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Fugitive methane emissions from leak-prone natural gas distribution infrastructure in urban environments *



POLLUTION

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A R T I C L E I N F O

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ABSTRACT

Fugitive emissions from natural gas systems are the largest anthropogenic source of the greenhouse gas methane (CH₄) in the U.S. and contribute to the risk of explosions in urban environments. Here, we report on a survey of CH₄ emissions from 100 natural gas leaks in cast iron distribution mains in Metro Boston, MA. Direct measures of CH₄ flux from individual leaks ranged from $4.0 - 2.3 \times 10^4$ g CH₄•day⁻¹. The distribution of leak size is positively skewed, with 7% of leaks contributing 50% of total CH₄ emissions measured. We identify parallels in the skewed distribution of leak size found in downstream systems with midstream and upstream stages of the gas process chain. Fixing 'superemitter' leaks will disproportionately stem greenhouse gas emissions. Fifteen percent of leaks surveyed qualified as potentially explosive (Grade 1), and we found no difference in CH₄ flux between Grade 1 leaks and all remaining leaks surveyed (p = 0.24). All leaks must be addressed, as even small leaks cannot be disregarded as 'safely leaking.' Key methodological impediments to quantifying and addressing the impacts of leaking natural gas distribution infrastructure involve inconsistencies in the manner in which gas leaks are defined, detected, and classified. To address this need, we propose a two-part leak classification system that reflects both the safety and climatic impacts of natural gas leaks.

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1. Introduction

Atmospheric methane (CH₄) concentrations have more than doubled in the past 150 years in conjunction with global industrialization and urbanization (NOAA, 2015). Methane, the primary constituent of natural gas, accounts for 10% of all U.S. greenhouse gas (GHG) emissions, approximately 30% of which are attributable to natural gas and petroleum systems (U.S. Environmental Protection Agency, 2015). Methane is a potent GHG whose global warming potential is 34 and 86 times greater than carbon dioxide (CO₂) over 100 and 20-year time horizons, respectively (IPCC, 2013). In terms of anthropogenic CH₄ emissions by source, emissions from natural gas systems are the highest (U.S. Environmental Protection Agency, 2015). As the U.S. shifts away from oil and coal, production of natural gas from shale gas reserves has increased by 35% from 2005 to 2013 (U.S. Energy Information Administration, 2015). Elucidating CH₄ emissions from natural gas systems will facilitate responsible management in keeping with national GHG mitigation goals (U.S. Global Change Research Program, 2014).

With the recent increase in hydraulic fracturing and horizontal drilling, carbon emissions associated with the upstream, midstream, and downstream sectors of the natural gas industry have become the subject of growing research interest (Alvarez et al., 2012; Miller et al., 2013; Brandt et al., 2014). Fugitive CH₄ emissions, attributed to venting or leakage across the life cycle of natural gas, make the climate benefits ascribed to natural gas questionable when compared to oil and coal. A majority of research to date has sought to constrain estimates of upstream and midstream fugitive CH₄ emissions (Allen et al., 2013, 2015; Brantley et al., 2014; Mitchell et al., 2015; Subramanian et al., 2015). However, downstream emissions associated with the processing and



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distribution of natural gas remain poorly characterized. Given the strain that increased production and consumption of natural gas places on aged U.S. distribution infrastructure (American Society of Civil Engineers, 2013; U.S. Department of Energy, 2015), this study assesses the impact of fugitive CH₄ emissions associated with leak-prone distribution infrastructure in urban environments.

Leak-prone distribution infrastructure is composed of outdated pipe material such as cast iron, wrought iron, and unprotected steel. often dating back to the mid 1800s and early 1900s (U.S. Environmental Protection Agency and Gas Research Institute, 1996). Iron mains make up 2.4% of the natural gas distribution system in the U.S. (PHMSA, 2015) yet contribute a majority of total pipeline emissions (U.S. Environmental Protection Agency and Gas Research Institute, 1996; Lamb et al., 2015). Leak-prone mains constitute up to 34% of natural gas distribution infrastructure in Eastern U.S. states (PHMSA, 2015). Urban mapping studies reveal that densely populated Eastern U.S. cities have thousands of natural gas leaks (Phillips et al., 2013; Jackson et al., 2014; Environmental Defense Fund and Google Earth Outreach, 2015; Gallagher et al., 2015). Despite progress made towards leak identification and mapping, quantification of fugitive CH₄ emissions from leak-prone distribution infrastructure remains poorly characterized. Bottomup approaches are limited by small sample sizes (U.S. Environmental Protection Agency and Gas Research Institute, 1996; Lamb et al., 2015), while top-down approaches (Townsend-Small et al., 2012; McKain et al., 2015) are not designed to resolve point source attribution.

