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Methane Emissions from United States Natural Gas Gathering and Processing

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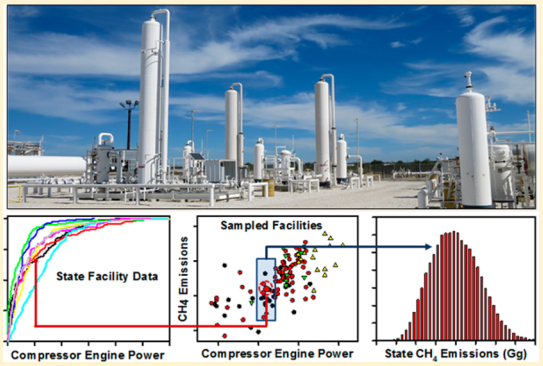
Supporting Information

ABSTRACT: New facility-level methane (CH_4) emissions measurements obtained from 114 natural gas gathering facilities and 16 processing plants in 13 U.S. states were combined with facility counts obtained from state and national databases in a Monte Carlo simulation to estimate CH_4 emissions from U.S. natural gas gathering and processing operations. Total annual CH_4 emissions of 2421 (+245/−237) Gg were estimated for all U.S. gathering and processing operations, which represents a CH_4 loss rate of 0.47% (±0.05%) when normalized by 2012 CH_4 production. Over 90% of those emissions were attributed to normal operation of gathering facilities (1697 +189/−185 Gg) and processing plants (506 +55/-52 Gg), with the balance attributed to gathering pipelines and processing plant routine maintenance and upsets. The median CH_4 emissions estimate for processing plants is a factor of 1.7 lower than the 2012 EPA Greenhouse Gas Inventory (GHGI) estimate, with the difference due largely to fewer reciprocating compressors, and a factor of 3.0 higher than that reported under the EPA Greenhouse Gas Reporting Program. Since gathering operations are currently embedded within the production segment of the EPA GHGI, direct comparison to our results is complicated. However, the study results suggest that CH_4 emissions from gathering are substantially higher than the current EPA GHGI estimate and are equivalent to 30% of the total net CH_4 emissions in the natural gas systems GHGI. Because CH_4 emissions from most gathering facilities are not reported under the current rule and not all source categories are reported for processing plants, the total CH_4 emissions from gathering and processing reported under the EPA GHGRP (180 Gg) represents only 14% of that tabulated in the EPA GHGI and 7% of that predicted from this study.



INTRODUCTION

Since 2005, domestic production of natural gas (NG) in the United States has increased by 26%.[1] Much of this increase is a consequence of major new resources that have become accessible for commercial production due to advances in hydraulic fracturing and horizontal drilling. During this same period, the U.S. has seen an increased demand for NG in the stationary electricity generation and transportation sectors.[2,3] Since NG has lower carbon per unit of energy than other fossil fuels, the potential exists for decreased greenhouse gas emissions when NG is substituted for coal or liquid fossil fuels because less carbon dioxide (CO_2) is emitted when NG is combusted in comparison to other fossil fuels. However, since NG is composed primarily of methane (CH_4)—a much more potent greenhouse gas than CO_2[4]—a certain amount of NG emitted throughout the entire supply chain can reduce the greenhouse gas benefits of lower CO_2 emissions. For example, recent studies suggest that substitution of new coal fired power plants with new NG plants would result in short-term climate benefits only if the total net CH_4 emission rates were less than 3 to 4%,[5,6] while substitution of heavy-duty diesel vehicles with compressed NG would require a net CH_4 emission rate of less than roughly 1% from well-to-wheels to ensure net short-term climate benefits.[5,7,8]

The NG supply chain consists of a vast network of operations (exploration, production, gathering, processing, transmission, and distribution) with a variety of CH_4 emission sources including: fugitive emissions from leaking valves, fittings, and compressors; venting from normal operations

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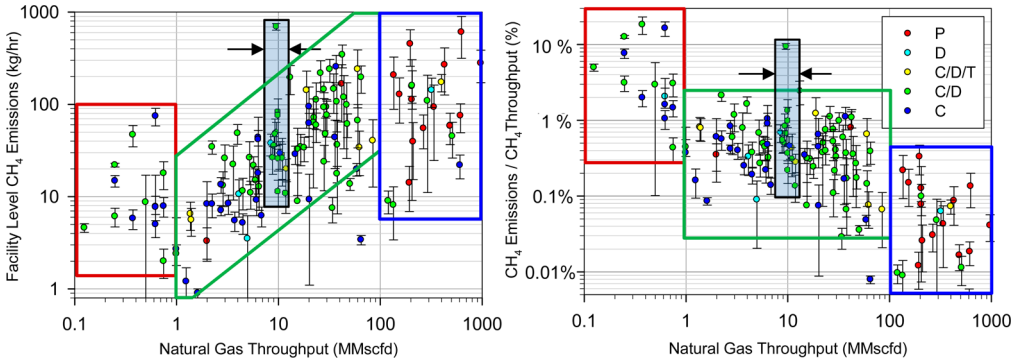

Figure 1. (a) Measured facility-level emission rates (FLER) of CH_4 (kg/h) and (b) throughput-normalized facility-level emission rates (tnFLER) of CH_4 (%) for 114 gathering facilities and 16 processing plants as a function of natural gas throughput (MMscfd) for each facility.[16] The blue shaded regions demonstrate the population of facilities from which CH_4 FLER would be drawn (and the associated tnFLER for the same population of facilities) for a 10 MMscfd facility in the Monte Carlo simulation. The red, green, and blue boxes highlight the differences in emissions for small (0.1 to 1 MMscfd), midsize (1 to 100 MMscfd), and large (>100 MMscfd) facilities.

