Rapid communication

Mapping urban pipeline leaks: Methane leaks across Boston

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ABSTRACT

Natural gas is the largest source of anthropogenic emissions of methane (CH_4) in the United States. To assess pipeline emissions across a major city, we mapped CH_4 leaks across all 785 road miles in the city of Boston using a cavity-ring-down mobile CH_4 analyzer. We identified 3356 CH_4 leaks with concentrations exceeding up to 15 times the global background level. Separately, we measured $\delta^{13}CH_4$ isotopic signatures from a subset of these leaks. The $\delta^{13}CH_4$ signatures (mean $= -42.8‰ \pm 1.3‰$ s.e.; $n = 32$) strongly indicate a fossil fuel source rather than a biogenic source for most of the leaks; natural gas sampled across the city had average $\delta^{13}CH_4$ values of $-36.8‰$ ($\pm 0.7‰$ s.e., $n = 10$), whereas CH_4 collected from landfill sites, wetlands, and sewer systems had $\delta^{13}CH_4$ signatures $\sim 20‰$ lighter ($\mu = -57.8‰, \pm 1.6‰$ s.e., $n = 8$). Repairing leaky natural gas distribution systems will reduce greenhouse gas emissions, increase consumer health and safety, and save money.

1. Introduction

Methane (CH_4) is a greenhouse gas more potent molecule for molecule than carbon dioxide (Shindell et al., 2012). In the United States, leaks of CH_4 from natural gas extraction and pipeline transmission are the largest human-derived source of emissions (EPA, 2012). However, CH_4 is not just a potent greenhouse gas; it also influences air quality and consumer health. CH_4 reacts with NO_x to catalyze ozone formation in urban areas (West et al., 2006). Incidents involving transmission and distribution pipelines for natural gas in the U. S. cause an average of 17 fatalities, 68 injuries, and $133 M in property damage each year (PHMSA, 2012). A natural gas pipeline explosion in San Bruno, CA, for instance, killed eight people and destroyed 38 homes in 2010. Detecting and reducing pipeline leaks of CH_4 and other hydrocarbons in natural gas are critical for reducing greenhouse gas emissions, improving air quality and consumer safety, and saving consumers money (West et al., 2006; Han and Weng, 2011; Shindell et al., 2012; Alvarez et al., 2012).

To assess CH_4 emissions in a major urban metropolis, we mapped CH_4 emissions over the entire 785 centerline miles of Boston's streets. To evaluate the likely source of the street-level CH_4 emissions, we also measured the $\delta^{13}C–CH_4$ carbon isotope composition, which can differentiate between biogenic (e.g., landfill, wetland, sewer) and thermogenic (e.g., natural gas) sources (Schoell, 1980).

2. Materials and methods

We conducted 31 mobile surveys during the period 18 August, 2011–1 October, 2011, covering all 785 road miles within Boston's city limits. We measured CH_4 concentration ([CH_4], ppm) using a mobile Picarro G2301 Cavity Ring-Down Spectrometer equipped with an A0491 Mobile Plume Mapping Kit (Picarro, Inc, Santa Clara, CA). This instrument was factory-calibrated on 15 August 2011, immediately prior to use in this study, and follow-up tests of the analyzer were made during 11–21 August, 2012, comparing analyzer output to a National Oceanic and Atmospheric Administration (NOAA) primary standard tank. In both pre- and post-checks, the analyzer output was found to be within 2.7 parts per billion of known [CH_4] in standard tanks, three orders of magnitude below typical atmospheric concentrations. Spectrometer and mobile GPS data were recorded every 1.1 s. To correct for a short time lag between instantaneous GPS location and a delay in [CH_4] measurement due to inlet tube length (~ 3 m), we used an auxiliary pump to increase tubing flow throughput to within 5 cm of the analyzer inlet; we also adjusted the time stamp on the [CH_4] readings based on a 1-s delay observed between analyzer response to a standard CH_4 source that we injected into the instrument while driving, and the apparent GPS location. We also checked the GPS-based locations of leaks with dozens of street-level sampling to confirm specific leak locations and the estimated sampling delay. Air was sampled through a 3.0 um Zefluor filter and Teflon tubing placed ~ 30 cm above road surfaces.

For our mobile survey data, we defined a "leak" as a unique, spatially contiguous group of [CH_4] observations, all values of which exceed a concentration threshold of 2.50 ppm. This was used as a threshold because it corresponded to the 90th

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percentile of the distribution of data from all road miles driven, and, relative to global background, is ~37% above 2011 mean mixing ratios observed at Mauna Loa (NOAA, 2012).

