

Natural Gas Pipeline Leaks Across Washington, DC

Robert B. Jackson,^{†,‡,*} Adrian Down,[†] Nathan G. Phillips,[§] Robert C. Ackley,^{||} Charles W. Cook,[†] Desiree L. Plata,[⊥] and Kaiguang Zhao[†]

[†]Duke University, Nicholas School of the Environment and Center on Global Change, Durham, North Carolina 27708 United States

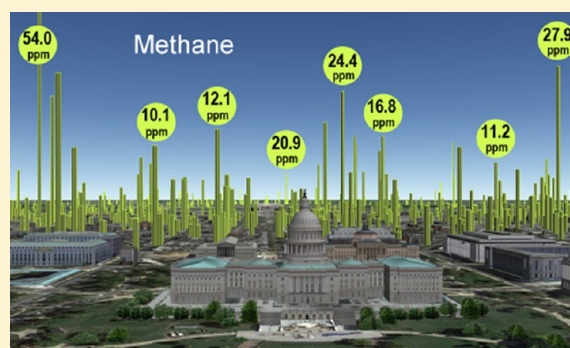
[‡]Stanford University, School of Earth Sciences, Stanford, California 94305 United States

[§]Boston University, Department of Earth and Environment, 675 Commonwealth Avenue, Boston Massachusetts 02215 United States

^{||}Gas Safety, Inc., Southborough, Massachusetts 01772 United States

[⊥]Duke University, Pratt School of Engineering, Durham, North Carolina 27708 United States

ABSTRACT: Pipeline safety in the United States has increased in recent decades, but incidents involving natural gas pipelines still cause an average of 17 fatalities and \$133 M in property damage annually. Natural gas leaks are also the largest anthropogenic source of the greenhouse gas methane (CH₄) in the U.S. To reduce pipeline leakage and increase consumer safety, we deployed a Picarro G2301 Cavity Ring-Down Spectrometer in a car, mapping 5893 natural gas leaks (2.5 to 88.6 ppm CH₄) across 1500 road miles of Washington, DC. The $\delta^{13}\text{C}$ -isotopic signatures of the methane ($-38.2\text{‰} \pm 3.9\%$ s.d.) and ethane (-36.5 ± 1.1 s.d.) and the CH₄:C₂H₆ ratios (25.5 ± 8.9 s.d.) closely matched the pipeline gas (-39.0‰ and -36.2‰ for methane and ethane; 19.0 for CH₄/C₂H₆). Emissions from four street leaks ranged from 9200 to 38 200 L CH₄ day⁻¹ each, comparable to natural gas used by 1.7 to 7.0 homes, respectively. At 19 tested locations, 12 potentially explosive (Grade 1) methane concentrations of 50 000 to 500 000 ppm were detected in manholes. Financial incentives and targeted programs among companies, public utility commissions, and scientists to reduce leaks and replace old cast-iron pipes will improve consumer safety and air quality, save money, and lower greenhouse gas emissions.



INTRODUCTION

Fugitive emissions of methane (CH₄) from oil and gas extraction and pipeline transmission are the largest anthropogenic source of methane in the United States and the second largest source globally.¹ Methane is a far more potent greenhouse gas (GHG), molecule for molecule, than CO₂,^{2–6} with an estimated global warming potential 86 times greater than CO₂ on a 20-year basis and 34 times greater on a 100-year basis.⁷

Considerable controversy surrounds the estimates of methane emissions from the extraction, distribution, and consumption of natural gas.^{3,4,8–11} In 2009, the U.S. Environmental Protection Agency (EPA) estimated that 2.4% (151 Tg CO₂-equivalents of CH₄) of total natural gas production in the United States leaked or was released to the atmosphere, with the estimate for 2011 being slightly smaller (145 Tg CO₂-equivalents).¹ New ground-based measurements from 150 natural gas production sites across the U.S. suggested that methane emissions during production are only 0.42% of gross gas production.¹² Estimates of methane losses during the production and transmission of natural gas in Russia were also relatively low, and comparable to U.S. networks.^{13,14}

In contrast to these lower estimates, new top-down measurements from the air over oil and gas fields in Colorado, Utah, and California suggest that leakage rates there may be

substantially higher than previously suggested.^{5,15–17} In the Uintah Basin of Utah, for instance, Karion et al.¹⁷ estimated that CH₄ emissions from a natural gas and oil production field were $55\,000 \pm 15\,000$ kg hr⁻¹, a rate corresponding to 6.2%–11.7% of total natural gas production in the sampling region. Peischl et al.¹⁶ ascribed 31.9 ± 6.5 Gg/yr of CH₄ emissions to leaks of local, unprocessed natural gas in the greater Los Angeles (LA) basin, an amount that is $\sim 17\%$ of local production; however, such estimates are especially complicated in LA because of the presence of natural geological seeps and extensive natural gas pipelines.

Methane losses from the pipelines that distribute natural gas are also uncertain.¹⁴ Decades ago, Mitchell et al.¹⁸ estimated that natural gas leakage from the natural gas distribution system in the United Kingdom in 1990 was between 1.9% and 10.8% (median estimate of 5.3%), compared to the estimate of 1% supplied by British Gas at the time. Lelieveld et al.¹⁹ estimated that methane losses from natural gas transport systems in Russia were much smaller than the British estimate, $\sim 1.4\%$, with a range of 1.0–2.5%. Recently, Phillips et al.²⁰ published

Received: October 8, 2013

Revised: December 6, 2013

Accepted: December 31, 2013

Published: January 16, 2014

the first comprehensive map of natural gas leaks for a city, documenting ~3400 pipeline leaks across the 785 road miles (1263 km) of Boston. The leaks had $\delta^{13}\text{CH}_4$ signatures consistent with the fossil-fuel gas from the pipelines rather than with a biogenic source of methane.²⁰

Along with reducing greenhouse gas emissions, repairing production and pipeline leaks would improve consumer health and safety and save money. Hydrocarbons lost to the atmosphere can react with NO_x to catalyze ozone formation in urban areas.²¹ Furthermore, incidents involving natural gas transmission and distribution pipelines in the U.S. cause an average of 17 fatalities, 68 injuries, and \$133 M in property damage annually.²²

In this study, we measured methane concentrations along each of the 1500 road miles (2,415 km) of Washington, DC to assess the frequency and extent of leakage from natural gas pipelines. To confirm the source of the natural gas, we measured the carbon isotope composition ($\delta^{13}\text{CH}_4$ and $\delta^{13}\text{C}_2\text{H}_6$) of leaks and the abundance of ethane (C_2H_6) and propane (C_3H_8), two gases that have no biological sources in this environment and that are not generated in wetlands or sewer systems.^{20,23} We also measured methane emission rates from street leaks and quantified the concentrations of methane in manholes associated with 19 leaks, determining the number with potentially explosive (Grade 1) concentrations. Finally, we analyzed data for national lost-and-unaccounted-for gas and cast-iron pipelines to place our observations in a broader scientific context. Such studies can help increase consumer safety and reduce fugitive methane emissions associated with the use of natural gas.