such as NG powered pneumatic devices; venting from periodic maintenance and upsets; and combustion emissions (uncombusted CH_4 released through the exhaust of devices fueled by NG). The Environmental Protection Agency (EPA) 2014 Greenhouse Gas Inventory (GHGI) estimates total net 2012 CH_4 emissions of 6,186 Gg/yr for the U.S. NG supply chain.[9] With the addition of an estimated 408 Gg/yr of CH_4 emissions from the production of associated gas in 2012, the EPA GHGI estimate represents a total rate of CH_4 loss of 1.3% from the U.S. NG supply chain (Supporting Information, SI, Section S.1.1). However, the EPA GHGI estimate remains uncertain,[5,10,11] as it is based largely on activity data and emission factors that were developed in the early 1990s.[12] Moreover, many studies suggest that CH_4 emission rates from the NG supply chain are dominated by a relatively small fraction of highly emitting components, and it is therefore inaccurate to assume a normal distribution for all emission sources.[11]

To improve our understanding of CH_4 emissions from the U.S. NG supply chain, a series of comprehensive studies have been organized in recent years focusing on CH_4 emissions from specific sectors of the NG industry: exploration and production;[10,13,14] gathering and processing,[15,16] (this study); transmission and storage;[17,18] distribution[19] and NG vehicles. In this study, recent facility-level CH_4 emissions measurements[15,16] obtained from 130 facilities (owned by five companies and located in 13 U.S. states) were combined with facility counts obtained from state and national databases in a Monte Carlo simulation to produce an estimate of total annual CH_4 emissions from all U.S. NG gathering and processing operations.

The gathering and processing sectors are defined as all NG industry assets and operations that take place between two custody transfer points: the well site delivery meter and the receipt meter to the transmission sector (or local distribution) (SI Section S.1.2). Gathering and processing systems include gathering pipelines, gathering facilities, and processing plants. Gathering facilities collect NG from multiple wells (typically 10 to 100), compress the gas to a higher pressure and discharge the gas toward its next destination, which could be another gathering facility, a transmission line (if no further processing of the gas is needed) or a processing plant. At gathering facilities, compressors are powered by reciprocating engines and/or gas turbines that operate on NG. In some cases, compressors are electrically powered. Gathering facilities also often include inlet separators (to remove liquid H_2O and/or hydrocarbon condensate), dehydration systems (to remove gaseous H_2O) and amine treatment systems (to remove CO_2 and/or hydrogen sulfide [H_2S]). Produced water and/or liquid hydrocarbon condensate are often stored in tanks onsite. Processing plants typically include all of the above operations/equipment but also include systems to remove ethane and/or natural gas liquids (NGLs).

Prior to the recent field campaign,[15,16] relatively little data had been published on CH_4 emissions from processing plants and even less for gathering facilities.[20−22] Shorter et al.[20] reported facility-level CH_4 emissions from seven processing plants and two gathering facilities, and two additional studies[21,22] reported CH_4 emissions from nine processing plants. While processing is reported as a distinct sector within the EPA GHGI, gathering is currently embedded within the EPA GHGI production inventory and therefore a national inventory of CH_4 emissions from gathering facilities does not exist. Moreover, unlike processing plants, for which national facility counts are available,[23,24] there are no national databases available on gathering facilities in the U.S. Accordingly, the objectives of this study were as follows: (1) compile facility data (count and NG throughput) for U.S. processing plants; (2) compile facility data (state counts and installed compressor engine power) for gathering facilities; (3) combine the facility data with new CH_4 emissions data[15,16] in a Monte Carlo simulation to estimate total annual CH_4 emissions from all U.S. gathering facilities and processing plants; and (4) compare the model results against the EPA GHGI and EPA Greenhouse Gas Reporting Program (GHGRP).[25]

METHODS

Field Campaign. As detailed in Mitchell et al.,[16] a 20-week field campaign was conducted from October 2013 through April 2014 during which total facility-level emission rates (FLER) of CH_4 were measured from 114 NG gathering facilities and 16 processing plants located in 13 U.S. states. The CH_4 FLER were measured using a dual gas downwind tracer flux technique,[15] which attempts to capture all CH_4 emissions from a facility (fugitive, vented, and uncombusted). The 114 gathering facilities were selected by the study team from randomly ordered lists of 738 such facilities—subject to constraints such as road access and wind direction—provided by four industry study partners (SI Section S.2). The 16

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sampled processing plants represented all accessible processing plants operated by two industry study partners and one randomly selected plant provided by another company who provided site access but did not provide funding for the study. Emissions from gathering pipelines were not measured. The dual tracer gas methodology and data analysis techniques employed in the field campaign are described in detail by Roscioli et al.[15] A detailed description of sampling strategy, facility selection process, analysis of CH_4 FLER data, and description of the effects of facility type and operating conditions on measured CH_4 FLER are presented by Mitchell et al.[16] and summarized in SI Section S.2.

Gathering facilities were defined as compression only (C); dehydration only (D); compression/dehydration (C/D); compression/dehydration/treatment (C/D/T); or dehydration/treatment (D/T). Processing plants (P) were defined in accordance with Title 40, Code of Federal Regulations, Part 60, Subpart KKK ("Subpart KKK processing plants"). Facilities exclusively dedicated to fractionation were omitted from the study since the process of fractionation of NGLs was considered to be outside the natural gas supply chain. Several of the processing plants sampled in the study did, however, have fractionation systems on site and any CH_4 emissions from these operations would have been captured as part of the CH_4 FLER.

Figure 1(a) is a plot of the CH_4 FLER from all sampled gathering facilities and processing plants as a function of NG throughput in MMscfd (1 MMscfd = 0.328 m^3/s). The CH_4 FLER data in Figure 1(a) represent weighted average results from multiple CH_4 plumes (2 to 42 plumes) acquired from each facility.[15,16] Figure 1(b) is a plot of throughput-normalized facility-level emission rates (tnFLER) for the same facilities, where tnFLER is defined as the measured CH_4 FLER divided by the facility CH_4 throughput, which was calculated from the total NG throughput and inlet CH_4 content (vol %), respectively. The data are tabulated in SI Section S.3 and are the focus of a recent paper by Mitchell et al.[16] The tnFLER represent the percentage of CH_4 handled by a given facility that is emitted on a particular day of sampling. Measured tnFLER were as low as 0.01% for some gathering facilities, but greater than 1% for 25 facilities and greater than 10% for four facilities. Five facilities had measurable CH_4 FLER with zero NG throughput. The median tnFLER was much lower for processing plants (0.08%) in comparison to gathering facilities (0.42%) and none of the 16 processing plants sampled had tnFLER greater than 0.80%. A discussion on the field campaign sample size and capture of "super-emitters" is included in SI Section S.3.1.

