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Attachment 2 of 2 for SoCalGas Comments on June 1, 2015 Workshop

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Direct Measurements Show Decreasing Methane Emissions from Natural Gas Local Distribution Systems in the United States

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Supporting Information

ABSTRACT: Fugitive losses from natural gas distribution systems are a significant source of anthropogenic methane. Here, we report on a national sampling program to measure methane emissions from 13 urban distribution systems across the U.S. Emission factors were derived from direct measurements at 230 underground pipeline leaks and 229 metering and regulating facilities using stratified random sampling. When these new emission factors are combined with estimates for customer meters, maintenance, and upsets, and current pipeline miles and numbers of facilities, the total estimate is 393 Gg/yr with a 95% upper confidence limit of 854 Gg/yr (0.10% to 0.22% of the methane delivered nationwide). This fraction includes emissions from city gates to the customer meter, but does not include other urban sources or those downstream of customer meters. The upper confidence limit accounts for the skewed distribution of measurements, where a few large emitters accounted for most of the emissions. This emission estimate is 36% to 70% less than the 2011 EPA inventory, (based largely on 1990s emission data), and reflects significant upgrades at metering and regulating stations, improvements in leak detection and maintenance activities, as well as potential effects from differences in methodologies between the two studies.

INTRODUCTION

Methane (CH_4) emissions from the natural gas supply chain account for approximately 30% of the total United States CH_4 emissions.[1] Recent developments in shale gas extraction have resulted in an increased use of natural gas and decreased use of coal and other fossil fuels.[2] Natural gas combustion results in lower carbon dioxide (CO_2) emissions compared to the combustion of coal or oil. However, an increase in throughput of natural gas may increase CH_4 emissions due to greater atmospheric losses. Because the global warming potential of CH_4 is 28 to 34 times greater than CO_2 on a 100 year time frame and up to 84 times greater over a 20-year time frame,[3] an increase in CH_4 emissions may diminish the CO_2 reduction benefit associated with using natural gas as an energy source.[4,5] Near-term reductions in CH_4 emissions are a vital tool for slowing the rate of climate change,[5] and as a complement to long-term reductions in CO_2. Therefore, an accurate estimate of the leak rate of CH_4 from natural gas infrastructure is needed to understand the climate impacts of natural gas use and to identify opportunities for overall reductions in CH_4 emissions.

Much of the data used by the US Environmental Protection Agency (EPA) to estimate CH_4 emissions from the natural gas industry were collected in the 1990s as part of a study by the Gas Research Institute (GRI) and EPA[6] (hereafter, GRI/EPA or GRI/EPA 1992 study, since the base year for the inventory was 1992). The GRI/EPA study compiled CH_4 emission factors (EFs) for components within the industry and developed estimates of the population of each component type (activity factors, AF) across the U.S. The products of EF × AF for each source category were used to compile a national estimate for CH_4 emissions from the natural gas industry. In the EPA emission inventory[1] for the year 2011 (hereafter, 2011 EPA inventory), current AFs are used with the original GRI/EPA EFs (with minor revisions) to calculate the annual CH_4 emission rate from natural gas infrastructure of 6890 Gg/yr,

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with local distribution systems accounting for 1329 Gg/yr, or 19% of the total from the natural gas supply chain and 0.33% of gas delivered to customers.

Considerable changes have occurred in local natural gas distribution systems since the 1990s. There have been substantial replacement and upgrades of equipment within metering and regulating (M&R) facilities along with reductions in miles of older cast iron (−38% to ∼33 000 total miles) and unprotected steel pipeline (−22% to ∼66 000 total miles), and increases in protected steel (+8% to ∼480 000 total miles) and plastic (+150% to ∼620 000 total miles) pipeline miles.[1,7] Leak survey methods have improved since the 1990s, with an increased emphasis on reporting of CH_4 emissions.[8] However, a new assessment of CH_4 emissions from U.S. natural gas distribution systems in response to these changes has not occurred.

Here we present results of direct measurements of CH_4 emissions from underground pipelines and M&R facilities across the U.S. These data were used to develop new EFs as the basis for a revised estimate of CH_4 emissions from natural gas distribution systems. We also compiled information from company surveys to update estimates for emissions from maintenance blow-downs and pipeline dig-ins. We use these results to provide a new estimate of the total amount of CH_4 emitted from the US natural gas distribution system.

EXPERIMENTAL SECTION

Scope of Study. At the beginning of the study, we used the available 2010 EPA emission inventory coupled with uncertainty estimates from the 1992 GRI/EPA study to develop a stratified random sampling plan targeting the largest CH_4 source categories (see Supporting Information SI Section S2.1). On the basis of this analysis, eight categories were targeted for sampling (in order of estimated emissions): M&R with inlet pressures between 100 and 300 psi, plastic mains, unprotected steel services, unprotected steel mains, cast iron mains, regulators >300 psi, M&R > 300 psi, and regulators 100−300 psi. This list includes Transmission−Distribution Transfer Stations (TDTS), also called city gates, where gas custody is transferred from the transmission pipeline to the distribution system. Sampling within these target categories occurred from May through November 2013 at 13 local distribution companies (LDCs) across the U.S. Wintertime conditions were not sampled, and there is little information available to suggest how this might bias the results. While these LDCs represent less than one percent of the 1400 distribution companies in the U.S.,[7] they have 19% of the distribution pipeline mileage (∼226 000 miles), 26% of the services (∼15 million services), and deliver 16% of the total gas delivered to customers in 2011. These companies, thus, account for a significant percentage of natural gas distributed nationally, although we recognize the potential bias in our results because companies volunteered to participate in this study, which was essential for access to facilities for measurements. Pipeline replacement rates for cast iron and unprotected steel mains in our partner companies are similar to other LDCs nationwide. Data from the DOT pipeline program[7] show that the miles of cast iron and unprotected steel mains have decreased due to replacement by 20% from 2005 through 2013 and that for our partner companies, the miles of cast iron and unprotected steel have remained a constant fraction (16% ± 1%) of the decreasing national total miles during this period.