MATERIALS AND METHODS

To estimate the extent of pipeline leaks across Washington, DC, we surveyed all 1500 road miles for methane concentrations in January and February of 2013 using a mobile Picarro G2301 Cavity Ring-Down Spectrometer equipped with an A0491 Mobile Plume Mapping Kit (Picarro, Inc., Santa Clara, CA). Our methods for the street mapping were similar to those described in Phillips et al.²⁰ Briefly, data obtained from the Picarro spectrometer sampled at ~0.5-m height above the road surface and mobile GPS unit (Hemisphere GNSS, Scottsdale, AZ; Model MD R100) were recorded every 1.1 s (0.91 Hz). We adjusted the time stamp on the $[\text{CH}_4]$ readings based on a 1-s delay observed between analyzer response to a CH_4 source injected into the instrument while driving and the apparent GPS location. We also used an auxiliary pump to increase tubing flow throughput to within 5 cm of the analyzer inlet to correct for a short time lag between instantaneous GPS location and a delay in $[\text{CH}_4]$ measurement due to inlet tube length (~3 m). Three sets of measurements immediately before, during, and after the campaign using standards of 0, 5, and 50 ppm CH_4 air yielded $r^2 > 0.99$ and a mean slope of 0.92 for the Picarro (i.e., our measurements of methane in air slightly underestimated actual values).

We defined a “leak” or other source as a separate, spatially contiguous set of $[\text{CH}_4]$ observations exceeding a concentration threshold of 2.50 ppm at >5-m spacing. This threshold was used by Phillips et al.²⁰ and corresponds to the 90th percentile of the distribution of data from all road miles driven. The background air in Washington, DC varied between 1.9 and 2.0 ppm. Relative to the global background, the 2.5-ppm threshold is ~37% above 2012 mean mixing ratios observed at Mauna Loa (~1.82 ppm).²⁴ The precise concentration

measured for any given leak depends on both wind dispersion and the leak rate.

To examine the source of the methane, we measured methane, ethane, and propane concentrations, as well as the isotopic signatures of methane and ethane ($\delta^{13}\text{CH}_4$ and $\delta^{13}\text{C}_2\text{H}_6$) in the air from 19 street leaks across the city. After mapping the leaks using the mobile methane analyzer, we first identified the location of the leak and the concentration of gases leaking directly from it (e.g., a curb or manhole). We then collected samples of each leak in 150-mL stainless steel canisters (Swagelok, Solon OH) and 1-L Tedlar bags with valve and septa fittings (Environmental Supply, Inc., Durham NC) in January and February 2013. Pipeline gas was collected in February 2013 at George Washington University. A Gas Sentry CGO-321 hand-held gas detector (Bascom-Turner, Norwood, MA) was first used to identify the area of highest ambient $[\text{CH}_4]$ at each leak location sampled for $\delta^{13}\text{CH}_4$ and, separately, to measure CH_4 concentrations of air in the manholes sampled at 19 locations. Evacuated sample bags were then filled at each leak location using a hand pump. The samples were analyzed for $\delta^{13}\text{CH}_4$, usually within two days and always within two weeks, using a Picarro G2112i Cavity Ring-Down Spectrometer²⁵ (CRDS) at the Duke Environmental Stable Isotope Laboratory (DEVIL). The CRDS was calibrated with a set of three known gas standards for concentration (Airgas Inc., Radnor, PA) and $\delta^{13}\text{C}$ (Isometrics Canada Inc., Victoria, BC) and was checked at least daily to ensure analyzer output was within 1‰ of a tank of CH_4 with $\delta^{13}\text{CH}_4$ measured by a private lab (Isotech Laboratories, Inc., Champaign, IL; published precision $\pm 0.1\%$). In cases where the concentrations in the samples exceeded the calibrated range of the instrument, a dilution with zero air was created using a glass in-line dilution vessel.

The concentrations and isotopic signatures of ethane and propane in air and their ratio to methane provide independent ways to identify biogenic and thermogenic methane sources.²⁶ Because microbes do not produce ethane or propane in the soil or shallow subsurface,^{23,27} lower ratios of methane to higher-chain hydrocarbons ($< \sim 100$, $[\text{CH}_4]/([\text{C}_2\text{H}_6] + [\text{C}_3\text{H}_8])$) are characteristic of hydrocarbons derived from a thermogenic source, such as natural gas. In contrast, high ratios ($\gg 1000$) are characteristic of a microbially derived biogenic methane source.²³

Ethane and propane concentrations were measured using an Agilent 7890A gas chromatograph (GC) system with a flame ionization detector (Agilent Technologies, Inc., Wilmington, DE). From each sample, 500 μL of gas was extracted from the Tedlar sampling bag or stainless steel flask. For cryo-focusing, the injected gases were condensed at liquid N temperatures onto a metal trap filled with *n*-octanol-coated silica (Porasil, Restek Inc., Bellefonte PA). After five minutes of freezing, the metal trap was immersed in a Dewar flask of near-boiling water to release the trapped methane, ethane, and propane onto the GC column. Samples were passed through a HP-PLOT/Q column (30 m length, 0.32 mm diameter, 1.3 mL/min flow rate) with the following temperature program: 5 min isothermal hold at 60 °C followed by 5 °C/min ramp to 120 °C. The GC was calibrated using 250 and 1000 μL injections of standard gases containing methane, ethane, and propane in the following concentrations: 10, 100, 1000 ppm. High purity 1 000 000 ppm standards of methane and ethane were also injected (Air Liquide, Paris, France). The resulting calibration

curves were run daily during the measurement period ($0.993 \leq r^2 \leq 1.000$ for each individual curve).

To determine the $\delta^{13}\text{C}_2\text{H}_6$ signatures of street leaks and pipeline gas, sample gases were delivered by continuous flow via open split in the GC-C (GC-combustion) unit to a ThermoFinnigan Delta+XL IRMS (Thermo Finnigan, Bremen, Germany). Raw $\delta^{13}\text{C}$ data were normalized to Vienna Pee Dee Belemnite (V-PDB) using tank ethane, as well as headspace gas in septum bottles, with known $\delta^{13}\text{C}$ values (Isotech Laboratories, Inc., Champaign IL). Samples were frequently measured with duplicate analyses, and results for each sample were averaged. The standard deviation of replicates of sample and standard gases ranged from 0.1 to 0.5‰ $\delta^{13}\text{C}$ relative to V-PDB. Statistical differences between air sampled from leaks and natural gas pipeline gas for methane and ethane concentrations and isotopic ratios were assessed by *t* test.