Independently of mobile street sampling of CH_4, we measured $\delta^{13}CH_4$ from a subset of the leaks with a Picarro G2112i Cavity Ring-Down Spectrometer (Crosson, 2008). This instrument is calibrated monthly using isotopic standards from Isometric Instruments (Victoria, BC, Canada). The instrument was checked at least once daily to ensure analyzer output was within 1‰ of a tank of CH_4 with $\delta^{13}CH_4$ measured by a private lab (Isotech Labs, IL). Samples were collected in 1-L Tedlar sampling bags with valve and septa fittings, manufactured by Environmental Supply Company (Durham, NC). A Gas Sentry CGO-321 handheld gas detector (Bascom-Turner, MA) was used to identify the area of highest ambient $[CH_4]$ at each site sampled for $\delta^{13}CH_4$. Sampling bags were pre-evacuated and filled at the area of highest ambient concentration at the sampling site using a hand pump. $\delta^{13}CH_4$ was analyzed using a Picarro G2112i with a sample hold time typically of a few days and always less than two weeks.

At a subset of sampling sites ($n = 12$), we collected duplicate samples in glass vials to assess potential leaking or fractionation by the Tedlar sampling bags. We also sent duplicate samples from a different subset of sampling sites ($n = 5$) to a private lab (Isotech Labs, IL) for independent $\delta^{13}CH_4$ analysis. These analyses suggest no significant fractionation or bias either from the sampling bags or the Picarro G2112i analyzer. Most samples were analyzed at less than the maximum hold time of two weeks, at which bag diffusion could account for a 1.2‰ drift in our measurements of $\delta^{13}CH_4$.

We compared $\delta^{13}CH_4$ of these locations with samples taken from area landfills, wetlands, and the Deer Island Water Treatment Facility. Sampling equipment and procedures, as well as laboratory analyses, for landfill and wetland sites were similar to those for $\delta^{13}CH_4$ sampling locations described above. Samples were collected from three capped, inactive landfills (there are currently no active landfills in the Boston area). At one former landfill site, samples were collected at approximately three-month intervals between September, 2011 and April, 2012. The $\delta^{13}CH_4$ signature of the landfill was consistent over this period (±3.4‰, s.e.). At all wetland sampling sites, a plastic chamber (10 cm × 25 cm × 5 cm) connected to a sampling tube was placed over the surface of exposed moist sediment or shallow (>5 cm) water. Sediment below the chamber was disturbed gently before drawing air samples from the headspace within the chamber. The sample from the Deer Island Treatment Facility was drawn from the headspace of a sample bottle of anaerobic sludge, collected onsite by Deer Island staff for daily monitoring of the facility's anaerobic sludge digesters.

3. Results and discussion

We identified 3356 CH_4 leaks (Figs. 1 and 2) exceeding 2.50 parts per million. Surface concentrations corresponding to these leaks ranged up to 28.6 ppm, 14-times above a surface background concentration of 2.07 ppm (the statistical mode of the entire concentration distribution). Across the city, 435 and 97 independent leaks exceeded 5 and 10 ppm, respectively.

Based on their $\delta^{13}CH_4$ signatures, the CH_4 leaks strongly resembled thermogenic rather than biogenic sources (Fig. 3). Samples of natural gas from the gateway pipelines to Boston and from other consumer outlets in the city were statistically indistinguishable, with an average $\delta^{13}CH_4$ signature of $-36.8‰$ (±0.7‰ s.e., $n = 10$; ‰ vs. Vienna Pee Dee Belemnite). In contrast, CH_4 collected from landfill sites, wetlands, and sewer systems reflected a greater fractionation from microbial activity and $\delta^{13}CH_4$ signatures ~20‰ lighter. Biogenic values ranged from $-53.1‰$ to $-64.5‰$ ($\mu = -57.8‰$, ±1.6‰ s.e., $n = 8$) for samples collected in four wetlands, three capped landfills, and the primary sewage facility for the city, Deer Island Sewage Treatment Plant, which had the heaviest sample observed for non-natural-gas sources ($-53.1‰$). Our results for biogenic CH_4 carbon isotope signatures are consistent with other studies of the $\delta^{13}CH_4$ signature of CH_4 from landfills (Bergamaschi et al., 1998; Borjesson et al., 2001) and wetlands (Hornibrook et al., 2000).