To develop a representative database, a random selection process was developed so that measurements were obtained within targeted, representative areas that we selected within each company's distribution system. Specific pipeline leaks and facilities were selected randomly from LDC leak survey data and facility lists for the targeted areas. Class 1 pipeline leaks were not measured since these leaks are repaired immediately for safety reasons. Because leaks are classified on the basis of safety (i.e., proximity to buildings) and not magnitude, class 1 leaks are not necessarily larger than class 2 or 3 leaks. Further information on the stratified random sampling plan and the partner LDCs is provided in SI Section S2.0. Our study does not address emissions downstream of customer meters or other portions of the natural gas supply chain in urban areas, including natural gas transmission lines and compressor stations, natural gas vehicles and fueling stations, and liquefied natural gas terminals and storage facilities.

Sampling Methods. The high-flow sampling method[9−11] was the primary measurement technique used to quantify leak rates on individual components at M&R stations (SI Section 3.1). The high flow sampler uses a high flow rate (6−8 standard cubic feet per minute) of air and a modified enclosure to completely capture the gas leaking from a component. Catalytic oxidation and thermal conductivity hydrocarbon sensors measure the CH_4 concentration in the air stream, and a thermal gas flow sensor measures sample flow rate. A version of the high-flow technique, modified to include a 1.2 × 1.2 m^2 surface enclosure and a CH_4 detector with a detection limit <100 ppmv was used to measure surface CH_4 emissions from underground pipeline leaks (SI Section 3.2). High-flow measurements were supplemented, for quality assurance purposes, by downwind tracer-ratio measurements with instruments mounted in a van[12] (SI Section S3.3). We found moderate (±50%) to excellent (±5%) agreement between the downwind tracer-ratio method and high-flow sampling methods (SI Sections S4.11 and S5.2). For six different pipeline leaks where we had tracer and direct measurements, total summed emissions measured were 4.85 and 5.83 g/min for the high flow and tracer methods, respectively, which yields an overall difference (19%) within the experimental uncertainties. Similarly, for eight M&R facilities, the total summed emissions were 66.0 and 51.6 g/min for the high-flow and tracer-ratio methods, respectively, a difference of 24%, within the range of the experimental uncertainty (see SI Sections 3.0 on methods, Sections S4.1.1 and S5.2 on tracer results and SI Appendix B on uncertainty analyses).

Statistical Methods. The population of measured leak rates generally shows marked asymmetry, with a few high emitters accounting for a large fraction of the total measured emissions, requiring highly skewed probability distributions as models. We considered eight different probabilistic models for each data category—Gaussian, log-normal, gamma, Weibull, hyperbolic, inverse Gaussian, Johnson, and generalized Tukey's lambda distributions—and compared them using the Bayesian Information Criterion (BIC), supplemented by inspection of QQ-plots (see SI Section 3.6 and references therein). Once a model was selected for a category, and its parameters estimated, 105 sample data sets were drawn from the fitted model in a manner that recognizes the uncertainty of the fitted parameters, where each of these samples was the same size as the original data set, and their averages were computed. The overall average from these bootstrap data sets was the estimate of the mean leak rate for the corresponding source category, and the 95th

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Table 1. Comparison of National Methane Emission Factor Estimates from Underground Pipeline Leaks Based on the Current Study and the 1992 EPA/GRI Study

	this study			1992 GRI/EPA		
pipeline material	*n*	emission factor (g/min)	95% UCL (g/min)	*n*	emission factor (g/min)	95% UCL (g/min)
			main pipelines			
cast iron	14	0.90	3.35	21	3.57[a]	5.60[a]
unprotected steel	74	0.77	2.07	20	1.91	3.70
protected steel	31	1.21	4.59	17	0.76	1.40
plastic	23	0.33	0.67	6	1.88	8.20
			services			
unprotected steel	19	0.13	0.19	13	0.34	0.54
protected steel	12	0.33	0.93	24	0.74	1.53
plastic	38	0.13	0.19	4	0.11	0.27

[a]GRI/EPA EF converted from SCF/mile to g/min/leak using cast iron pipeline miles and equivalent leaks from this study.

Table 2. Comparison of National Methane Emission Factors for Metering and Regulating Facilities Based on the Current Study and the 1992 EPA/GRI Study

	this study			1992 GRI/EPA		
facilities	*n*	emission factor (g/min)	95% UCL (g/min)	*n*	emission factor (g/min)	95% UCL(g/min)
			M&R stations			
>300 psi	59	4.06	7.67	31	57.4	79.6
100−300 psi	10	1.88	1.88	6	30.5	64.6
<100 psi	0	–	–	3	1.4	4.5
			regulating stations			
>300 psi	41	1.64	4.85	13	51.6	81.4
100−300 psi	41	0.27	0.73	7	12.9	21.4
40−100 psi	13	0.31	0.73	7	0.32	0.60
<40 psi	1	0.0	0.0	0	–	–
vaults[a]	23	0.10	0.13	28	0.03- 0.41	0.06−1.18

[a]All pressure categories are combined for underground vaults.

percentile of the same set of samples, defined as the 95% upper confidence limit (UCL), expresses the uncertainty associated with this overall mean. Further details are provided in SI Section 3.6.