To quantify methane emissions from individual street leaks, we used the Picarro “Plume Scanner” technology in June of 2013. The Plume Scanner technique is a direct emission measurement that utilizes (1) a mobile Picarro cavity ring-down spectrometer and a gas sampling system based on AirCore technology for recording short plume events, replaying them more slowly for improved isotope accuracy,²⁸ (2) a GPS tracker (Hemisphere GNSS, Scottsdale, AZ; Model R100); and (3) a 2-D sonic anemometer (Climatronics Corp. Bohemia, NY). As the Plume Scanner vehicle drives downwind across the plume, the air is simultaneously sampled at four heights (0.43, 1.09, 1.75, and 2.44 m) above the road surface on a mast mounted on the vehicle. When a plume is detected, the AirCore System’s valves are triggered into playback mode, in which the analyzer is connected in sequence to each of the four tube lengths. The emission estimate, $E(t)$, is determined by integrating over the concentration in the *y*-axis (i.e., the direction of the car motion) and the *z*-axis (height), after the background concentration has been subtracted (i.e., $C(y,z) - C_0$) is the density of the vector field of CH_4 gas and after multiplying the integrated values by the lateral wind speed *u* as measured by the anemometer mounted on the vehicle roof: $E(t) = \iint (C(y,z,t) - C_0) \times u(x,y,z) \, dy \, dz$. In this way, the methane spatial distribution can be reconstructed as an intensity map or plume image and fugitive emission rates of localized sources such as street leaks or natural gas production pads can be made quickly (~7 min per measurement). Verification using controlled methane releases of 15 L min⁻¹ under stable atmospheric conditions (Pasquill²⁹ atmospheric stability class C or D with strong and steady winds) gave a mean Plume Scanner measurement of 15.3 L min⁻¹ (± 2.3 L min⁻¹ s.d.); under somewhat less stable atmospheric conditions (Pasquill stability class A or B), 16 L min⁻¹ controlled releases gave a mean measurement of 14.9 L min⁻¹ (± 8.7 L min⁻¹ s.d.). Weather conditions on measurement days in June of 2013 were overcast with wind speeds that ranged from 1 to 3 m/s, conditions consistent with Pasquill stability classes B and C.

RESULTS AND DISCUSSION

Washington, DC had 5893 pipeline leaks across 1500 road miles of the city (Figures 1 and 2). There were 1122 street leaks >5 ppm CH_4 , roughly 2.5-times the background concentration of ~2 ppm CH_4 in the city (Figure 2). Additionally, there were 334 leaks >10 ppm CH_4 and 67 leaks >25 ppm CH_4 (Figure 2). The mean leak concentration observed was 4.6 ppm CH_4 (median value of 3.1 ppm CH_4), and the maximum leak

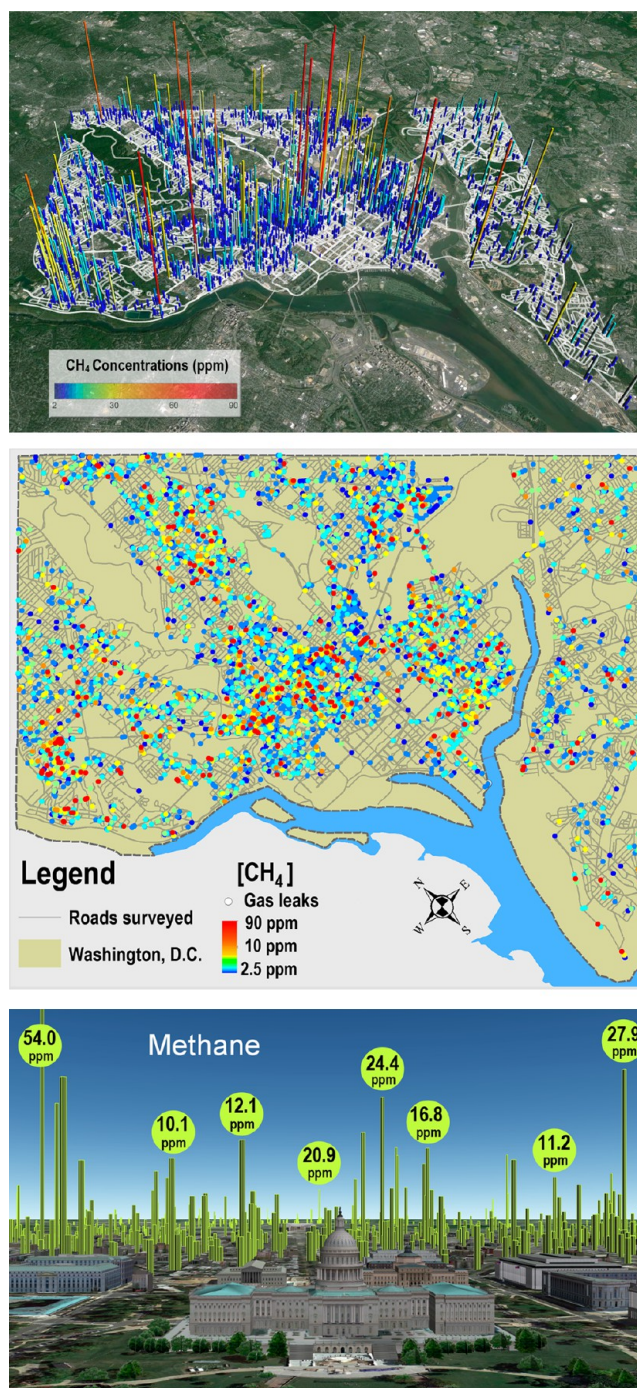


Figure 1. Methane leaks (5893 spikes >2.5 ppm) mapped across the ~1500 road miles of Washington, DC (upper panel: white = roads driven) and in planimetric view (middle panel). A close-up of leaks near the U.S. Capitol Building (lower panel) showing high leak densities east of the building but few leaks over the National Mall, where very few natural gas pipelines exist.

concentration was 88.6 ppm CH_4 , ~45-times higher than background CH_4 concentrations in air.