Further, very little is known about the nature of the statistical distribution of sizes of gas leaks in distribution pipeline systems in terms of CH₄ flux. Current industry practice is to use emissions factors that carry an implicit assumption of an average leak size based on a normal distribution (U.S. Environmental Protection Agency and Gas Research Institute, 1996). However, results from midstream and upstream studies increasingly show evidence for a skewed distribution of leak size (Brandt et al., 2014; Brantley et al., 2014; Allen et al., 2015; Mitchell et al., 2015; Subramanian et al., 2015). There also remains a lack of consensus regarding the volume of fugitive CH₄ emissions lost from leak-prone distribution infrastructure, the frequency of leaks per road mile, and the severity of the safety hazard posed by potentially explosive (Grade 1) natural gas leaks in urban environments.

In this study we made direct measurements of CH₄ emissions from 100 natural gas leaks in cast iron distribution mains within Metro Boston, MA in order to assess the nature of the distribution of gas leak sizes, in particular whether they are characterized by a normal or skewed distribution. We took flux chamber measurements at individual leak sites to constrain estimates of fugitive CH₄ emissions from leak-prone distribution infrastructure. We resampled a subset of these leaks in summer and winter to evaluate seasonal variation in CH₄ flux. We assessed the hazard potential of each leak surveyed, reporting those that qualified as Grade 1 to local utility companies. These results can be used to prioritize pipeline repair and replacement, stem GHG emissions, safeguard against pipeline explosions, and efficiently distribute and consume natural gas.

2. Materials and methods

To estimate CH₄ emissions from leak-prone natural gas distribution infrastructure we made direct measures of 100 natural gas leaks in cast iron distribution mains within Metro Boston, MA [Table S1; see Field Sampling section of Supplementary Materials (SM) for details]. We selected sampling sites based on three criteria: 1) cast iron pipe material, 2) a proportion of pipeline operating pressures representative of the total distribution

network, and 3) detection of elevated atmospheric [CH₄] (Fig. S1). We obtained the location, age, operating pressure, and diameter of buried cast iron mains from natural gas distribution infrastructure maps provided by National Grid (2013). We identified 45 natural gas leaks using the results of our 2011 on-road atmospheric CH₄ survey (Phillips et al., 2013) and an additional 55 leaks in Boston, Brookline, and Newton through real-time on-road atmospheric CH₄ surveys following the same methodology. We checked the calibration on the mobile Picarro G2301 Cavity Ring-Down Spectrometer (Picarro, Inc., Santa Clara, CA) with 0 and 5 ppm CH₄ test gas (Balance: air; Spec Air Specialty Gases, Auburn, ME; reported precision $\pm 10\%$) periodically throughout our sampling campaign. We sampled leaks over cast iron distribution mains operating at 0.5 (n = 93), 2 (n = 3), 22 (n = 3), and 60 (n = 1) pounds per square inch gage (PSIG; see Pipeline Operating Pressure section of SM for details).

We defined a leak as any detected atmospheric [CH₄] above a threshold of 2.5 ppm, consistent with Phillips et al. (2013), Jackson et al. (2014), and Gallagher et al. (2015). We further defined a leak as 1) at least 3.7 m (12 ft) in distance from adjacent leaks emanating from the same distribution main; 2) spatially distinct from leaks in parallel distribution mains; 3) spatially distinct from leaks in associated service lines; and 4) attributable to natural gas due to a recognizable odor of mercaptan. Distribution main segments are 3.7 m in length, attached by joints at either end (U.S. Environmental Protection Agency and Gas Research Institute, 1996). Applying a horizontal 3.7 m buffer reduces the risk of double counting leaks on the same distribution main. We avoided double counting leaks from parallel distribution mains running under the same street by excluding leaks that we could not confidently assign to one main or the other. Natural gas leaks also arise from service lines, which attach directly to the distribution main. We similarly excluded leaks that we could not confidently assign to either the distribution main or the service line.