RESULTS AND DISCUSSION

Emissions from Underground Pipelines. Methane emissions from 230 individual underground pipeline leaks were measured to form the basis for new pipeline EFs. This sample of leak measurements is twice as large as that used in the 1992 GRI/EPA database, although it is still a small fraction of total leaks in the U.S. On the basis of our stratified sampling plan, the emission rate measurements were from cast-iron, unprotected steel, cathodically protected steel, and plastic main and service pipelines. Typically, cast-iron and unprotected steel pipe have more leaks per mile than protected steel and plastic pipe.[6] Emission factors from pipeline mains ranged from 0.3 to 1.2 g/min/leak, while EFs from pipeline services ranged from 0.1 to 0.3 g/min/leak (Table 1). The estimated 95% UCLs on these EFs were factors of 2 to 4 times larger than the mean EFs.

We found that three large leaks (34.9, 22.2, and 4.9 g/min, from unprotected steel main, protected steel main, and cast iron main leaks, respectively, accounted for 50% of the total measured emissions from pipeline leaks. This type of distribution, where a few leaks account for a large fraction of the total CH_4 emitted, is not unexpected, and it has been observed in other emission studies.[6,13,14] For these skewed distributions, as described previously, the estimated mean for a sampled population and the corresponding UCL are best estimated from explicit probabilistic modeling of the skewed distribution of the measurements to find the distribution type which best matches the observations in each category (see SI, section S3.6, Figures S3.12, S4.1, S4.5).

Our EFs for underground pipeline leaks were about two times lower than reported in the 1992 GRI/EPA study[6] (see Table 1). The maximum emission rates measured in our study were similar to those in the GRI/EPA study, on the order of 30 g/min/leak. For smaller leaks, the GRI/EPA results were larger than the emission rates measured in the current study (median emission rate of 0.6 g/min/leak, versus 0.06 g/min/leak, respectively). Therefore, it is clear that our leak distribution has much lower leak rates than the GRI/EPA study (see SI Figure S4.5).

There are important categorical differences between our measurements and the 1992 GRI/EPA study. The EF for plastic mains in the GRI/EPA work was almost seven times larger than our estimate (0.33 g/min/leak). In this case the GRI/EPA plastic main EF was based on a relatively small sample size of six including one very large leak. Furthermore, recent measurements by the Gas Technology Institute[14] (GTI) also suggest lower EFs (1.0 ± 1.2 g/min/leak) than the rate used by EPA for plastic mains (1.88 g/min/leak) and the GTI rate is similar to our EF for plastic mains when corrections are made for the GTI detection limit (see SI Section S4.2). For leaks from cast iron mains, the GRI/EPA EF was reported on a per foot basis, which makes it difficult to compare to our measurements of emissions per leak. For protected steel mains and plastic services, our EFs were slightly higher than GRI/EPA. The reasons for these differences include better leak

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Table 3. Comparison of Results for High Emitting City Gates in the GRI/EPA Study with Results from Re-Visiting These Same Sites in This Study[a]

facility	GRI/EPA methane ER (g/min)	this study methane ER (g/min)	ratio (1992/2013)	facility modifications
A	162	30.3	5.30	
B	118	6.14	19.3	rebuilt
C	62	8.49	7.2	
D	40	56.2	0.70	no changes
E	29	0.543	53	
F	27	14.6	1.9	
G	24	5.19	4.6	
H	23	1.30	18	rebuilt
I	18	6.14	2.9	
			13	**average ratio**
totals for revisited sites	**504**	**129**	**3.9**	**ratio of totals**

[a]Blank cells indicate no information available from facility operators.

detection technology now compared to the 1990s, replacement of older pipelines, better maintenance activities, and, possibly, methodological differences between this study and the GRI/EPA work.

In the GRI/EPA study, leak rates were measured by digging and isolating pipe sections to measure leak flow rates, which were then adjusted empirically to account for oxidation of CH_4 in the soil.[6] The soil oxidation correction varied from a few percent for large leaks to as much as 40% for leaks from cast iron mains.[15] In this work, a surface enclosure was used to measure the emissions at the ground surface with no disturbance of the pipe and no corrections needed to account for soil oxidation. Considerable care was taken to completely map and then measure the surface expression of each leak using a series of gridded enclosure placements (SI Figure S3.3), and we also found good agreement between the surface enclosure method and an independent tracer-ratio approach (see SI Section S5.2). In the GRI work, LDCs conducted the pipe isolation leak measurements on sections of pipe scheduled for replacement, and audits were conducted to ensure that each company used consistent methods. In our work, leaks were selected randomly from the company leak survey database within the general area we had selected from each LDC service region (SI Section 2.0). It is not possible to determine how these differences might have affected the results in terms of the overall sample population or individual measured leak rates, although GTI showed good agreement between their surface enclosure measurements and a pipe isolation method[14] similar to that used in the GRI/EPA study.

Emissions from Metering and Regulating Stations. We completed measurements at 229 different M&R facilities including 48 TDTS stations (city gates). In the GRI/EPA 1992 study, 55 such facilities were measured. Emission factors for M&R stations are summarized in Table 2 for the different facility categories used in the emission inventory. We found higher emissions for facilities with higher inlet pressures, and lower emissions for vaulted (i.e., below grade) facilities. For facilities with inlet pressures >100 psi, the EFs range from 0.3 to more than 4 g/min/station. For vaulted facilities, the emissions are less than 0.1 g/min/station. In each case, the distribution of measured emission rates is skewed with median emission rates much less than the mean.