The isotopic signatures of the methane and ethane sampled from 19 street leaks in Washington, DC closely matched the signature of the pipeline gas in the city (Figure 3). The $\delta^{13}\text{C}_{\text{CH}_4}$ of the leaks had a mean value of -38.2‰ ($\pm 3.9\text{‰}$ s.d.), statistically indistinguishable from the value of -39.0‰ (s.d. = 0.6‰) for the pipeline natural gas ($P = 0.42$ by *t* test). In

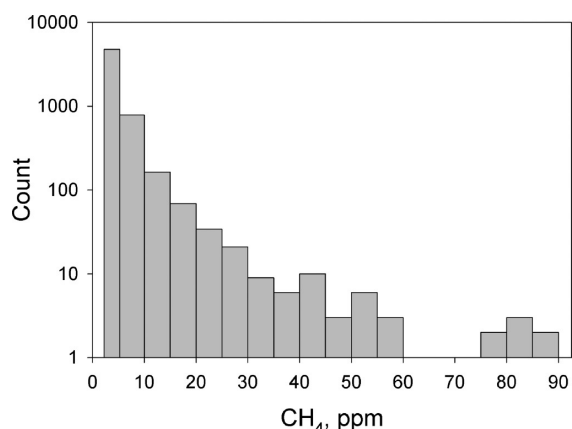


Figure 2. A histogram of methane concentrations for the 5893 leaks observed across the roads of Washington, DC.

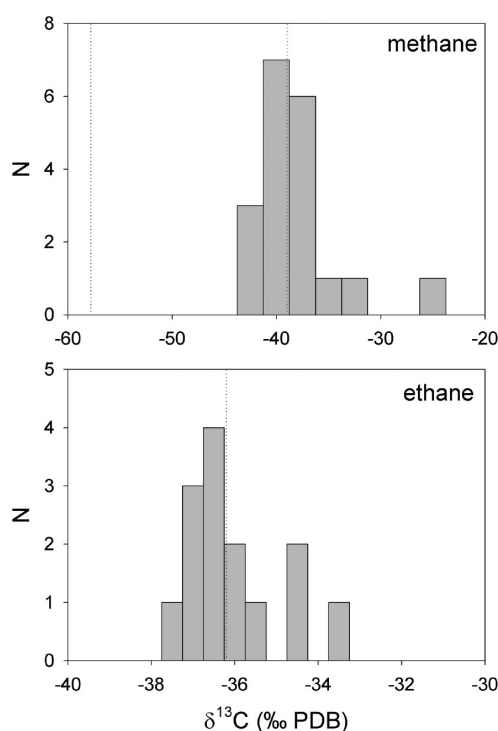


Figure 3. The $\delta^{13}\text{C}_{\text{CH}_4}$ (upper panel, $n = 19$) and $\delta^{13}\text{C}_{\text{C}_2\text{H}_6}$ (lower panel, $n = 14$) values of gas samples collected from street leaks in Washington, DC. The mean $\delta^{13}\text{C}$ values for methane and ethane were -38.2‰ ($\pm 3.9\text{‰}$ s.d.) and -36.5‰ ($\pm 1.1\text{‰}$ s.d.), respectively, relative to Vienna Pee Dee Belemnite. The dotted lines within the distributions are the values measured for pipeline natural gas in the city (-39.0‰ and -36.2‰ for CH_4 and C_2H_6 , respectively). The dotted line to the left in the upper panel is a typical $\delta^{13}\text{C}_{\text{CH}_4}$ value for biogenic methane sources from landfills, sewers, and wetlands (-57.8‰ ; Phillips et al.²⁰). Fourteen of 19 samples had sufficient $[\text{C}_2\text{H}_6]$ to measure $\delta^{13}\text{C}_{\text{C}_2\text{H}_6}$ accurately.

comparison, the typical $\delta^{13}\text{C}_{\text{CH}_4}$ value for biogenic methane sources from landfills, sewers, and wetlands are substantially lighter (e.g., an average value of -57.8‰ in Boston;²⁰ see additional references^{30–32}). Similar to the methane isotope values, the isotopic signatures of ethane ($\delta^{13}\text{C}_{\text{C}_2\text{H}_6}$) in our leak samples also matched the $\delta^{13}\text{C}_{\text{C}_2\text{H}_6}$ of the pipeline closely: -36.5 (± 1.1 s.d.) and -36.2 , respectively (Figure 3).

Methane concentrations assessed by GC analysis in air from the 19 leaks sampled from the sources at street level ranged from 2530 ppm to 258 000 ppm CH_4 , with average and median CH_4 concentrations of 77 000 and 69 000 ppm, respectively. Ethane concentrations in the same air samples ranged from 39 to 9960 ppm C_2H_6 , with propane values between 6 and 1120 ppm C_3H_8 . The average $\text{CH}_4/(\text{C}_2\text{H}_6 + \text{C}_3\text{H}_8)$ ratio across 19 street leaks in Washington, DC, was 25.5 (± 8.9 s.d.), characteristic of a fossil fuel source and consistent with our pipeline samples (19.0 ± 0.84 s.d.).

Pipeline distribution companies grade leaks based on the hazards that the leaks pose, often using the criteria set by the Gas Pipeline Technology Committee of the American Gas Association.³³ A grade 1 leak represents “an existing or probable hazard to persons or property, and requires immediate repair or continuous action until the conditions are no longer hazardous.”³⁴ A grade 1 leak is often assessed in the field using a portable combustible gas analyzer (CGA) and, when measured in a manhole or other confined space, is defined with a lower limit of 40 000 ppm methane or 4% gas, which corresponds to a threshold of least 80% of the lower explosion limit (LEL) of methane in air ($\sim 5\%$ methane or 50 000 ppm).

In January and February of 2013, we used a CGA in the field to measure the gas concentrations in manholes at 19 high-concentration leak locations. At that time, 12 of the 19 locations had manholes with air categorized as grade 1 leaks (Table 1). The values were as high as 50% methane (500 000

Table 1. Location and Concentration of CH_4 Measured in 12 Grade 1 Leaks Across Washington, DC, in an Initial Sampling (January and February, 2013) and in Follow-up Sampling in June of 2013^a

latitude	longitude	address	% CH_4 Jan/Feb 2013	% CH_4 June 2013
38.90306	-77.002068	1003 Third Street NE	6	9
38.90375	-77.00589	First Street NE at L Street NE	11	12
38.905439	-77.04737	M Street NW at New Hampshire Ave NW	11	n/a
38.908599	-77.008173	O Street NE by North Capitol Street	13	<0.05
38.912621	-77.015438	R Street at 3st NW	28	21
38.921077	-76.96647	2401 30th Street NE at Adams Street	47	11
38.924723	-77.038229	1630 Fuller Street NW	10	4
38.929994	-77.034788	3221 Hiatt Place NW	11	9
38.938177	-77.101682	5074 Sedgwick Street NW at Tilden Street NW	40	31
38.955283	-77.080354	5028 41st Street NW	21	0.25
38.959218	-77.024337	5720 Eighth Street NW on Marietta Place NW	50	5
38.984136	-77.036272	16th Street NW at Jonquil Street NW	9	11