We surveyed leaks in June and December of 2012, September and November of 2013, January of 2014, and June-September of 2014. We used a flame ionization unit (FIU; Dafarol A500 Flame Ionization Unit, Dafarol Inc., Hopedale, MA) to determine the spatial extent of each leak and the location of individual gas escape points within a sampling site (e.g., manhole, utility access point, road or sidewalk crack, curb, tree well, urban lawn, roadway drill hole). We checked the calibration on the FIU daily using 50 ppm CH₄ test gas (Balance: air; Spec Air Specialty Gases, Auburn, ME; reported precision $\pm 5\%$). After taking flux chamber measurements at all gas escape points we used a combustible gas indicator (CGI; Gas Sentry[®], model CGI-201, Bascom-Turner Instruments, Inc., Norwood, MA) to measure [CH₄] in soil gas and in the headspace of voids under manholes, gas and water valve boxes, electrical access points, and storm water drains. The CGI was calibrated every 30 days with 2.5% CH₄ test gas (MC-105 Methane & CO Calibration Gas; Bascom-Turner Instruments, Inc., Norwood, MA; reported precision $\pm 2\%$). We reported all leaks that qualified as potentially explosive (Grade 1) to local utility providers.

The Pipeline and Hazardous Materials Safety Administration (PHMSA, 2015), a U.S. Department of Transportation agency, classifies natural gas leaks into three grades, Grade 1 through 3 with Grade 1 being the most dangerous, based on their proximity to persons and property and the concentration of CH_4 gas detected in nearby air samples (Table S4; see Leak Grading section of SM for details). The lower explosive limit (LEL) and upper explosive limit (UEL) for natural gas in air are five and 15%, respectively. Natural gas is flammable at 5–15% in open air and explosive at 5–15% when found in a confined space. If a natural gas leak is proximate to people or property, where gas may accumulate to explosive levels (80% LEL) in confined spaces or migrate inside or around buildings,

it is considered unsafe. Alternatively, a natural gas leak that occurs in a well-ventilated area removed from people and high-value property is considered relatively low risk. Here, we follow the leak classification standards published by PHMSA and classify leaks as Grade 1 if we detected \geq 4% natural gas in the air sampled from confined, person-sized spaces (e.g. manholes), or 80% LEL (Table 1; Table S4; PHMSA, 2002). We also classify leaks as Grade 1 if we detected any gas within 1.5 m (5 ft) or less of a building (Table 1; Table S4; PHMSA, 2002).

We made direct measures of CH₄ efflux from gas leaks using a chamber-based method (see Chamber Measurements section of SM for details). Natural gas is lighter than air and migrates up and away from the leak origin (Okamoto and Gomi, 2011). As distribution pipes are buried under impervious surface (i.e., roads and sidewalks), leaked natural gas migrates underground along paths of least resistance for escape. We designed four chambers (55.6, 17.2, 16.1, and 14.0 L) to quantify CH₄ emissions escaping from manholes, utility access structures, curbs, soil, and cracks in asphalt and cement (Fig. S2; Table S2; Table S3). Two Swagelok-fitted vent holes located at the top of each chamber facilitated gas sampling from the chamber headspace via ¼ in plastic tubing. We fit a third vent hole with a 'pigtail' extension to reduce pressure anomalies resulting from wind turbulence (Bain et al., 2005). We equipped the chambers with plastic skirts, which were weighted down with gravelfilled burlap tubes to create a seal with the sampling surface. To ensure that the sample air was well mixed, we placed batteryoperated fans inside each chamber. We fit a simple linear regression to plotted chamber data and used the slope of this line ($[CH_{A}]$ • sec^{-1}) to approximate CH₄ flux at gas escape points.