M&R stations sometimes have vented devices, such as odorizers and pneumatic controllers, designed to emit natural gas as part of their normal operation. We measured emissions from these devices at M&R facilities and found that they have highly variable emission rates over short periods of time (SI Section S5.1). Therefore, measurements were collected using a high-flow sampling system coupled to a data system to record emissions over 15 to 30 min periods. The emissions from vented devices typically represent a significant fraction of the total emissions for facilities so equipped (SI Section S5.1). The EFs for odorizers and pneumatic controllers measured during our study were 2.2 and 4.9 g/min as compared to whole facility EFs that ranged from less than 1 g/min to more than 4 g/min.

There are significant differences between the emission factors from the GRI/EPA 1992 study and our measurements (Table 2) for M&R facilities. For the larger emitting categories, the GRI/EPA EFs are more than 14 times larger than our EFs. These differences are apparent in the frequency distribution of emissions from all M&R stations, where the maximum emission rate measured in the GRI/EPA work was 157 g/min/station while the maximum emission rate measured in our study was 56 g/min/station (SI Section S5.3; Figure S5.6). The large differences in the EFs are due to the upper 20% of the sites measured in the GRI/EPA work, since the median value in both studies is essentially identical at 0.3 g/min/station.

To understand the large reductions found in this work relative to the GRI/EPA results, we identified nine facilities from among the larger emitting sites measured during the GRI/EPA 1992 program to resample with our high-flow and tracer-ratio techniques (Table 3). These results show substantial reductions in emissions from each individual station (factors of 2 to 50) from 1992 to the present, with one exception. In two cases, the local operator indicated that significant equipment changes had occurred at the site; while at a third site, the local operator indicated that there had been no equipment upgrades at the site in the past 20 years. This particular site was the only site without a significant reduction in emissions. No information was available for equipment changes at the remaining sites. The data collected by resampling these facilities support our findings of substantial reductions in emissions from M&R facilities.

Because of the importance of facility equipment upgrades, we next surveyed the study partner LDCs and other LDC members of the American Gas Association (AGA) to determine how M&R sites have been upgraded since 1992 (SI Appendix G). Results obtained from five partner LDCs for 90 M&R sites of the 229 sites sampled in this study showed that approximately 60% of the 90 facilities had undergone some equipment change since 1992. Information on upgrades was not available for the remainder of the sampled sites. Our

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Table 4. Summary of the Overall Emission Inventory for U.S. Natural Gas Distributions Systems for This Study and the 2011 EPA GHG Inventory (1)

	this study		EPA 2011
category	methane emissions (Gg)	95% upper confidence limit (Gg)	methane emissions (Gg)
		pipelines	
mains	132	431	429
services	63.6	124	194
pipeline subtotal	197	554	623
		equipment	
M&R facilities	42.3	82.9	552
customer meters[a]	112	150	112
maintenance	1.6	2.5	3.7
upsets	41.6	64.1	38.9
equipment subtotal	197	300	706
total	393	854	1329

[a]EPA emission factor used for this category.

random sampling approach did not consider facility upgrades in sampling location selection. In addition, 14 LDC members from the AGA reported equipment upgrade activities with a total of 5267 out of 12 788, or 41% of facilities having upgrades. Furthermore, 43% of the responding companies reported rebuilding whole stations since the 1990s. It was clear from our interactions with M&R personnel that maintenance activities and attention to leaks have increased, in part, due to the GHG reporting requirements implemented in the past several years.[8] These results highlight the importance of making periodic emission measurements to account for upgrades and changes in the natural gas system, and point to the power of reporting requirements in helping to reduce emissions.

The GTI measured emissions from M&R stations using similar methods during 2008.[16] Our current EF for TDTS stations is lower than the GTI results, but within the large uncertainties associated with these measurements (SI Table S5.13). When the measurements are integrated over all M&R stations on a weighted basis to match the GTI results, the current EFs are approximately half of the GTI results, but still within the uncertainty estimates of the EF. For pressure regulating stations, a significant decrease occurs from the GRI/EPA data to the GTI data and from GTI to our study (see SI Table S5.13). Overall, the GTI results are consistent with our results and with significant upgrades in equipment and procedures for M&R stations from 1992 to the present, although there may be differences in how the GTI study selected stations and integrated the results.

National Emission Inventory. The U.S. natural gas distribution system has undergone modernization and growth since 1992.[1,7] Pipelines mains and services have increased by 44% to 1.2 million miles and 63 million services, respectively. Modernization of the system has led to substantial reductions in the miles of cast iron (−38% to ∼33 000 miles) and unprotected steel pipelines (−22% to ∼66 000 miles). At the same time, there has been an increase in miles of plastic mains (150% to ∼620 000 miles), protected steel mains (8% to ∼480 000 miles), and plastic and copper services (150% to 43 million services and 352% to 1 million services, respectively). For M&R stations, there has been an increase in the number of stations by about 8% to approximately 150 000 stations. For customer meters, there has been an increase of 6% in the number of residential meters to 42 million meters, but a 5% decrease in commercial meters to 4 million meters.