^aA value of 1% CH_4 is equivalent to 10 000 ppm CH_4 ; 10% CH_4 is 100 000 ppm CH_4 .

ppm) in the manhole air. As recommended, we notified the local distribution company of these leaks for immediate remediation. Subsequently in June of 2013, we returned to each of the dozen locations and rechecked the manholes. Of the twelve grade 1 leaks found initially, nine remained as grade 1 leaks four months later, two were below the grade 1 threshold

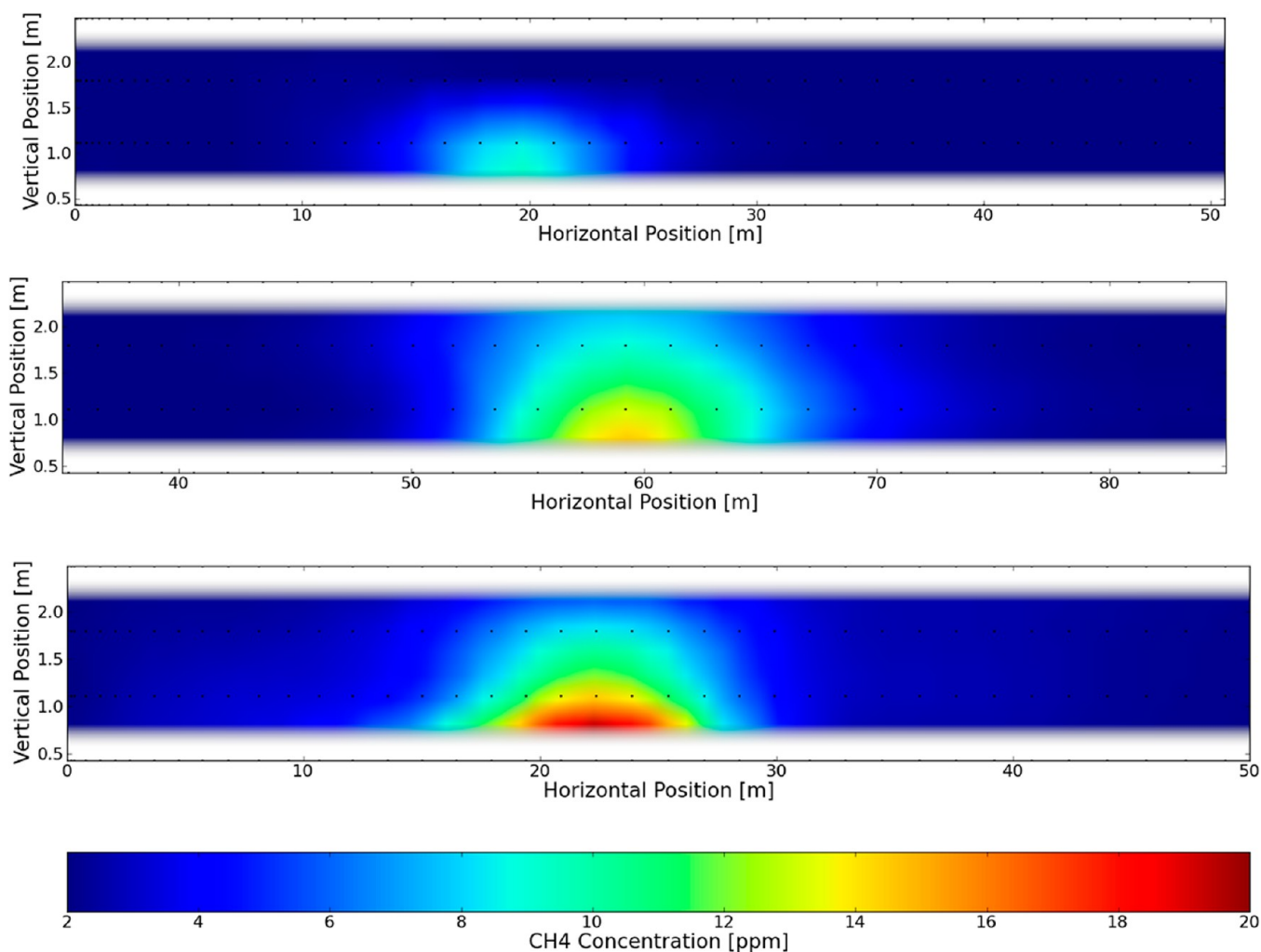


Figure 4. Methane concentrations observed in three street leaks in Washington, DC. The estimated emission rates (top to bottom for the three images) are 9200, 38 200 and 30 200 L CH₄ day⁻¹. Although the measured CH₄ concentrations were higher in the bottom panel than in the middle panel, the wind speed was also lower (1.23 versus 1.75 m/s), resulting in a higher estimated emission rate for the leak in the middle image. The four sampling heights are represented by the vertical boundaries of the image and the two dashed lines for the intermediate sampling heights (1.09 and 1.75 m height above the road surface).

(concentrations of <0.05% and 0.25% methane), and one was inaccessible to us because of road construction. Concentration values in the manholes in June were as high as 31% CH₄ (310 000 ppm).

As a first attempt at measuring emission rates from pipeline leaks in Washington, DC, we also quantified methane emissions at four street leaks, using the new Picarro Plume Scanner technology (see Methods). The estimated emission rates from the four leaks were 9200, 15 000, 30 200, and 38 200 L CH₄ day⁻¹ (330, 530, 1070, and 1350 standard ft³ CH₄ day⁻¹) (Figure 4). The estimated leak rate of 15 000 L CH₄ day⁻¹ is a lower bound for this measurement because the plume was taller than the mast on the vehicle measuring it. According to the American Gas Association,³³ the average home in the U.S. uses 5470 L (193 ft³) of natural gas per day. Given that estimate, the four leaks quantified here represent the approximate daily amounts of natural gas used by 1.7, 2.7, 5.5, and 7.0 homes, respectively, lost to the atmosphere.

The density of leaks in Washington, DC was comparable to that observed previously in Boston, MA, but the concentrations measured were higher. Previously, Phillips et al.²⁰ mapped 3356 leaks across the 785 road miles of Boston, MA. On the basis of

the 5893 leaks found across Washington, DC's 1500 road miles (Figures 1 and 2), the two cities had similar leak densities: 3.9 and 4.2 leaks per road mile for Washington, DC and Boston, respectively (and measured with similar sampling heights 0.5 m above road surface and vehicle speeds, typically 20 mi h⁻¹ (32 km h⁻¹)). The biggest difference was the greater CH₄ concentrations mapped in Washington, DC, which had 51 leaks with concentrations greater than 28.6 ppm CH₄, the highest value we observed in Boston.²⁰ Washington, DC, also had a maximum value of 88.6 ppm CH₄, more than three times the highest concentration observed in Boston. Both high leak densities and high emission rates of methane contribute to extensive amounts of lost-and-unaccounted-for natural gas in Washington, DC.