While the tnFLER is a useful metric to assess the rate of CH_4 loss on a facility-by-facility basis, a more effective metric to characterize emissions from a population of facilities is the sampled CH_4 loss rate (SMLR), defined as $\Sigma(CH_4\ FLER)/\Sigma(CH_4\ throughput)$ for the population of facilities. For gathering facilities and processing plants sampled in this study, the SMLR were 0.20% ± 0.01% and 0.078% ± 0.01%, respectively. However, these SMLR values cannot be extrapolated to the national population of gathering and processing facilities because a fraction of NG flows through multiple gathering facilities and the CH_4 emissions (both FLER and tnFLER) were found to be dependent on facility NG throughput.

Monte Carlo Model. The colored boxes in Figure 1 highlight the result that low throughput facilities (<1 MMscfd) exhibited lower FLER (2 to 75 kg/h) and higher tnFLER (1% to >10%) than high throughput facilities (>100 MMscfd), which had higher FLER (8 to 606 kg/h) but lower tnFLER (<0.8%). The midsize facilities (1 to 100 MMscfd), which represent the largest number of facilities, exhibited a wide spread in emissions. While the CH_4 FLER was found to vary with facility throughput (R^2 = 0.38), the wide spread in emissions observed for a given throughput range suggests that CH_4 emissions from some sources are independent of gas throughput. Indeed, CH_4 emissions are a function of facility type, facility design (e.g., number of pneumatic devices, prime mover types), facility size/complexity (e.g., number of fugitive leaks), facility operating pressures (e.g., magnitude of fugitive leaks), facility pressure ratio (e.g., number of stages of compression), total facility throughput (e.g., number of compressors; combustion emissions), and operating state (e.g., normal, maintenance, etc.). Mitchell et al.[16] includes a detailed discussion on how the CH_4 FLER was found to vary with facility type, equipment and facility operations. The modeling approach employed in this study accounts for both the throughput dependence and variability in CH_4 emissions for a given throughput range. To account for the variation in observed CH_4 FLER for a given throughput range, a Monte Carlo model was developed whereby CH_4 FLER values were randomly drawn from the measurement data set and assigned to "similar" facilities as compiled in partner, state and national inventories (SI Section S.4). The CH_4 FLER values were randomly drawn based on facility type and average daily NG throughput (for all processing plants and partner gathering facilities) or installed compressor engine power (for nonpartner gathering facilities). The experimental uncertainty for each sampled facility was accounted for by drawing a CH_4 FLER value from a normal distribution centered on the weighted average FLER given the unbiased weighted standard deviation for each measurement.

The modeling framework integrates multiple data sources: measured CH_4 FLER data, operating data and equipment inventory from each sampled facility (N = 130); operating data and equipment inventory from all study partner facilities (N = 766); gathering facility counts from eight U.S. states and national processing plant counts from the Energy Information Administration (EIA) and the Oil and Gas Journal[23,24] (SI Section S.4). State-level counts of gathering facilities were developed by analyzing publically available air permit data obtained from Arkansas, Colorado, Louisiana, New Mexico, Oklahoma, Pennsylvania, Texas, and Wyoming. These states accounted for 83.5% of all U.S. NG gathered (and 82.2% of marketed production) in 2012.[1] The process by which facilities in the air permit data sets were identified as gathering facilities and the quantification of confidence intervals for this process is detailed in SI Section S.4. The confidence intervals for the number of gathering facilities in each state were used to randomly vary the state facility counts for each iteration in the simulation. Table S13 (SI Section S.9) contains a list of the state facility data sets used in the simulation. For seven of the eight states, the gathering facility counts identified in each state data set were adjusted based on the fraction of known partner gathering facilities found in each data set (SI Section S.4.3). For Texas, the total number of gathering facilities was estimated by extrapolating the gathering facility count identified in a recent study[26] on the Barnett Shale (N = 259) based on the fraction of Texas NG produced in the Barnett Shale over the period 2009 through 2014 (SI Section S.4.3). For processing plants, national

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facility data were available[23,24] resulting in the identification of 578 unique Subpart KKK processing plants with NG throughput available for 512 of these plants (SI Section S.4.1).

The Monte Carlo simulation consists of three submodels (Figure S10, SI Section S.4.5): a partner gathering facility submodel, a nonpartner gathering facility submodel and a processing plant submodel. All three submodels employ a "nearest neighbors"[27] Monte Carlo scheme to account for the throughput dependence and variation in measured CH_4 FLER for a given throughput range. In the gathering facility submodels, facilities are drawn from each state database and identified as a partner facility or nonpartner facility. If identified as a partner facility, then the gathering facility type is identified along with its 2012/2013 average daily NG throughput (as provided by the partner companies). A measured CH_4 FLER value is then randomly drawn from one of the 10 sampled facilities of that type that are closest in NG throughput (i.e., the "nearest neighbors"). The blue shaded region in Figure 1(a) indicates the region of population from which CH_4 FLER would be drawn for a 10 MMscfd throughput facility. The nonpartner gathering facility submodel is used when the facility is not identified as a partner facility. In this case, the 10 "nearest neighbors" are identified based on installed compressor engine horsepower and no distinction is made between gathering facility types. The observed variation in CH_4 FLER with facility throughput is taken into account through the use of installed compressor engine power, which was found to correlate well with throughput (R^2 = 0.71) for sampled partner facilities (Figure S8, SI Section S.4.2).