To quantify the total CH_4 emitted from underground pipelines in the U.S., we developed an AF for underground pipelines (units of leaks per mile of pipe or number of services). Because such information is not available on the national scale, we use the concept of an "equivalent leak" where an equivalent leak represents a leak that exists for one year.[6] For each LDC, equivalent leaks account for the number of annual leak indications (including customer call-ins), an estimate of the number of actual leaks based on leak indications (assuming the company does not know about all of their outstanding leaks), the number of annual leak repairs, and the average time between leak indication and leak repair. In the GRI/EPA study, survey results from four LDCs were used as the basis for equivalent leak calculations. We followed the same approach and obtained data from six of the study partner LDCs (see SI Section 4.3 and SI Appendix D). Class 1 leaks were not measured, but were included in the equivalent leak calculations. Since Class 1 leaks are repaired as soon as possible, including them in the equivalent leak calculations is conservative in that it will result in greater national emissions.

The annual CH_4 emissions in each category for the U.S. are calculated by multiplying the AF (number of equivalent leaks) in each pipeline category by the appropriate EF (Table 1). On the basis of our estimates, the national total is 197 Gg/yr with a 95% UCL of 554 Gg/yr, where the UCL only accounts for the uncertainty in the EF values. The uncertainty in AFs in this study and the 1992 GRI/EPA study are similar, on the order of ±30%. These uncertainties are due to variability among the companies surveyed regarding the number of leak repairs, the time between leak detection and repair, and the number of pipe miles. Annual emissions due to leaks in pipeline mains account for 67% of the total underground pipeline emissions. Even though cast iron and unprotected steel mains represent less than 10% of national distribution system pipeline miles, the emissions from these two categories account for 46% of the total emissions from pipeline mains.

The annual emissions from pipeline leaks estimated in our study are approximately 32% of the 2011 EPA estimates of 623 Gg/yr (Table 4) and approximately 26% of the 1992 GRI/EPA estimates of 751 Gg/yr (SI Table S4.8). This is due to a combination of lower EFs (CH_4 emitted per leak) and lower AFs (equivalent leaks in the U.S.). The GRI/EPA 1992 total estimate of 751 Gg/yr decreases to 483 Gg/yr when the GRI/EPA EFs are used with EPA 2011 AFs (SI Table 4.8). Therefore, roughly half of the decrease from the 1992 estimate

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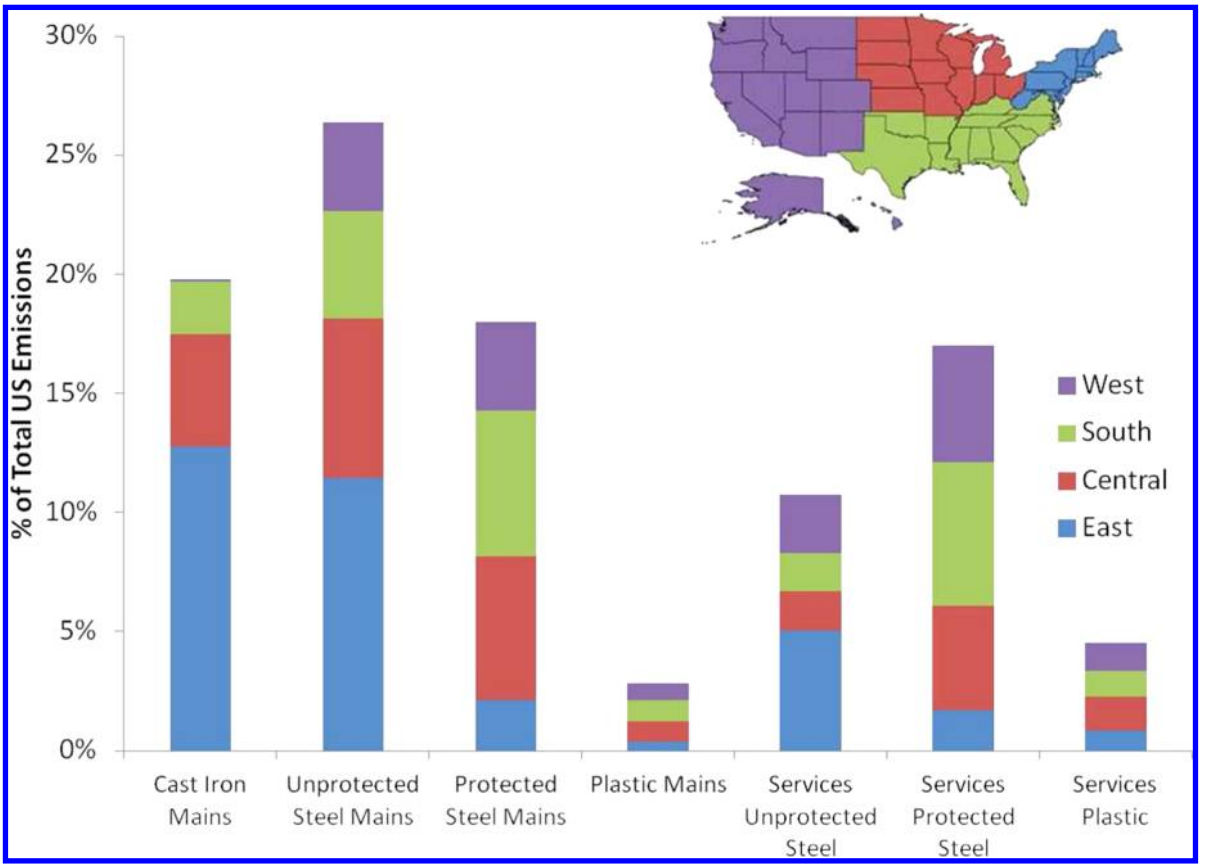

Figure 1. Percentage of total U.S. methane emissions from underground pipeline leaks by region and by pipeline type and category. The total U.S. emission estimate for pipeline leaks is 197 Gg/yr with a 95% upper confidence limit of 554 Gg/yr.

is due to reductions in AF and the other half is due to the aforementioned reductions in EFs.