The Pipeline and Hazardous Materials Safety Administration (PHMSA), a regulatory agency within the U.S. Department of Transportation, keeps track of pipeline safety and natural gas that is lost or unaccounted for during distribution.²² As defined by PHMSA, lost and unaccounted (LAU) gas is the difference between the amount of gas purchased (e.g., what enters the gateway to a city) and the amount of gas sold (e.g., what is

metered to consumers). Pipeline leaks and errors in metering both contribute to estimates of LAU gas.

On the basis of the PHMSA data for 2011, 174 companies in the United States had natural gas distribution systems of at least 1000 pipeline miles.²² Across those companies, the average LAU gas term reported by each company in 2011 was 1.6% (Figure 5). There were 23 companies (13%) that reported zero

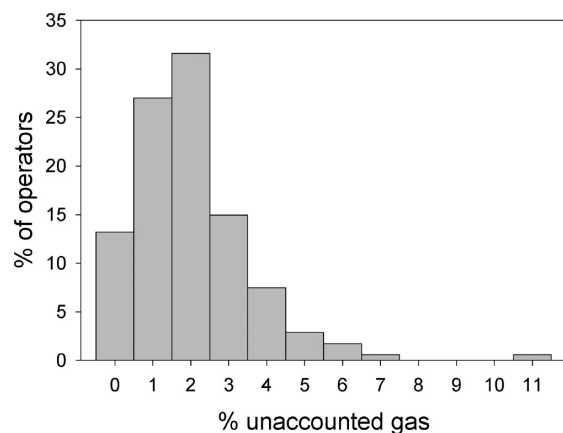


Figure 5. Histogram of lost-and-unaccounted-for natural gas in 2011 reported by distribution companies to the Pipeline and Hazardous Safety Materials Administration (PHMSA²²). The histogram shows data for providers with a minimum of 1000 pipeline miles.

LAU gas, 47 (27%) that reported losses between 0 and 1% LAU gas, 55 companies (32%) between 1 and 2% LAU gas, 26 companies (15%) between 2 and 3%, and 23 companies (13%) with losses >3%, including a maximum of 11% LAU gas. LAU for the primary service provider in Washington, DC was substantially higher than the average across companies, 4% in 2011 and ~3.5% averaged across the previous decade.

PHMSA data^{22,35} also shed light on the potential cause of the high LAU term and leak rate observed in Washington, DC, particularly the prevalence of older cast-iron piping (Table 2).

Table 2. Length (In Miles) For States with >400 Miles of Cast-Iron Mains and the Percentage of Cast-Iron Distribution Mains by State and for the District of Columbia (PHMSA³⁵)

location	miles cast-iron mains	% cast-iron mains
New Jersey	5044	15
New York	4417	9
Massachusetts	3792	18
Pennsylvania	3221	7
Michigan	3101	5
Illinois	1744	3
Connecticut	1467	19
Maryland	1399	10
Alabama	1383	5
Missouri	1113	4
Texas	934	1
Rhode Island	859	27
Ohio	582	1
Nebraska	503	4
Louisiana	470	2
District Of Columbia	419	35
Virginia	406	2

Eleven percent of reported incidents for U.S. gas distribution mains between 2005 and 2011 involved cast-iron pipes, even though such pipes comprised only 2.6% of distribution mains.²² In Boston, the frequency of leaks per road mile from natural gas pipelines was associated primarily with the presence of cast-iron mains that were as much as a century old ($r^2 = 0.79$, $P < 0.001$; Phillips et al.²⁰). Leak rates did not differ statistically with socioeconomic indicators such as median income or poverty level. As a state, Massachusetts has 3792 miles of cast-iron piping, the third highest after New Jersey and New York (5044 and 4,017 miles, respectively).³⁵ Pennsylvania, Michigan, Illinois, Connecticut, Maryland, Alabama, and Missouri complete the top ten, each with >1000 miles of cast-iron mains (Table 2). Washington, DC would rank 16th on the list if it were a state, with 419 cast-iron miles. However, it ranks first in the percentage of distribution mains made of cast iron, 35%, compared to states with the highest percentages: Rhode Island (27%), Connecticut (19%), Massachusetts (18%), New Jersey (15%), and Maryland (10%) (Table 2). The prevalence of cast-iron mains likely explains at least part of the high LAU term for Washington, DC and the relatively high number of leaks observed per mile.

Providing financial incentives to fix leaks will save money, particularly incentives to replace cast-iron, bare steel, and other older, unprotected mains. Recently, the U.S. Energy Information Administration determined that \$3.1B worth of natural gas was lost and unaccounted for annually in the United States between 2005 and 2010.³⁶ A more recent report estimated that U.S. consumers paid more than \$20 billion between 2000 and 2011 for lost and unaccounted for natural gas.³⁷ Several barriers to pipeline repair and replacement exist, however, as cost recovery for pipeline repairs by distribution companies is often capped by Public Utility Commissions (PUCs). Furthermore, consumers often pay for all or most of the lost-and-unaccounted-for gas through user fees, meaning that the local distribution company has less financial incentive to fix leaks than might be predicted from the value of lost gas alone. To overcome the barriers to fixing pipeline leaks, PUCs could allow distribution companies to recover funds to accelerate pipeline replacement faster than a typical 40-year replacement cycle. For instance, New Hampshire implemented a Cast Iron/Bare Steel (CIBS) replacement program that allows the distribution companies to recover repair costs.³⁸ Other mechanisms that have been suggested include the application of carbon-offset programs and placing a price on carbon emissions (e.g., a carbon fee or a cap-and-trade system).¹⁴

Fixing pipeline leaks will also improve consumer safety and save lives. Pipeline safety is already improving in the U.S. Between 1991 and 2001, major accidents and incidents reported for gas distribution systems declined by half on average.²² Nevertheless, 17 fatalities, 68 injuries, and \$133 M in property damage are caused each year on average by incidents involving transmission and distribution pipelines for natural gas.²² A natural gas explosion in San Bruno, CA, in 2010 killed eight people and destroyed 38 homes. An explosion in Allentown, PA, in 2011 killed five people and destroyed eight homes; after a 16-month investigation, the Pennsylvania Public Utility Commission concluded that the explosion was caused by a leak from an 83-year-old cast-iron pipe.