For the processing plant submodel, a total Subpart KKK processing plant count of 592 ± 14 was used, which accounted for the discrepancy between the 606 processing plants as compiled in the EPA GHGI[9,23] and the 578 plants identified in this study as Subpart KKK plants (SI Section S.4.1). For each iteration of the Monte Carlo simulation, the processing plant count is randomly varied (between 578 and 606) and that count is populated using the NG throughput distribution from the national data set of 512 Subpart KKK processing plants for which NG throughput was found.[23,24] For each plant in the national list, a measured CH_4 FLER value is randomly drawn from one of the seven sampled plants and C/D/T facilities that are closest in NG throughput. The CH_4 FLER measured at C/D/T gathering facilities were integrated into the processing plant submodel because the throughput distribution of sampled processing plants had a much lower percentage of low throughput plants in comparison to the national distribution. The use of C/D/T facilities as a proxy for low throughput Subpart KKK plants is discussed in detail in SI Section S.4.1. A discussion on use of seven nearest neighbors in the processing plant simulation is included in SI Section S.5.

RESULTS

The Monte Carlo simulation results for gathering facilities in the eight states are shown in Figure 2. The figure contains cumulative distribution functions (CDFs) of the predicted total CH_4 emissions (Gg) from each state from 50 000 iterations of the simulation. The slope of each CDF curve is indicative of the confidence intervals in facility count, which were estimated as described in SI Section S.4.4. For example, the model predictions for Colorado suggest that the total annual emission of CH_4 from gathering facilities in Colorado is 69 Gg (+25/−21 Gg). The former value represents the median value at 50% probability from the model predictions, while the latter values

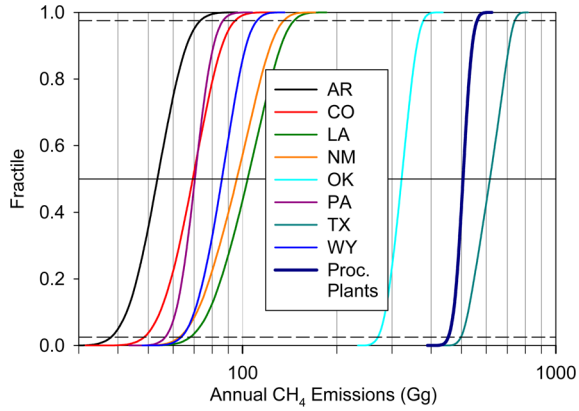

Figure 2. Cumulative distribution functions (CDFs) of predicted annual emission of methane (Gg) from gathering facilities in the states of AR, CO, LA, NM, OK, PA, TX, and WY and U.S. Subpart KKK processing plants (N = 592 ± 14) from the Monte Carlo simulation. The median values and confidence intervals are indicated by the intersection between the CDFs and the solid (50%) and dashed lines (2.5%, 97.5%), respectively.

represent the confidence intervals in the model results. The confidence intervals are defined as the difference between the median prediction and the prediction at 2.5% and 97.5% probability, respectively. The magnitude of the confidence intervals is influenced primarily by the uncertainty in the facility counts and, to a lesser extent, by the experimental uncertainty in each individual CH_4 FLER measurement. Incorporating the experimental uncertainty in the model resulted in widening of the state level confidence intervals by 5 to 15%. A similar process was performed for all eight states in the model and the predicted CH_4 emissions for each state ranged from a low of 53 Gg (+20/−15 Gg) for Arkansas to a high of 616 Gg (+124/−118 Gg) for Texas. The results are summarized in Table 1.

The predicted annual total emission of CH_4 from gathering facilities in the eight states explicitly included in the model is 1417 (+158/−154) Gg. Normalizing this value by the total CH_4 gathered in 2012 from these eight states (351 Tg) yields a modeled CH_4 loss rate (MMLR) from gathering facilities of 0.40% (+0.05%/-0.04%), which is substantially higher than the SMLR (0.20% ± 0.01%) from the 114 gathering facilities sampled in this study. The higher model predictions are indicative of the fact that a percentage of NG is handled by more than one gathering facility as it travels from the well to the transmission (or distribution) sector. It should also be noted that the SMLR for gathering facilities was heavily influenced by the presence of five very high throughput/low tnFLER facilities; indeed, the remaining 109 gathering facilities had an SMLR of 0.41%. The total amount of CH_4 gathered in the eight states included in the model represents 83.5% of the 421 Tg of CH_4 gathered in the U.S. 2012. Assuming that the CH_4 emissions from facilities that gather the remaining 16.5% of CH_4 occur at the same CH_4 loss rate (0.40%), a total of 1697 (+189/−185) Gg of CH_4 emissions is predicted for all U.S. gathering facilities.

Table 1 also includes the total number of gathering facilities as derived from the eight state data sets (3797 +768/−587) and an extrapolated estimate of total U.S. gathering facilities (4549 +921/−703), which was developed by assuming that the total number of gathering facilities scales linearly with CH_4 gathered (SI Section S.4.3 and S.4.4). Since accurate gathering facility counts were not available prior to this study, a comparison was

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Table 1. Total Annual Emission of CH_4 (Gg) and Modeled CH_4 Loss Rate (MMLR %) from Gathering Facilities in 8 U.S. States, which Account for 83.5% of U.S. Natural Gas Gathered

state	number of gathering facilities	annual emission of CH_4 from gathering facilities (Gg)	2012 CH_4 gathered[b] (Gg)	modeled CH_4 loss rate (%)
AR	214 (+43/−38)	53 (+20/−25)	19 723	0.27% (+0.10%/−0.08%)
CO	193 (+46/−35)	69 (+25/−21)	28 261	0.25% (+0.09%/−0.07%)
LA	364 (+62/−124)	104 (+42/−36)	50 207	0.21% (+0.08%/−0.07%)
NM	282 (+82/−82)	96 (+40/−33)	20 215	0.47% (+0.20%/−0.16%)
OK	1103 (+132/−132)	322 (+56/−52)	34 263	0.94% (+0.16%/−0.15%)
PA	247 (+22/−7)	70 (+16/−14)	37 676	0.19% (+0.04%/−0.04%)
TX	1012 (+304/−101)	616 (+124/−118)	126 552	0.49% (+0.10%/−0.09%)
WY	382 (+77/−66)	86 (+25/−22)	34 414	0.25% (+0.07%/−0.06%)
total states in model	3797 (+768/−587)	1417 (+158/−154)	351 310	0.40% (+0.05%/−0.04%)
total U.S.[a]	4549 (+921/−703)	1697 (+189/−185)	420 906	0.40% (+0.05%/−0.04%)