The primary reason for reductions in AFs is the replacement of older cast-iron and unprotected steel pipe with plastic (see SI Section 4.3 and Tables S4.5−8). Specifically, the number of pipeline leaks has decreased between 25% and 16% for pipeline mains and services, respectively, due to the use of better pipe materials, efforts to seal cast iron joints, and enhanced leak detection and repair procedures. A survey of AGA LDCs made during this study indicates that substantial cast-iron pipe replacement and joint-sealing activities are being conducted in the U.S. In fact, over half of the 20 gas companies who provided information during the survey reported sealing roughly one cast-iron joint per mile of cast-iron pipe in 2011 (SI Appendix G).

As previously mentioned, our emission rate measurements in each category exhibited a skewed distribution, and while this is typical of CH_4 emission studies, these distributions result in large upper confidence limits. In our case, our national emission estimate of 197 Gg/yr has a 95% upper confidence limit of 554 Gg/yr that is within ∼10% of the 2011 EPA emission estimate of 623 Gg/yr. Given the effect that just a few large leaks have on the mean EF, it is important to recognize the upper bound as an integral part of any comparison with other emission estimate methods.

We also examined how emissions from pipeline leaks varied on a regional basis in the U.S. due to differences in pipeline type and miles by region (see SI Section S4.3; there was no statistical difference in EFs by region). The eastern region accounts for 34% of the total U.S. CH_4 from pipeline leaks, while the western region contributes less than 20% (Figure 1). In the eastern region, emissions are dominated by leaks from cast iron and unprotected steel characteristic of older systems. As such, leaks from cast iron and unprotected steel pipe account for 70% of the eastern emissions and almost half of total U.S. emissions. In the western region, systems are newer with more miles of plastic and protected steel pipe, and leaks from these systems contribute less than 5% of the total U.S. emissions. These regional variations and the low emissions associated with plastic pipes are significant as the U.S. moves toward replacement of older pipelines with plastic and uses plastic for new distribution expansion.

To extrapolate to a national level for the M&R emissions, we use the same categories as used in the 2011 EPA GHG emission inventory along with current AF for each category. For the present study, the results indicate a total CH_4 emission rate from M&R stations of 42 Gg/yr with a 95% UCL of 83 Gg/yr (Table 4). The top two contributing categories are M&R (>300 psi) stations, which includes TDTS stations, and M&R (100−300 psi) (see SI Table 5.14), and these account for more than half of the estimated emissions.

Our annual CH_4 emission total for M&R stations in the U.S. is significantly lower than the 2011 EPA estimate (552 Gg/yr) by factors of 7 to 13 (Table 4). These differences are large, but are supported by significant differences in emissions at the revisited large emitting sites from the GRI/EPA study and from industry information, which indicates significant improvements in equipment and maintenance. These differences are also supported by the results from the GTI study,[16] which also

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showed significant decreases in emissions for M&R facilities compared to the GRI/EPA 1992 work.

For comparison to the EPA inventory for distribution systems, we used results from surveys of AGA companies to update estimates for maintenance and mishaps (see SI Appendix G). Together, our estimates for CH_4 losses from pipeline leaks, M&R facilities, maintenance activities, and mishaps, along with the EPA estimate for customer meters, address emissions from U.S. local distribution systems up to and including the customer meter. Our estimate for these categories for the total U.S. emission rate is 393 Gg/yr with a 95% UCL of 854 Gg/yr. The UCL on this new inventory is approximately 36% less than the EPA 2011 emission inventory, while the mean emission total is 70% less than the EPA estimate. The reduction in the national total is due to a combination of lower EFs and AFs. Changes in EF are clearly linked to equipment upgrades at M&R stations and to changes in pipeline leak survey methods, replacement of older pipe, and better maintenance efforts. There may also be a difference in EFs due to the differences in sampling methodologies used here vs the original GRI/EPA work, but the effects of these differences in methods are difficult to determine. The 2012 EPA inventory, currently in draft form, shows a decrease of 100 Gg/yr compared to the 2011 EPA inventory, which does not substantially change the comparison. Our new estimate represents 0.10% to 0.22% of the CH_4 delivered via the distribution system. Our results also show considerable differences on a regional basis throughout the U.S. because of differences in pipeline types and miles by region.

The magnitude of the UCL is due to the skewed distribution of measurements collected in this study and is typical of emission rate measurements from the natural gas distribution system. The upper limit also includes uncertainties for customer meters, maintenance, and mishaps (e.g., accidental dig-ins) that were estimated from company surveys in a manner similar to that used in the GRI/EPA study. For customer meters, GTI conducted high-flow measurements on 2800 customer meters in 2008.[16] If the GTI EFs are used in place of the EPA 2011 emission estimate for customer and commercial meters, then the U.S. total emissions for these meters decreases from 112 Gg/yr to 81 Gg/yr.