Reducing greenhouse gas emissions and improving air quality provide additional reasons to repair pipeline leaks. CH₄ is a potent greenhouse gas with an estimated 20-year global warming potential 86 times greater than CO₂.⁷ Ozone

formation in urban areas can be catalyzed by CH₄ reacting with NO_x²¹ and reducing hydrocarbon concentrations in urban areas could reduce rates of respiratory and cardiopulmonary disease associated with tropospheric ozone production.^{2,21} The greenhouse gas and public health benefits of reducing natural gas leaks would be in addition to other benefits of natural gas compared to coal, such as fewer SO₂, mercury, and particulate matter emissions² and lower CO₂ emissions per unit energy.

Through bottom-up surveys, such as this one, and complementary top-down approaches,^{5,15,16} fugitive emissions of methane and other hydrocarbons in natural gas can be reduced, to the economic and environmental benefit of gas companies, municipalities, and consumers. Further campaigns to map and repair urban pipeline leaks around the world are critical for reducing greenhouse gas emissions, improving air quality and consumer safety, and saving consumers money.^{2,4,20,21,39,40}

AUTHOR INFORMATION

Corresponding Author

*Phone: 1-919-660-7408; fax 1-919-660-7425; e-mail: jackson@duke.edu; rob.jackson@stanford.edu.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

Duke University's Nicholas School of the Environment and Center on Global Change funded this research. We thank Tracy Tsai of Picarro, Inc., for scientific assistance, Jon Karr of the Duke Environmental Isotope Laboratory (DEVIL) for assistance with isotope analyses, and William Chameides, Dean of the Nicholas School, for his scientific input and support. The Jackson lab and anonymous reviewers provided helpful comments on the manuscript.

REFERENCES

- U. S. Environmental Protection Agency. *Methane Emissions*; Washington, DC, 2012; <http://epa.gov/climatechange/ghgemissions/gases/ch4.html>.
- Shindell, D.; Kuylenstierna, J. C. I.; Vignati, E.; van Dingenen, R.; Amann, M.; Klimont, Z.; Anenberg, S. C.; Müller, N.; Janssens-Maenhout, G.; Raes, F.; Schwartz, J.; Faluvegi, G.; Pozzoli, L.; Kupiainen, K.; Höglund-Isaksson, L.; Emberson, L.; Streets, D.; Ramanathan, V.; Hicks, K.; Oanh, N. T. K.; Milly, G.; Williams, M.; Demkine, V.; Fowler, D. Simultaneously mitigating near-term climate change and improving human health and food security. *Science* **2012**, *335* (6065), 183–189.
- Howarth, R.; Shindell, D.; Santoro, R.; Ingraffea, A.; Phillips, N.; Townsend-Small, A. Methane Emissions from Natural Gas Systems. National Climate Assessment. Ref. no. 2011–0003, 2012.
- Alvarez, R. A.; Pacala, S. W.; Winebrake, J. J.; Chameides, W. L.; Hamburg, S. P. Greater focus needed on methane leakage from natural gas infrastructure. *Proc. Natl. Acad. Sci. U. S. A.* **2012**, *109* (17), 6435–6440.
- Townsend-Small, A.; Tyler, S. C.; Pataki, D. E.; Xu, X.; Christensen, L. E. Isotopic measurements of atmospheric methane in Los Angeles, California, USA: Influence of “fugitive” fossil fuel emissions. *J. Geophys. Res.* **2012**, *117* (D7), D07308.
- Schlesinger, W. H.; Bernhardt, E. S. *Biogeochemistry: An Analysis of Global Change*; Academic Press: Waltham, MA, 2013.
- Intergovernmental Panel on Climate Change, AR5. Climate Change 2013: The Physical Science Basis. Working Group I Contribution to the IPCC 5th Assessment Report.
- Wunch, D.; Wennberg, P. O.; Toon, G. C.; Keppel-Aleks, G.; Yavin, Y. G. Emissions of greenhouse gases from a North American megacity. *Geophys. Res. Lett.* **2009**, *36* (15), 16 DOI: 10.1029/2009GL039825.
- Miller, S. M.; Wofsy, S. C.; Michalak, A. M.; Kort, E. A.; Andrews, A. E.; Biraud, S. C.; Dlugokencky, E. J.; Eluszkiewicz, J.; Fischer, M. L.; Janssens-Maenhout, G.; Miller, B. R.; Miller, J. B.; Montzka, S. A.; Nehrkorn, T.; Sweeney, C. Anthropogenic emissions of methane in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, DOI: 10.1073/pnas.1314392110.
- Osborn, S. G.; Vengosh, A.; Warner, N. R.; Jackson, R. B. Methane contamination of drinking water accompanying gas-well drilling and hydraulic fracturing. *Proc. Natl. Acad. Sci. U. S. A.* **2011**, *108*, 8172–8176, DOI: 10.1073/pnas.1100682108.
- Jackson, R. B.; Vengosh, A.; Darrah, T. H.; Warner, N. R.; Down, A.; Poreda, R. J.; Osborn, S. G.; Zhao, K.; Karr, J. D. Increased stray gas abundance in a subset of drinking water wells near Marcellus shale gas extraction. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110*, 11250–11255, DOI: 10.1073/pnas.1221635110.
- Allen, D. T.; Torres, V. M.; Thomas, J.; Sullivan, D. W.; Harrison, M.; Hendler, A.; Herndon, S. C.; Kolb, C. E.; Fraser, M. P.; 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. Natl. Acad. Sci. U. S. A.* **2013**, DOI: 10.1073/pnas.1304880110.
- Dedikov, J. V.; Akopova, G. S.; Gladkaja, N. G.; Piotrovskij, A. S.; Markellov, V. A.; Salichov, S. S.; Kaesler, H.; Ramm, A.; Müller von Blumencron, A.; Lelieveld, J. Estimating methane releases from natural gas production and transmission in Russia. *Atmos. Environ.* **1999**, *33* (20), 3291–3299.
- Lechtenböhrer, S.; Dienst, C.; Fishedick, M.; Hanke, T.; Fernandez, R.; Robinson, D.; Kantamaneni, R.; Gillis, B. Tapping the leakages: Methane losses, mitigation options and policy issues for Russian long distance gas transmission pipelines. *Int. J. Greenhouse Gas Control* **2007**, *1*, 387–395.
- Pétron, G.; Frost, G.; Miller, B. R.; Hirsch, A. I.; Montzka, S. A.; Karion, A.; Trainer, M.; Sweeney, C.; Andrews, A. E.; Miller, L.; Kofler, J.; Bar-Ilan, A.; Dlugokencky, E. J.; Patrick, L.; Moore, C. T.; Ryerson, T. B.; Siso, C.; Kolodzey, W.; Lang, P. M.; Conway, T.; Novelli, P.; Masarie, K.; Hall, B.; Guenther, D.; Kitzis, D.; Miller, J.; Welsh, D.; Wolfe, D.; Neff, W.; Tans, P. Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study. *J. Geophys. Res.* **2012**, *117* (D4), D04304.
- Peischl, J.; Ryerson, T. B.; Brioude, J.; Aikin, K. C.; Andrews, A. E.; Atlas, E.; Blake, D.; Daube, B. C.; de Gouw, J. A.; Dlugokencky, E.; Frost, G. J.; Gentner, D. R.; Gilman, J. B.; Goldstein, A. H.; Harley, R. A.; Holloway, J. S.; Kofler, J.; Kuster, W. C.; Lang, P. M.; Novelli, P. C.; Santoni, G. W.; Trainer, M.; Wofsy, S. C.; Parrish, D. D. Quantifying sources of methane using light alkanes in the Los Angeles basin, California. *J. Geophys. Res.* **2013**, *118* (10), 4974–4990.
- Karion, A.; Sweeney, C.; Pétron, G.; Frost, G.; Hardesty, R. M.; Kofler, J.; Miller, B. R.; Newberger, T.; Wolter, S.; Banta, R.; Brewer, A.; Dlugokencky, E.; Lang, P.; Montzka, S. A.; Schnell, R.; Tans, P.; Trainer, M.; Zamora, R.; Conley, S. Methane emissions estimate from airborne measurements over a western United States natural gas field. *Geophys. Res. Lett.* **2013**, *40* (16), 4393–4397.
- Mitchell, C.; Sweet, J.; Jackson, T. A study of leakage from the UK natural gas distribution system. *Energy Policy* **1990**, *18* (9), 809–818.
- Lelieveld, J.; Lechtenböhrer, S.; Assonov, S. S.; Brenninkmeijer, C. A. M.; Dienst, C.; Fishedick, M.; Hanke, T. Greenhouse gases: Low methane leakage from gas pipelines. *Nature* **2005**, *434*, 841–842.
- 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.
- West, J. J.; Fiore, A. M.; Horowitz, L. W.; Mauzerall, D. L. Global health benefits of mitigating ozone pollution with methane emission controls. *Proc. Natl. Acad. Sci. U. S. A.* **2006**, *103* (11), 3988–3993.