Peaks of $[CH_4]$ detected in the road surveys strongly reflected the signature of natural gas rather than biogenic sources (Table 1). The average $\delta^{13}CH_4$ value for peaks was $-42.8‰ ± 1.3‰$ ($n = 32$), reflecting a dominant signal from natural gas, likely altered in some cases by minor fractionation of natural gas traveling through soils and by mixing with background air ($\delta^{13}CH_4 = -47‰$; Dlugokencky et al., 2011). A minority of samples had $\delta^{13}CH_4$ more negative than
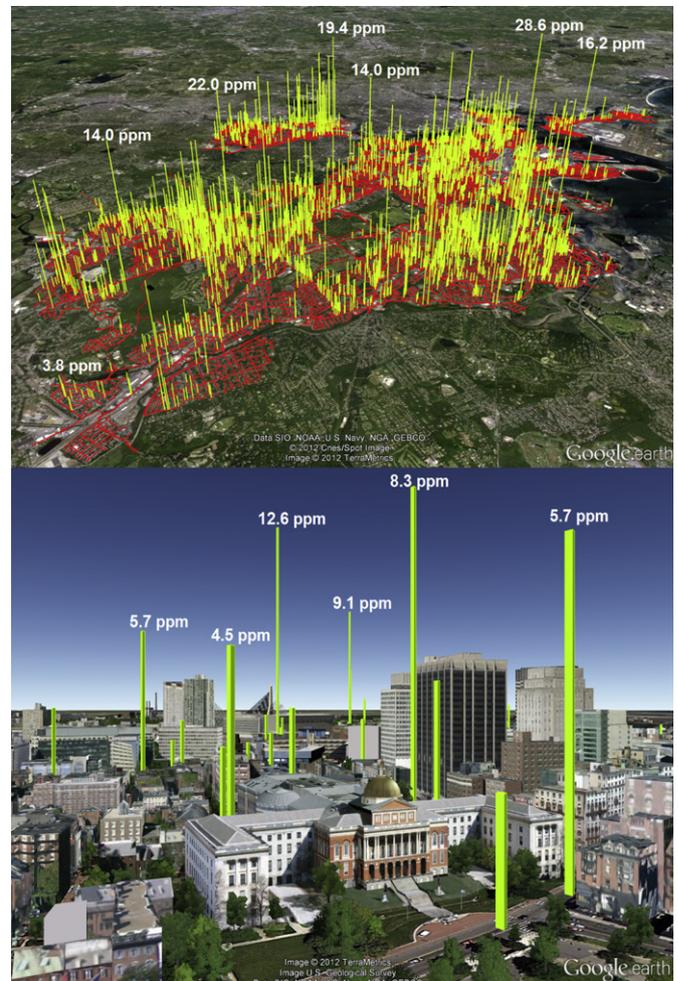

Fig. 1. Upper Panel: Methane leaks (3356 yellow spikes > 2.5 ppm) mapped on Boston's 785 road miles (red) surveyed in this study. Lower Panel: Leaks around Beacon Hill and the Massachusetts State House. Sample values of methane concentrations (ppm) are shown for each panel. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

that of background air, reflecting apparent influence of biogenic CH_4. Most samples emitted a distinct odor of the mercaptan additive associated with natural gas, including those with a larger apparent biogenic influence on $\delta^{13}CH_4$.
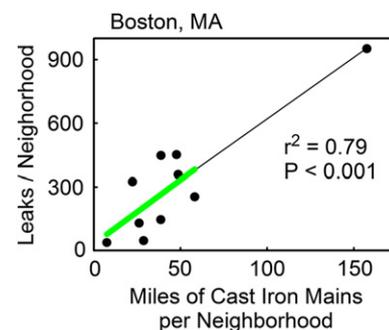
Fig. 2. Leak prevalence is associated with old cast iron pipes across ten Boston neighborhoods. (The combined line is the regression across all ten neighborhoods ($P < 0.001$); the green regression line [$r^2 = 0.34$; $P = 0.08$], which eliminates the influence of the leverage point [Dorchester neighborhood], has a slope and intercept indistinguishable ($P > 0.10$) from the combined regression.). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
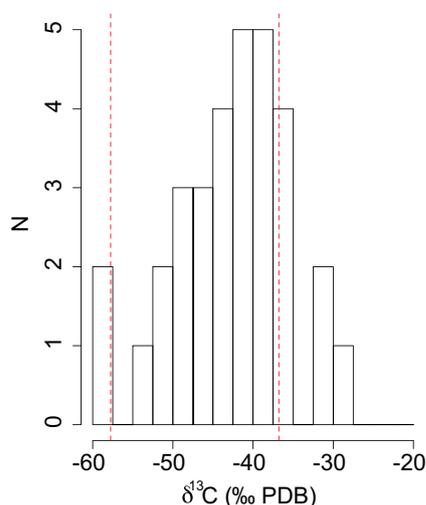
Fig. 3. $\delta^{13}CH_4$ of [CH_4] peaks detected in road surveys ($n = 32$). Red lines represent means of thermogenic ($-36.8‰$, $\pm0.7‰$ s.e., $n = 10$) and biogenic ($-57.8‰$, $\pm1.6‰$ s.e., $n = 8$) sources, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Leaks across Boston (Fig. 1), were associated primarily with cast iron mains that were sometimes over a century old (Fig. 2). Across ten Boston neighborhoods, leak frequency was linearly related to number of miles of cast iron mains ($r^2 = 0.79$, $P < 0.001$; Fig. 2), but only marginally to miles of non-cast-iron piping ($r^2 = 0.27$; $P = 0.12$, data not shown). Leak counts did not differ statistically by neighborhood or by socio-economic indicators for the neighborhoods obtained from the 2010 US Census ($P > 0.1$ for number of housing units and ethnicity) or the 2000 US Census ($P > 0.1$ for median income and poverty rate).