[a]Total number of gathering facilities and total predicted CH_4 emissions scaled linearly by total natural gas gathered in the U.S. in 2012. [b]Natural gas gathered for each state was calculated by subtracting lease fuel from the total marketed production.[1] CH_4 gathered was calculated from natural gas gathered assuming an average CH_4 content of 90 vol %.

made between the facility counts derived from the state air permit databases to the known partner facility count. The partner companies reported handling a total of 5.9 Tscf of NG in 2012, which represents 103 Tg of CH_4 (assuming an average of 90 vol % CH_4) or 24% of the total 421 Tg of CH_4 gathered during the same period. The 738 partner facilities would therefore extrapolate to a total of 3022 gathering facilities nationally. However, the total amount of CH_4 handled by the partner facilities is calculated from the sum of the annual CH_4 throughput from each facility and is not directly equivalent to the "CH_4 gathered" by the partner companies since NG can be handled by multiple gathering facilities in transit through the gathering sector.

Since the gathering facility submodel draws CH_4 FLER data from the same set of 114 sampled facilities for all states in the model, the variation in predicted MMLR among the eight states is driven primarily by the ratio of gathering facility count to CH_4 gathered for each state. Oklahoma, with a ratio of 32.2 facilities per Tg CH_4 gathered, exhibited the highest MMLR (0.94%). Conversely, states such as LA, CO, and PA all have less than 7.3 facilities per Tg CH_4 gathered and MMLRs less than 0.25%. Figure S12 (SI Section S.5), which is a plot of MMLR as a function of the ratio of facility count to CH_4 gathered (Tg^{-1}) for each state in the model, shows that the MMLR varies linearly ($R^2 = 0.84$) with this ratio and that the 0.40% national estimated MMLR falls along this same line.

The Monte Carlo simulation results for all U.S. Subpart KKK processing plants are also shown in Figure 2. The results suggest that the total annual emission of CH_4 from Subpart KKK processing plants is 506 (+55/−52) Gg for 2012. Normalizing this value by the total CH_4 processed in 2012 (287 Tg, SI Section S.8) yields a modeled CH_4 loss rate (MMLR) from processing plants of 0.18% (±0.02%).

Potential Sources of Bias in Model Predictions. Several sources of uncertainty exist that potentially bias the CH_4 emissions results as predicted by the Monte Carlo simulation. The potential sources of bias include: multiple episodic CH_4 emission events captured during maintenance/blowdowns at gathering facilities that were not included in the CH_4 FLER data,[16] incomplete capture of uncombusted CH_4 entrained in lofted exhaust plumes[16] and uncertainty in electric compressor station facility counts. As discussed in detail in SI Section S.5.1, the bias from the missing episodic CH_4 emissions events and incomplete capture of CH_4 entrained in lofted plumes were quantified and the results suggest that the model predictions are biased slightly low. To estimate the impact of the missing episodic emission events (10 plumes out of 1442 plumes acquired during the field campaign) on the gathering facility model predictions, a separate Monte Carlo model was constructed that showed that these events yield an additional 169 (+426%/−96%) Gg/yr of CH_4 emissions, which is less than 10% of the total predicted CH_4 emissions from gathering facilities. In terms of incomplete capture of lofted emissions, Mitchell et al.[16] estimated that the CH_4 FLER data were biased low by a maximum of 15% for processing plants and 17% for gathering facilities, respectively. It is unclear how the uncertainty in electric compressor station facility counts biases the model predictions. In summary, the effects of the missed episodic emissions, incomplete capture of lofted plumes, and electric compressors were not incorporated into the model, and it is therefore likely that the model predictions are overall biased slightly low.

DISCUSSION

The total estimated annual CH_4 emissions from U.S. gathering facilities and processing plants as predicted by the Monte Carlo simulation were compared to the 2012 CH_4 emissions as reported in the 2014 EPA GHGI (SI Section S.6) and reported under the EPA GHGRP (SI Section S.7).

Comparison to EPA GHGI. *Processing.* Since the EPA GHGI explicitly includes NG processing as a separate sector in the U.S. NG supply chain, a reasonably direct comparison can be made with our results (506 +55/−52 Gg). In the 2014 EPA GHGI, the 2012 total net U.S. CH_4 emissions from NG processing are estimated to be 892 Gg, which includes 851 Gg of emissions during normal operations and 40 Gg of emissions from routine maintenance (see SI Section S.6.1).

The results of this study suggest that CH_4 emissions from processing plants during normal operations are lower (by a median factor of 1.7) than the EPA GHGI estimate. The discrepancy can be largely attributed to differences in compressor activity data in the GHGI in comparison to that observed during the field campaign. The EPA GHGI lists 5624 reciprocating and 906 centrifugal compressors among 606 plants, which yields a value of 9.3 reciprocating compressors per plant and a ratio of 6.2 reciprocating to centrifugal compressors (SI Section S.6.1). During the field campaign, for the 16 sampled plants we observed fewer reciprocating compressors

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per plant (6.4) and a smaller ratio of reciprocating to centrifugal compressors (2.6). Including field observations for the C/D/T facilities—whose CH_4 FLER values were part of the Monte Carlo processing plant simulation—we also observed fewer reciprocating engines per plant (5.4) and a smaller ratio of reciprocating to centrifugal compressors (3.3) (SI Section S.6.1, Table S6). Mitchell et al.[16] found that CH_4 FLER at facilities with centrifugal compressors driven by gas turbines was 75% lower than at facilities with engine-driven reciprocating compressors. Similar observations were found in recent studies of CH_4 emissions from the U.S. transmission and storage sector.[17,18] Since 90% of all CH_4 emissions in the EPA GHGI processing inventory are from compressor fugitives and engine/turbine exhaust emissions (SI Section S.6.1, Table S5), it is likely that the GHGI's assumed activity factors for reciprocating and centrifugal compressors lead to an overestimate of CH_4 emissions from processing plants.