We utilized a closed dynamic chamber method (Bain et al., 2005) for quantifying CH₄ emissions from relatively low flux gas escape points (where flux was \leq 96 g CH₄•day⁻¹). Of 535 individual chamber measurements made over the course of this study, 26% employed this chamber methodology (capturing 11% of all CH₄ emissions sampled). For these measurements, we used a Picarro G2301 Cavity Ring-Down Spectrometer to collect CH₄ flux data. As this analyzer resolves [CH₄] the nearest parts per billion and has an upper [CH₄] limit of ~40 ppm, it is particularly well suited for quantifying CH₄ emissions from relatively low flux gas escape points.

We utilized a modified closed dynamic chamber method (Bain et al., 2005) for quantifying CH₄ emissions from relatively high flux gas escape points (where flux was $\leq 1.6 \times 10^4$ g CH₄•day⁻¹). A majority (74%) of the chamber measurements made during this study employed this technique (capturing 89% of all CH₄ emissions sampled). For these measurements, we used a CGI to collect CH₄ flux data. This analyzer is less precise than the Picarro G2301 Cavity Ring-Down Spectrometer, but it is capable of measuring up to 100% CH₄ gas. The CGI collects [CH₄] data at 0.01% gas intervals (100 ppm), making it particularly well suited for quantifying CH₄ emissions from relatively high flux gas escape points.

3. Results and discussion

3.1. Leak size is skewed

Direct measures of CH₄ flux from 100 natural gas leaks originating from cast iron distribution infrastructure in Metro Boston,

Table 1

We report the conditions found at fifteen sites that qualify for Grade 1 leak classification that were identified during leak surveys of cast iron distribution mains in Boston, Brookline, and Newton, MA in column one (Table S4; PHMSA, 2002). The Environmental Defense Fund and Google Earth Outreach also identified fourteen of the same leaks during street mapping in March and June of 2013, but deemed all of them nonhazardous (Roston, 2014; Wong, 2014). The Environmental Defense Fund and Google Earth Outreach assessed leaks according to low (700–9,000 L CH₄•day⁻¹), medium (9,000–60,000 L CH₄•day⁻¹), or high (>60,000 L CH₄•day⁻¹) estimated CH₄ flux, reported in column two (Environmental Defense Fund and Google Earth Outreach, 2015).

| Leak assessment | | Leak location | |
|---|--|---|---------------|
| Boston university | Environmental defense fund and google earth outreach | Block address | Lat long |
| 9% CH4 in manhole | Low | 400 block Dudley St., Boston, MA 02119 | 42°19′30.94″N |
| | | | 71°04′30.09″W |
| >30% CH ₄ in three manholes | Medium | 800 block Centre St., Boston, MA 02130 | 42°18′33.24″N |
| | | | 71°07′13.78″W |
| 13% CH ₄ in manhole | Low | 4100 block Washington St., Boston, MA 02131 | |
| | | | 71°07′32.93″W |
| 6% CH ₄ in manhole | Low | 1900 block Columbus Ave., Boston, MA 02119 | 42°18′54.76″N |
| | | | 71°06′00.47″W |
| 7% CH ₄ in manhole | Low | 4300 block Washington St., Boston, MA 02131 | 42°17′04.73″N |
| | | | 71°07′50.28″W |
| 8% CH ₄ in manhole | Medium | 400 block Hyde Park Ave., Boston, MA 02131 | 42°17′12.70″N |
| | | | 71°07′07.29″W |
| 5% CH4 in manhole | Not detected | 100 block Mt. Pleasant Ave., Boston, MA | 42°19′36.37″N |
| | | 02119 | 71°04′46.92″W |
| 42% CH ₄ in soil \leq 1.5 m from build | ing Low | 600 block Centre St., Boston, MA 02130 | 42°19′15.37″N |
| | | | 71°07′21.04″W |
| 6% CH ₄ in manhole | Low | 1900 block Dorchester Ave., Boston, MA | 42°16′58.95″N |
| | | 02124 | 71°03′54.21″W |
| 25% CH ₄ in soil \leq 1.5 m from build | ing Low | 1900 block Dorchester Ave., Boston, MA | 42°16′59.52″N |
| | | 02124 | 71°03′53.54″W |
| 25% CH ₄ in soil \leq 1.5 m from build | ing Low | 0 block Lanark Rd., Boston, MA 02135 | 42°20′23.64″N |
| | | | 71°08′45.28″W |
| 6% CH ₄ in manhole | Low | 100 block Lochstead Ave., Boston, MA 02130 | |
| | | | 71°06′54.26″W |
| 17% CH ₄ in manhole | Low | 100 block Tappan St., Brookline, MA 02445 | 42°20′00.50″N |
| | | | 71°08′03.17″W |
| 66% CH ₄ in manhole | Low | 100 block Claflin Rd., Brookline, MA 02445 | 42°20′14.35″N |
| | | | 71°08′10.43″W |
| 5% CH ₄ in manhole | Medium | 200 block Hammond St., Newton, MA 02467 | 42°19′51.61″N |
| | | | 71°10′12.15″W |

