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Methane Emissions from Natural Gas Infrastructure and Use in the Urban Region of Boston, Massachusetts

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Methane emissions from natural gas delivery and end use must be quantified to evaluate the environmental impacts of natural gas, and to develop and assess the efficacy of emission reduction strategies. We report natural gas emission rates for one year in the urban region of Boston, MA, using a comprehensive atmospheric measurement and modeling framework. Continuous methane observations from four stations are combined with a high-resolution transport model to quantify the regional average emission flux, 18.5 ± 3.7 (95% CI) g CH4 m-2 yr-1. Simultaneous observations of atmospheric ethane, compared with the ethane to methane ratio in the pipeline gas delivered to the region, demonstrate that natural gas accounted for > 60–100% of methane emissions, depending on season. Using government statistics and geospatial data on natural gas use, we find the average fractional loss rate to the atmosphere from all downstream components of the natural gas system, including transmission, distribution, and end use, was 2.7 ± 0.6% in the Boston urban region, with little seasonal variability. This fraction is notably higher than the 1.1% implied by the most closely comparable emission inventory.

Natural gas distribution | greenhouse gas emissions | cities

Atmospheric methane (CH4) is an important greenhouse gas (1) and major contributor to elevated surface ozone concentrations worldwide (2). Current atmospheric CH4 concentrations are 2.5 times greater than preindustrial levels due to anthropogenic emissions from both biological and fossil-fuel sources. The growth rate of CH4 in the atmosphere slowed beginning in the mid-1980s and plateaued in the mid-2000s, but growth has resumed since 2007. The factors responsible for the observed global increase and interannual trends, and the spatiotemporal distribution of sources remain uncertain (3).

Loses of natural gas (NG) to the atmosphere are a significant component of anthropogenic CH4 emissions (3), with important implications for resource use efficiency, worker and public safety, air pollution and human health (4), and for the climate impact of NG as a large and growing source of energy. A major focus area of the U.S. Climate Action Plan is reduction of CH4 emissions (5), but implementation requires identification of dominant source types, locations, and magnitudes. A recent review and synthesis of CH4 emission measurements in North America, spanning scales of individual components to the continent, found that inventory methods consistently underestimate CH4 emissions, that fossil fuels are likely responsible for a large portion of the underestimate, and that significant fugitive emissions may be occurring from all segments of the NG system (6).

The present study quantifies CH4 fluxes from NG in the urbanized region centered on Boston, Massachusetts, USA. Elevated CH4 concentrations in urban environments have been documented around the world for decades (7, Table S1) and attributed to a variety of anthropogenic source types. Recent studies of urbanized regions in California, using diverse atmospheric observing and modeling approaches, consistently found that CH4 emission rates were larger than those estimated by regional bottom-up inventories (8-12). In Boston, elevated CH4 concentrations have been observed at street-level and attributed to > 3,000 NG pipeline leaks from antiquated infrastructure (13), but associated CH4 emission rates were not quantitatively assessed.

In this study, we combine four key quantities in an atmosphere-based analytical framework: (1) atmospheric CH4 enhancements above background (∆CH4) were determined from measurements at a network of continuous monitoring stations, inside and upwind of the urban core (Fig. 1), for 12 months in 2012-13; (2) the NG fraction of the observed ∆CH4 was quantified for cool and warm seasons by measuring atmospheric ethane (C2H6), a tracer of thermogenic CH4, and comparing ratios of C2H6 and CH4 in the atmosphere and in the pipeline gas flowing through the region; (3) total CH4 emissions were derived from an atmospheric transport model, which quantitatively links surface fluxes with observed ∆CH4 using assimilated meteorology; and (4) the fraction of delivered NG lost to the atmosphere was estimated by comparing CH4 emissions to spatially explicit data on NG consumption. The result encompasses NG losses from the entire urbanized region, including emissions from NG transmission, storage, distribution, end use, and liquefied NG importation.

Methane Concentrations in the Boston Atmosphere

Atmospheric CH4 concentrations were measured continuously from September 2012 through August 2013 at two locations near the urban center (BU and COP) and two locations outside of Boston (HF and NHT) (Fig. 1, Table S2, SI Appendix 1). Background concentrations in air flowing into the city were estimated by randomly sampling from a range (5-35‰) of lower

Significance

Most recent analyses of the environmental impact of natural gas have focused on production, with very sparse information on emissions from distribution and end use. This study quantifies the full seasonal cycle of methane emissions and the fractional contribution of natural gas for the urbanized region centered on Boston, Massachusetts. Emissions from natural gas are found to be several times larger than predicted by existing inventory methodologies and industry reports. Our findings suggest that natural gas consuming regions may be larger sources of methane to the atmosphere than is currently estimated and represent areas of significant resource loss.