Comparison to EPA GHGI. *Gathering.* For gathering facilities, a direct comparison with the EPA GHGI is complicated because gathering systems (gathering pipelines, facilities, and associated equipment) are embedded within the GHGI production inventory. Some of the source categories in the EPA GHGI production inventory (e.g., gathering pipelines) can be assigned solely to gathering and in these cases it is reasonable to assign 100% of the activity and emissions data for those categories to the gathering sector. Other categories, such as gathering compressors, engine exhaust, pneumatic devices, dehydration units, chemical pumps, and condensate tanks are common to both production sites and gathering facilities[10,13,14] and assumptions must be made to assign a percentage of the activity data and emissions to each sector. Table S7 (SI Section S.6.2) is a compilation of 2012 CH_4 emission source categories from the 2014 EPA GHGI production inventory that are present at both production sites and gathering facilities. Sources common to both production and gathering systems (which would constitute an absolute upper bound on gathering emissions in the GHGI) account for 1,431 Gg of 2012 CH_4 emissions in the 2014 EPA GHGI, which is less than the model prediction of 1697 (+189/−185) Gg for gathering facilities alone.

We apportioned the fraction of activity data for each source category in SI Table S7 that could be assigned to the gathering sector using equipment data from the 738 partner gathering facilities, detailed equipment surveys of the 114 sampled gathering facilities (which included pneumatic device counts, inlet separator counts, condensate/water tank counts, dehydrator counts, installed/operating compressor engine power, etc.) and the estimated total count of 4549 U.S. gathering facilities (SI Section S.6.2). Given these assumptions, a total of 404 Gg of CH_4 emissions were assigned to gathering, which represents 20% of the total net 2012 CH_4 emissions (1992 Gg) in the 2014 EPA GHGI production inventory.[9] For the gathering facilities (i.e., subtracting out the GHGI estimated 178 Gg of gathering pipeline emissions), a total CH_4 emission of 226 Gg is estimated (Table S8, SI Section S.6.2) in comparison to our model results of 1697 Gg for gathering facilities. While there is uncertainty in determining gathering facility emissions from the EPA GHGI, the results of this study suggest that the GHGI substantially underestimates emissions from gathering facilities.

Comparison to EPA GHGRP. The EPA GHGRP requires annual reporting of GHG emissions from large direct emission sources and suppliers of certain fossil fuels and industrial gases in the United States. Processing plants are required to report CH_4 emissions from multiple source types under 40 CFR Part 98 Subparts W and C if their total greenhouse gas emissions from these sources are in excess of 25 000 t per year of CO_{2e}. Gathering facilities are required to report CH_4 emissions only from stationary combustion sources (under 40 CFR Part 98 Subpart C) and only if their total greenhouse gas emissions from these sources are in excess of 25 000 t per year of CO_{2e}.[28] A detailed discussion of GHGRP reporting requirements for processing plants and gathering facilities is included in SI Section S.7 and elsewhere.[28]

Comparison to EPA GHGRP. *Gathering.* In 2013, 404 gathering facilities reported a total of 0.53 Gg of CH_4 emissions under the GHGRP Subpart C, which represents a very small fraction (0.03%) of the total national CH_4 emissions from gathering facilities predicted in this study. This large difference is mainly because the total number of reporting gathering facilities represents less than 10% of the national facility count and only combustion emissions from those facilities are reported. Also, a recent study on CH_4 emissions from the NG transmission and storage sector suggests that the emission factor used in the GHGRP to estimate CH_4 combustion emissions for reciprocating engines is unrealistically low.[17,18] The EPA has proposed a revision to the GHGRP rule that would require the gathering sector to begin reporting CH_4 emissions from numerous additional sources under Subpart W (SI Section S.7.1).[29]

Comparison to EPA GHGRP. *Processing.* The estimate of processing plant emissions from this study is also larger than that reported under the GHGRP. Figure 3 is a comparison of measured annualized CH_4 FLER (Mg) and the 3-year average (2011 to 2013) of CH_4 emissions reported under the GHGRP for the 16 processing plants sampled in this study. The total emission of CH_4 based on the FLER measurements from the 16 sampled processing plants when extrapolated to an annual value represents a total of 23.7 ± 4.0 Gg. The total annualized CH_4 emissions reported under the GHGRP for those same

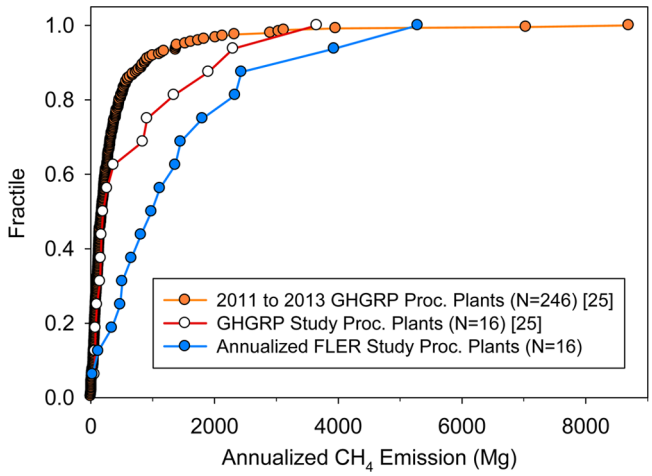

Figure 3. Comparison of cumulative distribution functions for the measured annual equivalent methane FLER (Mg, CH_4) for the processing plants sampled in this study (N = 16; blue circles), average annual emissions reported under the 2011 to 2013 EPA Greenhouse Gas Reporting Program[25] for the same processing plants (N = 16; white circles) and all processing plants that reported emissions under the GHGRP from 2011 through 2013 for which average daily natural gas throughput was available (N = 246).[25]