MA ranged from $4.0 - 2.3 \times 10^4$ g CH₄•day⁻¹ (Table S1). The distribution of leak size is positively skewed, with a long right-hand tail anchored by a few superemitter leaks that contribute a large proportion of fugitive CH₄ emissions (Fig. 1). The left-hand mass of the distribution is composed of many small flux leaks. The lognormal mean leak rate is 1.2×10^3 g CH₄•day⁻¹ for all leaks surveyed. We found no significant difference in CH₄ flux between leaks sampled in the winter versus summer seasons (n = 13, p = 0.56).

A positively skewed distribution of leak size across leak-prone distribution infrastructure in Metro Boston, MA is inconsistent with earlier work that implicitly assumes a normal distribution of leak size (U.S. Environmental Protection Agency and Gas Research Institute, 1996), but consistent with the distribution of direct measures of natural gas leaks made by Lamb et al. (2015) within local distribution systems in the U.S. The emission factor reported for cast iron mains by Lamb et al. (2015; 1.3×10^3 g CH₄•day⁻¹) is also consistent with the log-normal mean leak rate that we report for our survey. A positive skew in fugitive CH₄ emissions across leak-prone distribution infrastructure, with many small leaks and few superemitter leaks, is also analogous to findings on CH₄ leakage from natural gas industry (Brandt et al., 2014; Brantley et al., 2014; Allen et al., 2015; Mitchell et al., 2015; Subramanian et al., 2015).

Our survey makes progress on improving our understanding of the distribution of leak size across leak-prone natural gas distribution infrastructure by virtue of its enhanced sample size of 100. Previous research by the U.S. Environmental Protection Agency and Gas Research Institute (1996) and Lamb et al. (2015) reported on 21 and 14 leaks, respectively, across cast iron distribution mains. A larger sample size increases the likelihood of capturing the furthest extent of a positively skewed leak distribution with bottom-up sampling approaches. Lamb et al. (2015) report a 95% upper confidence limit of 4.8×10^3 g CH₄•day⁻¹ for an emission factor for cast iron distribution mains, while the largest leak surveyed in Metro Boston, MA was 2.3×10^4 g CH₄•day⁻¹. Notably, our reported natural gas leakage rates are likely an underestimate of actual leakage rates across natural gas distribution pipelines as subsurface leaks from distribution mains in urban environments are highly complex. The heterogeneous patchwork of pervious and impervious surfaces and the abundance of buried, collocated nongas utility structures make it unlikely that we have captured all natural gas emissions during our sampling events.

3.2. Top 7% of leaks contribute 50% of total CH₄ emissions

We found that seven superemitter leaks contributed 50% of all fugitive CH₄ emissions captured in this survey. Of these superemitter leaks, five were sampled over mains operating at 0.5 PSIG, including the largest leak surveyed. The two remaining superemitter leaks were sampled over mains operating at 22 and 60 PSIG, respectively. Mean CH₄ flux appeared to correlate positively with pipeline operating pressure ($R^2 = 0.85$, p = 0.03, n = 5). Nevertheless, leak size data remain skewed even when leaks sampled over mains operating at pressures greater than 0.5 PSIG are excluded ($\mu = 5.4$, $\sigma = 1.8$, log-normal mean = 1.0×10^3 g $CH_4 \bullet day^{-1}$, n = 92, Pearson's coefficient of skewness for flux data = 8.0). Further, the distribution of leak size remains skewed even when all superemitter leaks are excluded (Fig. 1). Resolving the relationship between leak size and pipeline operating pressure remains an open area of research for future studies of natural gas distribution systems.