While our study provides a significant increase in the number of measurements for pipeline leaks and M&R facility emissions, additional sampling would improve our understanding of the frequency distribution of leaks, particularly for the few large leaks that seem characteristic of the distribution. As noted previously, we were limited to LDCs which volunteered to participate in this work; uncertainties remain regarding leak rates in other locations. However, we might expect leak frequency to differ among LDCs due to maintenance and pipeline material differences, but the actual leak rates (EFs) might be expected to be similar. We were also limited to nonwinter sampling conditions; the effects of frozen soils upon pipeline leak rates and greater natural gas throughput in winter months have not been addressed in this work. Looking forward, technology that would allow rapid leak detection and direct measurement of emission rates would expand the database of leaks and reduce the uncertainty in EFs. Additional efforts to develop AFs by surveying more companies would also help to reduce uncertainties in these bottom-up estimates.

Top-down emission estimates, which infer emission rates from ambient CH_4 observations, are vital in constraining emission estimates. These approaches typically provide larger emissions estimates than bottom-up approaches,[17] which indicates that further work is required to address sources not explicitly included in our direct source measurements. For example, McKain et al.[18] have reported top-down CH_4 and ethane measurements in Boston, MA with inverse modeling analyses that suggest natural gas sources account for 60% to 100% of the enhanced CH_4 levels depending on the season of the year. Similar results have been reported elsewhere from top-down studies,[19,20] and this seems to be supported by nonquantitative city street surveys of CH_4 concentrations.[21,22] Further work on reconciling bottom-up emission inventories with top-down emission estimates is needed to address all of the sources contributing to CH_4 emissions from the natural gas supply chain in urban areas since top-down methods cannot yet provide specific source attributions. These include emissions downstream of customer meters from industrial facilities, commercial structures, and residential housing, emissions from pipeline leaks that migrate into sewer lines and vents, emissions from transmission lines and compressor stations within urban areas, from natural gas vehicles and refueling stations, from liquefied natural gas terminals and storage facilities, or other unidentified sources. Such efforts are underway in Indianapolis, IN,[23] among other urban areas, and EDF is sponsoring emission studies of several of these source sectors. Additional work is needed to treat seasonal differences such as reported in Boston.[18]

In summary, this survey of methane emissions from a sample of the natural gas distribution systems of the U.S. is based on direct measurements and is the most comprehensive since that of the 1990s. Instances of significant emissions reductions have been quantified, in particular, reductions ranging from approximately a factor of 2 to 50 for some M&R stations, and illustrate the impact of two decades of advances in technologies and changes to operational procedures that reduce emissions.

■ ASSOCIATED CONTENT

Supporting Information

Project overview; study scope; local natural gas distribution systems; sampling approach; sampling strategy; stratified random sampling; representativeness of the LDC sampling regions; sample selection; emission rate measurement methods; high flow component leak measurements at TDTS and M&R facilities; surface enclosure underground pipeline leak measurements; tracer ratio leak methods for facility and pipeline leaks; isotopic methane sampling; mobile methane mapping; probabilistically modeling of mean and upper bound emission factors; probabilistic modeling—methods; probabilistic modeling—results; underground pipeline emissions; pipeline leak measurement results; tracer ratio results; isotopic analyses; pipeline emission factor analysis; national extrapolation of emissions from pipeline leaks; M&R facility emissions; vented emission measurements; odorizers; pneumatic controllers; tracer ratio results; M&R facility emission factor analysis; national extrapolation of emissions from M&R facilities; national emission inventory; references; glossary; and additional figures and tables. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare the following competing financial interest(s): Brian Lamb served on the Science Advisory Panel for the University of Texas study for natural gas emissions from production facilities and is a co-author on the Allen et al. (2013) paper describing that work. B.K.L., T.H., C.E.K., M.R.H. were participants in the 1992 GRI/EPA national sampling program (Harrison et al., 1996). Conestoga-Rovers & Associates (T.W.F., T.H., and W.D.) have a number of natural gas production and distribution companies as clients. C.E.K. and M.R.H. were members of the measurement team for the University of Texas led study and were co-authors of the Allen et al. (2013) PNAS article. C.E.K. is also a member of the Science Advisory Board for the ongoing Environmental Defense Fund organized Methane Monitoring Technology Initiative..

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■ REFERENCES

(1) *Inventory of U. S. Greenhouse Gas Emissions and Sinks: 1990–2010*; EPA430-R-12–001; United States Environmental Protection Agency: Washington, DC, 2013; http://www.epa.gov/climatechange/ghgemissions/usinventoryreport.html.

(2) *Annual Energy Outlook 2013 with Projections to 2040*; DOE/EIA-0383; United States Energy Information Administration: Washington, DC, 2013; http://www.eia.gov/.

(3) Myhre, G. et al. Anthropogenic and natural radiative forcing. In *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*; Stocker, T. F.; et al., Eds; Cambridge University Press: Cambridge, U.K., 2013.

(4) Wigley, T. M. L. Coal to gas: The influence of methane leakage. *Clim. Change* **2011**, *108*, 601–608.

(5) 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. Nat. Acad. Sci.* **2012**, *109*, 6435–6440.

(6) *Emissions from the Natural Gas Industry Vol. 1: Executive Summary*; 94/0257; Gas Research Institute/Environmental Protection Agency; Harrison, M. R., Shires, T. M., Wessels, J. K., Cowgill, R. M., Eds.; Chicago, 1996; http://www.epa.gov/gasstar/documents/emissions_report/1_executiveummary.pdf.

(7) United States Department of Transportation Pipeline and Hazardous Materials Safety Administration Website; http://phmsa.dot.gov/pipeline/library/data-stats.

(8) *USEPA Greenhouse Gas Reporting Program 2013—Petroleum and Natural Gas Systems*; United States Environmental Protection Agency: Washington, DC, 2013; http://www.epa.gov/ghgreporting/ghgdata/reported/petroleum.html.