Reducing CH_4 leaks will promote safety and help save money. Although our study was not intended to assess explosion risks, we observed six locations where gas concentrations in manholes exceeded an explosion threshold of 4% [CH_4] at 20 °C (concentrations measured using a Gas Sentry CGO-321 handheld gas detector; Bascom-Turner, MA). Moreover, because CH_4, ethane (C_2H_6), and propane (C_3H_8) interact with NO_x to catalyze ozone formation, reducing these hydrocarbon concentrations should help reduce urban ozone concentrations and respiratory and cardiopulmonary disease (West et al., 2006; Shindell et al., 2012). CH_4 is also a potent greenhouse gas, with an estimated 20-year global warming potential 72 times greater than CO_2 (Alvarez et al., 2012; Townsend-Small et al., 2012). Replacing failing natural gas mains will reduce greenhouse gas emissions, thereby providing an additional benefit to the fewer mercury, SO_2 and particulate emissions that natural-gas burning emits compared to coal (Shindell et al., 2012). Finally, leaks contribute to $3.1 B of lost and unaccounted natural gas annually in the United States (EIA, 2012; 2005–2010 average).

Our ongoing and future research evaluates how surface [CH_4] values correspond to individual, and city-wide, urban leak rates and greenhouse-gas emissions. Two approaches to this question are useful: "bottom-up" chamber measurements taken on representative samples of individual leaks, and "top-down" atmospheric mass-balance estimates from rooftops of the collective urban leak rate that exploit the known isotopic signature of natural gas versus that of biogenic sources and other fossil fuel sources. The instrumentation used in this study is well-suited for both approaches.

We propose that a coordinated campaign to map urban pipeline leaks around the world would benefit diverse stakeholders, including companies, municipalities, and consumers. Repairing the leaks will bring economic, environmental, and health benefits to all.

Acknowledgments

Table 1
Locations and isotopic values from discrete street leak samples.

Latitude	Longitude	$\delta^{13}CH_4$ (‰ PDB)
42.3654	−71.0612	−53.959
42.3439	−71.2628	−47.898
42.3493	−71.2265	−57.590
42.3583	−71.1749	−40.818
42.3411	−71.2440	−37.323
42.3543	−71.2441	−38.241
42.3559	−71.1898	−39.412
42.3513	−71.2092	−41.978
42.3515	−71.2081	−39.531
42.3614	−71.2314	−41.796
42.3426	−71.1012	−44.100
42.3443	−71.0949	−41.848
42.3328	−71.0761	−37.516
42.3360	−71.0738	−46.414
42.3441	−71.0673	−45.490
42.3303	−71.0569	−37.476
42.3409	−71.0542	−40.029
42.3524	−71.0445	−43.127
42.3799	−71.0272	−48.182
42.3722	−71.0361	−57.693
42.3785	−71.0681	−48.429
42.3730	−71.0632	−37.471
42.3593	−71.0629	−42.689
42.3584	−71.0644	−52.033
42.3546	−71.1271	−47.241
42.2943	−71.1891	−52.028
42.2793	−71.1514	−37.648
42.2887	−71.1428	−32.467
42.3285	−71.0792	−28.251
42.3215	−71.0692	−36.214
42.3269	−71.0796	−30.662
42.3553	−71.0573	−43.836
	Mean	**−42.793**
	Standard error	**1.259**

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