The positively skewed distribution of leak size across aged distribution infrastructure has important policy implications. Fixing superemitter leaks will stem a large fraction of fugitive CH₄ emissions from natural gas infrastructure that are known to contribute to the GHG profile of urban centers (Brandt et al., 2014; McKain et al., 2015). Many cities, including Boston, MA, do not currently factor in CH₄ emissions from natural gas systems when accounting for citywide GHG emissions, or when setting specific GHG reduction goals (City of Boston, 2014). However, awareness of the issue is growing, in part due to research published on the topic. Understanding how leak size is distributed allows urban stakeholders to prioritize leak repair towards meeting climate change goals, improving efficiency in urban energy systems, and reducing utility rate inflation associated with lost and unaccounted for (LAUF) gas. Top-down measurements of fugitive CH₄ emissions in the

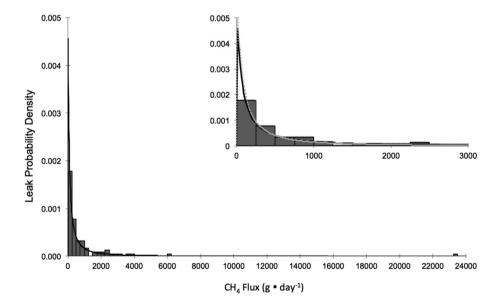


Fig. 1. The distribution of leak size is skewed (n = 99; Pearson's coefficient of skewness for flux data = 7.5). The black line represents a fitted, log-normal distribution (main and inset plot; $\mu = 5.4$, $\sigma = 1.8$, log-normal mean = 1.2×10^3 g CH₄•day⁻¹). The distribution of leak size is skewed even when superemitter leaks are excluded (grey dotted line in inset plot represents all leaks excluding the top 7% that contribute 50% of total CH₄ emissions; $\mu = 5.2$, $\sigma = 1.7$, log-normal mean = 7.4×10^2 g CH₄•day⁻¹, n = 92, Pearson's coefficient of skewness for flux data = 1.8). See Leak Size Distribution section of SM for details.

Boston urban region estimate that the average annual loss rate from all downstream components of the natural gas system is 2.7%, or roughly \$90 million worth of natural gas fuel (McKain et al., 2015). Addressing superemitter leaks is an effective way to revitalize aged infrastructure while still meeting energy needs and adhering to GHG reduction targets. To the extent that the cost of both LAUF gas and pipeline repair are folded into natural gas utility rates, as they currently are in MA, fixing superemitter leaks will benefit consumers by reducing LAUF gas through relatively high benefit-tocost pipeline repair projects.

3.3. Flux is not an indicator of safety

Of the 100 natural gas leaks surveyed, 15% qualified as potentially explosive (Grade 1; Table 1). Notably, we found no significant difference in CH₄ flux between Grade 1 leaks and all remaining leaks surveyed based on the result of a two-tailed, heteroscedastic *t*-test. Here, we compare CH₄ flux from fifteen Grade 1 leaks to all remaining leaks surveyed (n = 85) and find that CH₄ flux is not significantly different between the two sample populations (p = 0.24).

Further, we found 10 cases of small leaks $(<1.2 \times 10^3 \text{ g} \text{ CH}_4 \cdot \text{day}^{-1})$ that qualified as potentially explosive (Grade 1). As small leaks have the potential to be hazardous, CH₄ flux is not an indicator of safety. Addressing superemitter leaks will stem GHG emissions, but all leaks must be assessed as small leaks cannot be disregarded as 'safely leaking.' This result has important implications for human health and safety, as well as for the future of leak detection and classification.