(22) PHMSA Data & Statistics (1992–2011 averages reported); U. S. Department of Transportation Pipeline and Hazardous Materials Safety Administration (PHMSA): Washington, DC, 2012; <http://www.phmsa.dot.gov/pipeline/library/data-stats>.

(23) Schoell, M. The hydrogen and carbon isotopic composition of methane from natural gases of various origins. *Geochim. Cosmochim. Acta* **1980**, *44* (5), 649–661.

(24) Annual Greenhouse Gas Index (AGGI). National Oceanic and Atmospheric Administration, Earth System Research Laboratory, Global Monitoring Division: Boulder, CO, 2012; <http://www.esrl.noaa.gov/gmd/aggi/>.

(25) Crosson, E. R. A cavity ring-down analyzer for measuring atmospheric levels of methane, carbon dioxide, and water vapor. *Appl. Phys. B: Laser Opt.* **2008**, *92* (3), 403–408.

(26) Prinzhofer, A. A.; Huc, A. Y. Genetic and post-genetic molecular and isotopic fractionations in natural gases. *Chem. Geol.* **1995**, *126* (3–4), 281–290.

(27) Bernard, B. B. Light hydrocarbons in marine sediments. Ph.D. Dissertation, Texas A&M University: College Station, TX, 1978.

(28) Karion, A.; Sweeney, C.; Tans, P.; Newberger, T. AirCore: An innovative atmospheric sampling system. *J. Atmos. Oceanic Technol.* **2010**, *27* (11), 1839–1853.

(29) Pasquill, F. The estimation of the dispersion of wind borne material. *Meteor. Mag.* **1961**, *90*, 33–49.

(30) Bergamaschi, P.; Lubina, C.; Knigstedt, R.; Fischer, H.; Veltkamp, A. C.; Zwaagstra, O. Stable isotopic signatures ($\delta^{13}\text{C}$, δD) of methane from European landfill sites. *J. Geophys. Res.* **1998**, *103* (D7), 8251–8265.

(31) Börjesson, G.; Chanton, J.; Svensson, B. H. Methane oxidation in two Swedish landfill covers measured with carbon-13 to carbon-12 isotope ratios. *J. Environ. Qual.* **2001**, *30* (2), 369–376.

(32) Hornibrook, E. R. C.; Longstaffe, F. J.; Fyfe, W. F. Evolution of stable carbon isotope compositions for methane and carbon dioxide in freshwater wetlands and other anaerobic environments. *Geochim. Cosmochim. Acta* **2000**, *64* (6), 1013–1027.

(33) American Gas Association. How to measure natural gas. Washington D.C.; 2013; <http://www.aga.org/KC/aboutnaturalgas/additional/Pages/HowtoMeasureNaturalGas.aspx>

(34) PHMSA. (2001) Training guide for operators of small LP gas systems (accessed 15 September 2013): <http://phmsa.dot.gov/staticfiles/PHMSA/DownloadableFiles/smallpgas-chapt10.pdf>

(35) PHMSA. (2013a) *Pipeline Replacement Updates: Cast and Wrought Iron Pipeline Inventory*; U. S. Department of Transportation Pipeline and Hazardous Materials Safety Administration: Washington, DC, 2013; http://opsweb.phmsa.dot.gov/pipeline_replacement/cast_iron_inventory.asp.

(36) Energy Information Administration Website; <http://www.eia.gov/>; http://www.eia.gov/dnav/ng/ng_sum_lsum_dcu_nus_a.htm; http://www.eia.gov/naturalgas/annual/pdf/table_a01.pdf; http://www.eia.gov/naturalgas/annual/archive/2009/pdf/table_a01.pdf.

(37) Markey, E. J. America pays for gas leaks: Natural gas pipeline leaks cost consumers billions; U. S. House Natural Resources Committee: Washington, DC, 2013.

(38) PHMSA. (2013b) *State Replacement Programs for Pipeline Infrastructure*; U. S. Department of Transportation Pipeline and Hazardous Materials Safety Administration: Washington, DC, 2013; <http://opsweb.phmsa.dot.gov/pipelineforum/pipeline-materials/state-pipeline-system/state-replacement-programs/>.

(39) Han, Z. Y.; Weng, W. G. Comparison study on qualitative and quantitative risk assessment methods for urban natural gas pipeline network. *J. Hazard. Mater.* **2011**, *189* (1–2), 509–518.

(40) Gurney, K. R.; Razlivanov, I.; Song, Y.; Zhou, Y.; Benes, B.; Abdul-Massih, M. Quantification of fossil fuel CO₂ emissions on the building/street scale for a large U.S. city. *Environ. Sci. Technol.* **2012**, *46* (21), 12194–12202.