmental Protection Agency: Washington, DC, 1996.
(6) Czepiel, P. M.; Shorter, J. H.; Mosher, B.; Allwine, E.; 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* (7), 593−598.
(7) Lamb, B.; Shorter, J. H.; McManus, J. B.; Kolb, C. E.; Mosher, B. W.; Harriss, R. C.; Allwine, E. J.; Blaha, D.; Siverson, R.; Howard, T.; 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.
(8) 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.
(9) Roscioli, J. R.; Yacovitch, T. I.; Floerchinger, C.; Mitchell, A. L.; Tkacik, D. S.; Subramanian, R.; Martinez, D. M.; Vaughn, T. L.; Williams, L.; Zimmerle, D.; et al. Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement methods. *Atmos. Meas. Tech.* **2014**, *7*, 12357−12406.
(10) Marchese, A. J.; Zimmerle, D.; Vaugh, T. L.; Martinez, D.; Williams, L. L.; Robinson, A. L.; Mitchell, A. L.; Subramanian, R.; Tkacik, D. S.; Roscioli, J. R.; Herndon, S. C. Methane emissions from United States natural gas gathering and processing. (near submission), **2015**.
(11) U.S. Energy Information Administration, Annual Energy Outlook, (Washington, DC, 2014).
(12) EPA. *Replace glycol dehydration units with methanol injection*; U.S. Environmental Protection Agency,: 2011; http://www.epa.gov/gasstar/documents/methanol_injection.pdf.
(13) Valerus Gas Dehydration. http://www.valerus.com/products-services/production-equipment/gas-dehydration (accessed August 1, 2014).
(14) U.S. Energy Information Administration. *Emissions of Greenhouse Gases in the United States, Appendix E*; U.S. Department of Energy: Washington, DC, 1997.
(15) Seinfeld, J. H.; Pandis, S. N. *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*; John Wiley & Sons Inc.: New York, 2006.
(16) Theresa, M. S.; Harrison, M. R. *Methane Emissions from Vented and Combustion Sources*; Radian International LLC: 1996.
(17) Kirchgessner, D. A.; Lott, R. A.; Michael Cowgill, R.; Harrison, M. R.; Shires, T. M. Estimate of methane emissions from the U.S. natural gas industry. *Chemosphere* **1997**, *35* (6), 1365−1390.
(18) Shorter, J. H.; McManus, J. B.; Kolb, C. E.; Lamb, B.; Siverson, R.; Allwine, E.; Westberg, H.; Mosher, B.; Harriss, R. *Results of Tracer Measurements of Methane Emissions From Natural Gas System Facilities*; Gas Research Institute: 1995.
(19) Picard, D. J.; M, S.; Ross, B. *Identification and Evaluation of Opportunities to Reduce Methane Losses at Four Gas Processing Plants*; Gas Technology Institute, U.S. Environmental Protection Agency: Des Plaines, IL, 2002; p 141.
(20) National Gas Machinery Laboratory. Clearstone Engineering Ltd.; Innovative Environmental Solutions, I. *Cost-Effective Directed Inspection and Maintenance Control Opportunities at Five Gas Processing Plants and Upstream Gathering Compressor Stations and Well Sites*; U.S. Environmental Protection Agency: 2006; p 74.
(21) U.S. Environmental Protection Agency. *Petroleum and Natural Gas Systems*. In *Final Rule: Subpart W of 40 CFR Part 98*, Washington, DC, 2012.
(22) Subramanian, R.; Williams, L.; Vaughn, T. L.; Zimmerle, D.; Roscioli, J. R.; Herndon, S. C.; Yacovitch, T. I.; Floerchinger, C.; Tkacik, D. S.; Mitchell, A. L.; et al. Methane emissions from natural gas compressor stations in the transmission and storage sector: Measurements and comparisons with the EPA greenhouse gas reporting program. *Environ. Sci. Technol.* **2015**, DOI: 10.1021/es5060258.
(23) 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.
(24) Pring, M. O.; Oommen, R.; Wilhelmi, J. *City of Fort Worth: Natural Gas Air Quality Study (Final Report)*; Easter Research Group, Inc.: Morrisville, NC, 2011; http://www.epa.gov/ttnchie1/conference/ei20/session6/mpring.pdf.
(25) Brandt, A. R.; Heath, G. A.; Kort, E. A.; O'Sullivan, F.; Pétron, G.; Jordaan, S. M.; Tans, P.; Wilcox, J.; Gopstein, A. M.; Arent, D.; et al. Methane leaks from North American natural gas systems. *Science* **2014**, *343* (6172), 733−735.
(26) Meyer, R. F.; Wallace, L. G.; Wagner, F. J. Geologic provinces code map. *AAPG Bull.* **1991**, *75*, (10).

DOI: 10.1021/es5052809
Environ. Sci. Technol. 2015, 49, 3219−3227