While good progress has been made towards revitalizing aged infrastructure, with 15% of remaining leak-prone distribution mains replaced in the U.S. since 2010, local utilities still rely on 29,359 miles of cast and wrought iron mains to distribute natural gas to consumers (PHMSA, 2015). As these pipes continue to age, the U.S. sees an average of 110 gas distribution pipeline incidents per year (2010-2014; PHMSA, 2015). Significant distribution pipeline incidents are characterized by a fatality or injury requiring in-patient hospitalization, or causing \$50,000 or more in total costs (PHMSA, 2015). In 2014 alone, there were 113 gas distribution pipeline incidents reported across the U.S., with 18 fatalities, 94 injuries, and almost \$75 M in property damage (PHMSA, 2015), exceeding the national 5-year average (2010-2014) reported for all categories. Since 2010, there have been 23 gas distribution pipeline incidents reported across Massachusetts, with one fatality, 18 injuries, and almost \$6 M in property damage (PHMSA, 2015). Most recently, a house explosion caused by a natural gas leak in Dorchester, MA on April 16, 2014 injured 12 people and destroyed a two-and-a-half story residence in an ensuing three-alarm fire. Although costly, our results indicate that reducing pipeline incidents requires fully revitalizing leak-prone distribution infrastructure and improving leak detection and monitoring.

Utility companies currently detect natural gas leaks following similar on-road driving surveys of elevated atmospheric [CH₄] as those employed by Phillips et al. (2013), Jackson et al. (2014), and Gallagher et al. (2015), yet do not employ additional equipment to measure meteorological conditions (e.g. wind speed and direction, boundary layer stability, mixing layer height, atmospheric pressure). Utilities also perform walking surveys of mains and service lines, although at less frequent intervals relative to driving surveys, and rely on the public to directly report suspected natural gas leaks in their vicinity in real-time. Grade 1 leaks detected by or reported to natural gas utility companies are currently prioritized for expedited repair (Table S4; PHMSA, 2002). As utility companies can only repair a finite number of leaks per year, a surplus of Grade 2 and predominantly Grade 3 leaks are monitored less frequently. Grade 2

and Grade 3 leaks are classified as non-hazardous at the time of detection and do not require reevaluation for another six and 15 months following the time of detection, respectively (Table S4; PHMSA, 2002). This leak management model is worrisome because there is no evidence to support a correlation between on-road atmospheric [CH₄] readings and CH₄ flux at leak sites (Table 1, Fig. S4), CH₄ flux is not an indicator of leak safety, and lesser leaks can quickly transform into Grade 1 leaks via mechanical disruption or as the result of frost heaves associated with prevalent winter freeze—thaw cycles in Northeastern states. A lesser gas leak may also be upgraded to a Grade 1 leak if 1) existing corrosion intensifies, leading to an increase in CH₄ flux; 2) operating pressure is increased, leading to an increase in CH₄ flux; 3) natural gas begins, or continues, to accumulate in a closed space to 80% LEL; and 4) natural gas begins, or continues, to spread into or around buildings.

3.4. Universal leak detection and classification methodology is required

There is currently no universal definition of what constitutes a natural gas leak, no universal leak detection methodology, and no universal standard for how leaks are classified according to severity. This lack of agreement amongst stakeholders poses a problem as urban environments are heterogeneous and natural gas leaks can be complex. For example, what is the relationship between individual gas escape points at the road's surface and the number of fissure points in the underlying pipeline? Leaks are now detected using on-road driving or walking surveys of atmospheric $[CH_4]$, but academic (Phillips et al., 2013; Jackson et al., 2014; Gallagher et al., 2015), utility (PHMSA, 2002), and environmental advocacy groups (Environmental Defense Fund and Google Earth Outreach, 2015) utilize different leak detection instruments and employ different leak detection methods. Leaks are also classified in a variety of ways, including via direct measurements of CH₄ flux (Lamb et al., 2015), estimated CH₄ flux based on 'controlled releases' (Environmental Defense Fund and Google Earth Outreach, 2015), and a combination of FIU and CGI readings (PHMSA, 2002).

The lack of universal leak detection and classification methodology amongst natural gas stakeholders limits our understanding of the magnitude of safety and climate concerns associated with aged natural gas distribution infrastructure. Current estimates of the total number of leaks within a particular region, or the frequency of leaks per road mile or pipe mile within that region, are ambiguous without universal leak detection and reporting criteria (Table 2). Further, the results of this study indicate that on-road driving surveys are not sufficient to classify leak severity. There is no reliable evidence to indicate that atmospheric [CH₄] correlate to CH₄ flux at a leak site (Fig. S4; see On-Road Driving Surveys vs. Flux Measurements section of SM for details) and CH₄ flux itself is not a reliable indicator of leak safety (Table 1). While mobile CH₄ surveys provide excellent information towards leak detection and location, small leaks may still go undetected during mobile surveys and all leaks require additional FIU and CGI readings to determine safety classifications (Table 1).