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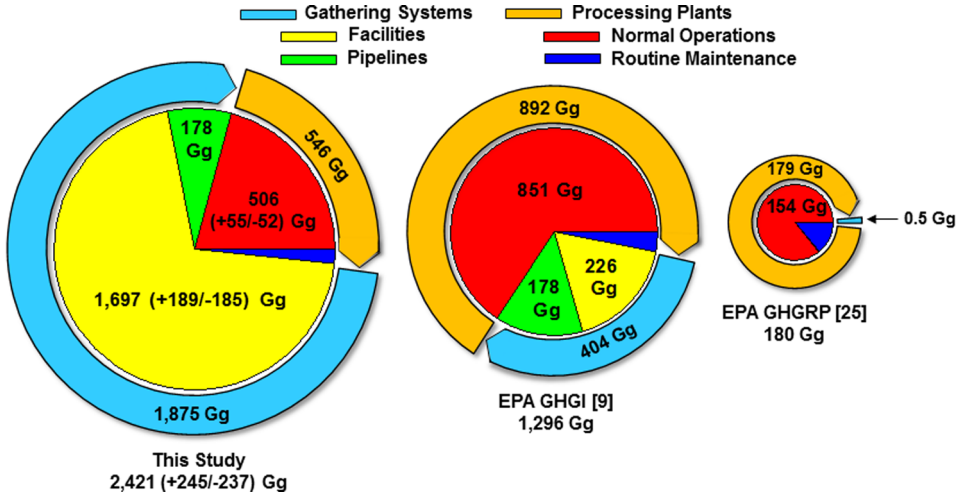

Figure 4. National estimate of total CH_4 emissions (Gg) from U.S. natural gas gathering systems (facilities and pipelines) and processing plants (normal operation and routine maintenance) from this study in comparison to that derived from the 2014 EPA GHGI[9] and reported under the 2013 EPA GHGRP,[9] respectively. CH_4 emissions from processing plant routine maintenance (blue wedges) are 40 Gg for the study results and EPA GHGI and 25 Gg for the EPA GHGRP.

plants was 12.6 Gg, which is 47% lower than the measurements. Normalizing by total CH_4 throughput for these 16 plants, the GHGRP-reported CH_4 loss rate from the 16 plants is 0.043% in comparison to the SMLR of 0.078% for these same plants.

A comparison was also made between the results of the Monte Carlo simulation for all U.S. plants and the total CH_4 emissions from all processing plants that reported to the GHGRP from 2011 to 2013 for which annual NG throughput was obtained (N = 246). The reported annualized average CH_4 emissions from these facilities are also plotted in Figure 3. The total reported annual CH_4 emissions from these 246 plants (computed as the average of 2011 through 2013) was 104 Gg. The total annualized NG processed by these plants was 14.5 Tscf. Assuming an average inlet CH_4 content of 85 vol % for these processing plants yields a total reported rate of CH_4 loss of 0.044%, which is less than 1/4 of our 0.18% MMLR. As shown in Figure 3, approximately 50% of the reported CH_4 emissions in both the larger GHGRP (N = 246) and the study plant data set (N = 16) were less than 200 Mg, whereas only two of 16 annualized CH_4 FLER measurements were below this value. One reason for the discrepancy between measured and reported GHGRP CH_4 emissions is that not all sources of CH_4 emissions at processing plants are required to be reported under the GHGRP. For example, CH_4 emissions from pneumatic controllers, pneumatic pumps, tanks and compressor fugitives during some operating modes are not reported, and for acid gas removal vents, only CO_2 emissions (and not CH_4) are reported (SI Section S.7). Also, as mentioned above, a recent study suggests that the emission factor used in the GHGRP to estimate CH_4 combustion emissions for reciprocating engines is unrealistically low.[17,18]

Total CH_4 Emissions and Loss Rate from U.S. Gathering and Processing. In this section the Monte Carlo simulation results are used to estimate the total annual CH_4 emissions (Gg) and rate of CH_4 loss (%) from all U.S. NG gathering and processing operations (SI Section S.8). Figure 4 summarizes the modeled estimate of total annual CH_4 emissions from U.S. gathering facilities (1697 +189/−185 Gg) and processing plants under normal operation (506 +55/−52 Gg) along with comparisons against the EPA GHGI and 2013 GHGRP. Methane emissions from gathering pipelines and processing plants under routine maintenance (i.e., blowdowns, which entail the venting of high pressure vessels and pipe sections) were not measured in this study and, for the purposes of a national estimate, the estimated 2012 CH_4 emissions in the EPA GHGI were assumed for these source categories (178 Gg and 40 Gg, respectively). On the basis of the results of this study, the total annual CH_4 emissions from U.S. natural gathering and processing are estimated at 2421 (+245/−237) Gg in comparison to 1,296 Gg of CH_4 emissions derived from the EPA GHGI and 180 Gg of CH_4 emissions reported under the 2013 EPA GHGRP, respectively. The 2013 GHGRP value includes 154 Gg of CH_4 emissions from 433 NG processing plants under normal operating conditions, 25 Gg from these same plants during routine maintenance and only 0.53 Gg from the 404 gathering facilities that reported in 2013.

While CH_4 emissions from processing plants appear to be substantially lower than that tabulated in the EPA GHGI, CH_4 emissions from gathering facilities appear to be substantially higher than current EPA GHGI estimates and it can be concluded from this study that gathering systems are a substantial contributor to total CH_4 emissions from the U.S. NG supply chain. Indeed, the total CH_4 emissions from gathering systems (facilities and pipelines) as predicted in this study (1875 +189/−185 Gg) are greater than that estimated for the transmission and storage sector in a recent study (1503 Gg +30%/−19%)[18] and are equivalent to 30% of the total net 2012 CH_4 emissions for the entire NG supply chain in the 2014 EPA GHGI (6186 Gg). Moreover, the predicted CH_4 emissions are most likely biased low as discussed above and in SI Section S.5.1.

Figure 4 also shows that the total CH_4 emissions from gathering and processing reported under the EPA GHGRP (180 Gg) represents only 14% of that tabulated in the EPA GHGI and 7% of that predicted from this study. As noted above and in the SI (Section S.7), the large discrepancies are due to reporting requirements (e.g., reporting threshold of 25 000 t CO_{2e} per facility) that capture very little CH_4 emissions from gathering (0.53 Gg total) as well as multiple source categories that are not reported for processing plants under the GHGRP.