(9) *Directed Inspection and Maintenance at Compressor Stations*; EPA 430-B-03–008; United States Environmental Protection Agency: Washington, DC, 2013; http://epa.gov/gasstar/documents/ll_dimcompstat.pdf.

(10) *A High-Flow Rate Sampling System for Measuring Leak Rates at Natural Gas Facilities*; GRI94/0257.38; Indaco Air Quality Services, Inc. and Gas Technology Institute: Chicago, IL, 1995.

(11) *Evaluation of the high volume collection system (HVCS) for quantifying fugitive organic vapor leaks*; EPA/600/SR-95/167; United States Environmental Protection Agency National Risk Management Research Laboratory, Research Triangle Park NC, 1996.

(12) Lamb, B. K.; McManus, J. B.; Shorter, J. H.; Kolb, C. E.; Mosher, B.; Harriss, R. C.; Allwine, E. G.; Blaha, D.; Howard, T.; Guenther, A.; Lott, R. A.; Siverson, R.; Westberg, H.; Zimmerman, P. Development of atmospheric tracer methods to measure methane emissions from natural gas facilities and urban areas. *Environ. Sci. Technol.* **1995**, *29*, 1468–1479.

(13) Allen, D. T.; Vincent, M.; Torres, J. T.; Sullivan, D.; Harrison, M.; Hendler, A.; Herndon, S. C.; Kolb, C. E.; Fraser, M.; Hill, A. D.; Lamb, B. K.; Miskimins, J.; Sawyer, R. F.; Seinfeld, J. H. Measurements of methane emissions at natural gas production sites in the United States. *Proc. Nat. Acad. Sci.* **2013**, *110* (44), 17768–17773 DOI: 10.1073/pnas.1304880110.

(14) *Improving Methane Emission Estimates for Natural Gas Distribution Companies: Phase II—PE Pipes*; Report No. 21044; OTD Project Number 7.10.c; Gas Technology Institute: Des Plaines, IL, 2013.

(15) Lamb, B. K.; Westberg, H.; Kashinkunti, R.; Czepiel, P.; Crill, P.; Harriss, R.; Kolb, C.; McManus, B. Methane oxidation in soils from underground natural gas pipeline leaks; Contract 5091 254 2148; Gas Research Institute: Chicago, IL, 1996.

(16) *Field measurement program to improve uncertainties for key greenhouse gas emission factors for distribution sources*; GTI 20497-OTD 7.7.b.GTI; Gas Technology Institute: Chicago, IL, 2009.

(17) 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.; Wofsy, S.; Brown, N. J.; Bradley, R.; Stucky, G. D.; Eardley, D.; Harriss, R. Methane leaks from North American natural gas systems. *Science* **2014**, *343*, 733–735 DOI: 10.1126/science.1247045.

(18) McKain, K.; Down, A.; Raciti, S. M.; Budney, J.; Hutyra, L. R.; Floerchinger, C.; Herndon, S.; Nehrkorn, T.; Zahniser, M. S.; Jackson, R. B.; Phillips, N.; Wofsy, S. C. Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proc. Nat. Acad. Sci.* **2015**, DOI: 10.1073/pnas.1416261112.

(19) Townsend-Small, A.; Tyler, S. C., Pataki, D. E., Xu, X., Christensen, L. E. Isotopic measurements of atmospheric methane in Los Angeles, Influence of "fugitive" fossil fuel emissions, California, USA. *J. Geophys. Res.* 2012, *117* (D7), D07308; doi:10.1029/2011JD016826.

(20) Wennberg, P. O.; Mui, W.; Wunch, D.; Kort, E. A.; Blake, D. R.; Atlas, E. L.; Santoni, G. W.; Wofsy, S. C.; Diskin, G. S.; Jeong, S.; Fischer, M. L. On the sources of methane to the Los Angeles atmosphere. *Environ. Sci. Technol.* **2012**, *46*, 9282–9289 DOI: 10.1021/es301138y.

(21) Phillips, N. G.; Ackley, R.; Crosson, E. R.; Down, A.; Hutyra, L. R.; Brondfield, M.; Karr, J. D.; Zhao, K.; Jackson, R. B. Mapping urban pipeline leaks: Methane leaks across Boston. *Environ. Pollut.* **2013**, *173*, 1–4.

(22) Jackson, R. B.; Down, A.; Phillips, N. G.; Ackley, R. C.; Cook, C. W.; Plata, D. L.; Zhao, K. Natural gas pipeline leaks across Washington, DC. *Environ. Sci. Technol.* **2014**, *48* (3), 2051–2058 DOI: 10.1021/es404474x.

DOI: 10.1021/es505116p
Environ. Sci. Technol. XXXX, XXX, XXX–XXX

(23) Cambaliza, M. O. L.; Shepson, P. B.; Bogner, J.; Caulton, D. R.; Stirm, B.; Sweeney, C.; Montzka, S. A.; Gurney, K. R.; Spokas, K.; Salmon, O. E.; Lavoie, T. N.; Hendricks, A.; Mays, K.; Turnbull, J.; Miller, B. R.; Davis, K.; Karion, A.; Moser, B.; Miller, C.; Obermeyer, C.; Whetstone, J.; Prasad, K.; Miles, N.; Richardson, S. Quantification and source apportionment of the methane emission flux from the city of Indianapolis. *Elementa* **2014**, DOI: 10.125952/journal.elementa.000037.

DOI: 10.1021/es505116p
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

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