This issue is as relevant for aged natural gas distribution infrastructure as it is for relatively 'young' natural gas systems. Currently 38% of U.S. natural gas distribution mains are composed of protected steel and 55% are composed of plastic (PHMSA, 2015). Even though protected steel and plastic pipes are not considered leakprone, proposed increases in operating pressure associated with increased supply and demand for natural gas fuels does place strain on even the most robust distribution systems. We report that mean CH₄ flux appears to correlate positively with pipeline operating pressure, suggesting that resolving the relationship between leak size and pipeline operating pressure is a vital next step for future

| Table 2 | |
|---|--|
| Street leak frequency reported for Boston, MA since 2005. | |

| Leaks/Road mile | Pipe material | Survey year | Data source |
|-----------------|---------------|-------------|---|
| 2.7 | All Materials | 2005 | Keyspan Corporation (now National Grid), 2005 |
| 4.3 | All Materials | 2011 | Phillips et al., 2013 |
| 1.0 | All Materials | 2013 | Environmental Defense Fund and Google Earth Outreach 2015 |
| 2.2 | All Materials | 2014 | National Grid, 2015 |

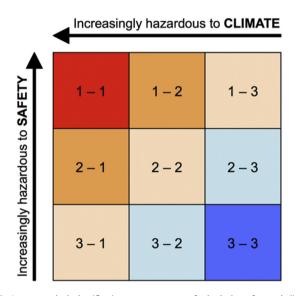


Fig. 2. A two-part leak classification system accounts for both the safety and climatic impacts of natural gas leaks (safety–climate). Warm colors indicate hazardous leaks with increasing explosive potential and large volumes of LAUF gas lost. Cool colors indicate non-hazardous leaks with a reduced safety risk and limited LAUF gas lost.

studies of natural gas distribution systems. Improving distribution pipeline safety and mitigating associated greenhouse gas emissions, regardless of the age of the distribution network, requires that U.S. regulators mandate that all public utilities companies adopt 1) a universal definition of what constitutes a natural gas leak; 2) universal leak detection methodology that employs both driving and walking surveys in order to detect and assess leaks of all sizes; 3) universal standards for how leaks are classified; and 4) universal action criteria for how leaks are addressed within appropriate timelines.

4. Conclusions

We report on a survey of CH_4 emissions from 100 natural gas leaks in cast iron distribution mains in Metro Boston, MA. This study has three results: 1) the distribution of leak size is skewed, 2) a small fraction of leaks contribute disproportionate CH_4 emissions, and 3) CH_4 flux at leak sites is not an indicator of safety. Key methodological impediments to quantifying and addressing this problem involve inconsistencies in the manner in which gas leaks are defined, detected, and classified. While leak definition and detection are beyond the scope of this research, here we propose one key advance in leak classification.

Natural gas leaks are now classified according to a three-tiered system that reflects explosive potential, with Grade 1 leaks posing the most serious threat to life and property and Grade 3 leaks posing no threat at the time of detection. Missing from this classification system is an assessment of the climatic and monetary consequences of LAUF gas. To address this need, we propose a two-part leak classification system that better reflects the full impacts of

natural gas leaks (Fig. 2). This classification system accounts for both the explosive potential (1 to 3, most to least dangerous) and climatic consequence (1 to 3, most to least LAUF gas lost) of natural gas leaks. For example, a Grade '3–1' leak is non-hazardous to life and property but emits large quantities of LAUF gas, while a Grade '3–3' leak is non-hazardous to life and property and emits little LAUF gas. With regard to a Grade '3–1' leak, the current leak classification system misses what may be called the 'Climatic Grade 1' designation of an otherwise non-hazardous Grade 3 leak. We propose improvements to leak classification in part to also encourage similar progress towards development of a universal leak definition and universal leak detection methodology.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.01.094.

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