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The results of the Monte Carlo simulation were used to estimate the MMLR (%) from all U.S. NG gathering and processing operations, and the individual subcategories of gathering facilities, gathering pipelines and processing plants (routine operations and blowdowns). The subsector MMLR for gathering facilities, gathering pipelines, processing plant normal operations, and processing plant blowdowns (retaining the EPA GHGI estimate for gathering lines and processing plant blowdowns) were estimated to be 0.40%, 0.042%, 0.18%, and 0.014%, respectively (SI Section S.8). These values represent the percentage of CH_4 lost during handling by each of those subcategories but cannot be added together to develop an estimate for the total rate of CH_4 loss from gathering and processing. Normalizing the CH_4 emissions from each subcategory by the total net CH_4 production of 513 Tg for 2012 (SI Section S.8), yields a supply chain MMLR of 0.33%, 0.035%, 0.099%, and 0.008% for gathering facilities, gathering pipelines, processing plant normal operations and processing plant blowdowns, respectively, and an overall MMLR of 0.47% for all U.S. gathering and processing. The MMLR is 87% higher than the 2012 CH_4 loss rate of 0.25% for all gathering and processing operations derived from the 2014 EPA GHGI. The results are summarized in Table S11 (SI Section S.8).

The results of the Monte Carlo simulation were also used to develop "facility-level emission factors" for gathering facilities and processing plants, respectively (SI Section S.8.1). For processing plants, the national estimate of 546 (+55/−42) Gg of CH_4 emissions from 592 ± 14 Subpart KKK processing plants, yields an estimated facility-level emission factor of 105 (+11/−10) kg/h-plant in comparison to 168 kg/h-plant as derived from the EPA GHGI. Similarly, for gathering facilities, the national estimate of 1697 (+189/-185) Gg of CH_4 emissions from 4549 (+921/−703) gathering facilities yields an estimated facility-level emission factor of 43 (+10/−8) kg/h-facility in comparison to 5.7 kg/h-facility as derived from our analysis of the EPA GHGI production inventory (SI Section S.6.2). These results are summarized in Table S12 (SI Section S.8.1).

In the context of evaluating the greenhouse gas emission implications of NG substitution into various energy sectors (e.g., coal power plants, heavy duty diesel vehicles, and light duty gasoline vehicles), an overall rate of CH_4 loss of 0.47% from gathering and processing is not trivial, particularly with respect to those applications with the lowest tolerance for CH_4 emissions in terms of net short-term climate benefits (i.e., heavy duty diesel vehicles).[5,7,8] However, the results also show that CH_4 emissions from NG processing plants during normal operation (506 +55/−52 Gg, 0.099% loss rate when normalized by production) represent a relatively small fraction of the CH_4 emissions from the overall NG supply chain. While emissions from gathering facilities (1697 +189/−185 Gg; 0.33% loss rate when normalized by production) represent a substantially higher fraction of the total U.S. CH_4 emissions in comparison to processing, observations from the field campaign[15,16] suggest that further reductions in CH_4 emissions can be realized by incorporating systems and/or practices to quickly identify and repair the high emitting gathering facilities.

Two areas of future work are recommended. First, as mentioned above, the EPA GHGI does not include gathering as a separate sector in its NG systems inventory. However, the results of this study indicate that CH_4 emissions from gathering facilities represent a sizable fraction of CH_4 emissions from the NG supply chain. The results of this study in conjunction with the results of recent studies on production sector emissions[10,13,14] can provide input to the EPA for a separate gathering sector inventory in future versions of the GHGI. A proposed rule change to the EPA GHGRP that would require reporting of CH_4 emissions from gathering and boosting stations (SI Section S.7.1) would provide further input for a separate GHGI gathering inventory. Second, emissions from gathering pipelines were not measured as part of this study. The 445 135 miles of gathering pipeline in the 2014 EPA GHGI is based solely on extrapolations from rough estimates of pipeline miles per well made in the early 1990s[12] and the emission factors (CH_4 emissions per mile) have never been accurately quantified for gathering pipelines. Accordingly, it is recommended that future studies be conducted to more accurately survey the length of and quantify the CH_4 emissions from the U.S. national gathering pipeline network.

■ ASSOCIATED CONTENT

Supporting Information

S.1 Background Information; S.1.1 EPA GHGI Estimate of Methane Emissions and Methane Loss Rate; S.1.2 Gathering and Processing Sector Definitions; S.2 Description of the Field Campaign; S.2.1 Definition of Facility Types; S.2.2 Natural Gas Compressor Engine Types and Uncombusted Methane; S.3 Facility-Level CH_4 Emissions Results; S.3.1 Field Campaign Sample Size and Frequency of Super-Emitters; S.4. Model Description; S.4.1 Processing Plant Data; S.4.2 Facility Natural Gas Throughput and Engine HP Relationship; S.4.3 Estimation of State Gathering Facility Counts; S.4.4 Estimation of Confidence Intervals for State Gathering Facility Counts; S.4.5 Monte Carlo Simulation Scheme for Gathering Facilities; S.4.6 Monte Carlo Simulation Scheme for Processing Plants; S.5 Model Results; S.5.1 Potential Sources of Bias in Model Predictions; S.6 Comparisons with EPA Greenhouse Gas Inventory; S.6.1 Natural Gas Processing; S.6.2 Natural Gas Gathering; S.7 Comparison with EPA Greenhouse Gas Reporting Program; S.7.1 Proposed Rule Changes to EPA GHGRP for Gathering and Boosting; S.8. National Estimate of CH_4 Emissions and Loss Rate from G&P Systems; S.8.1 Facility-Level Emission Factors; S.9 State Gathering Facility Data; Data Tables in excel format: SI-Tables20150615.xlsx; Gathering and processing facility lists: facility-list20150615.xlsx This material is available free of charge via the Internet at http://pubs.acs.orgThe Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b02275.

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Author Contributions

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■ ABBREVIATIONS

FLER	Facility-level emission rate (weighted average of multiple plumes) [kg/h]
MMLR	Modeled methane loss rate [%]
MMscfd	Million cubic feet per day of natural gas at standard conditions (60 °F and 1 atm)
SMLR	Sampled methane loss rate [%]
tnFLER	Throughput normalized facility-level emission rate [%]
Tscf	Trillion cubic feet of natural gas at standard conditions (60 °F and 1 atm)

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