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Methane concentrations were measured with a laser spectrometer (15) at BU for three months in the fall and winter of 2012-13 and one month in the late spring of 2014 (Fig. S5). Covariances between observed variations in atmospheric CH$_4$ and CH$_4$ were determined from the daily slopes of a linear model that minimizes $\chi^2$ (16) of five-minute median afternoon data (Fig. 4, SI Appendix 2.1). The median of the daily slopes of atmospheric CH$_4$ versus CH$_4$ was 2.6 (2.5, 2.8) % during the cool season and 1.6 (1.4, 1.7) % during the warm season, obtained from days with a coefficient of determination ($R^2$) > 0.75 (~50% of the days).

The average C$_2$H$_6$ and CH$_4$ ratio in the NG flowing into the region during the two atmospheric measurement periods was 2.7 ± 0.0 % in the fall and winter of 2012-13 and 2.4 ± 0.1 % in the spring of 2014, determined from hourly gas quality data from the three pipelines that serve the region (17-18, Figs. S6 and S7, SI Appendix 2.2). The quotient of the C$_2$H$_6$ and CH$_4$ ratios in the atmosphere and pipeline demonstrates that NG contributed 98 (92, 105) % and 67 (59, 72) % of the ΔCH$_4$ in Boston in the cool and warm seasons, respectively. This result is insensitive to assumptions about the relative contribution of the three pipelines that supply the region and to the filtering criteria for the atmospheric data (SI Appendix 2.3).

### Methane and Natural Gas Emissions in Greater Boston

Methane enhancements were modeled at BU and COP with the Stochastic Time-Inverted Lagrangian Transport model (19), coupled to the Weather Research and Forecasting meso-scale meteorological model run at 1 km$^2$ grid resolution (WRF-STILT, 20, SI Appendix 3.1). WRF-STILT generates footprints (with units ΔCH$_4$ per unit surface flux, Fig. S8), which represent the sensitivity of each measurement point in space and time to upward surface fluxes. Both urban measurement sites were sensitive to emissions from the greater Boston region, with COP sensitive to a larger area than BU due to its higher altitude (Figs. 1 and S8, Table S2).

A spatially resolved (1 km$^2$) annual inventory of CH$_4$ emissions was constructed for the study region (SI Appendix 3.2, Fig. S12, Table S5) and combined with WRF-STILT footprints to generate a set of simulated ΔCH$_4$ values for each hour at each measurement station. The emission inventory was scaled for each season to equalize mean afternoon (11-16 h EST) modeled and observed ΔCH$_4$, providing optimized CH$_4$ emission rates for the region. Detailed methods and results for the model framework, including details on the emissions error quantification and results from alternative methodological approaches, are given in the SI Appendix, sections 3 and 4. Observation-model comparisons are shown in Figures 3, S13, and S14.

The mean annual optimized emission rate for the study area was 18.5 ± 3.7 g CH$_4$ m$^{-2}$ yr$^{-1}$ from all sources (Fig. 5A). Seasonal variations of total CH$_4$ emissions were modest, with fluxes in spring and summer marginally higher than in fall at the 95% confidence level (Fig. 5A). The weak seasonality of observed ΔCH$_4$ (Fig. 2) and the CH$_4$ flux rate is consistent with the finding that most of the emissions are from thermogenic gas, rather than biological processes, which would likely depend more strongly on season (21-22). When data from each urban site are analyzed independently, CH$_4$ emission results are equal within statistical uncertainty (Fig. S15), notwithstanding the large differences in ΔCH$_4$ (Fig. 2) and modeled footprints (Figs. 1, S8) between the two sites. This result provides strong support for the observation-model framework, which is further strengthened by the robustness of the emission result to adoption of different model frameworks (SI Appendix 4.2-4.3).
To assess the fraction of delivered NG emitted to the atmosphere, we constructed a spatially explicit estimate of NG consumption in the region (Fig 6, SI Appendix 3.2.1). Fractional loss rates for the region were obtained by multiplying optimized emissions by the fractional contribution of NG to the atmospheric signal, as indicated by the ethane tracer data, and dividing by the mean NG consumption in the region (Fig. 5AB). The inferred mean annual NG loss rate in the study area was 2.7 ± 0.6 % of the delivered gas in 2012-2013, with little seasonal dependence (Fig. 5C). Uncertainties in the average loss rates were calculated by summing in quadrature the relative errors for the average emissions, atmospheric NG fraction, and NG consumption terms (SI Appendix 3.2.1).

The modest seasonality of the inferred NG loss rate (Fig. 5C) is driven by the small seasonal variability in total NG consumption (Fig. 5B). Our analysis makes no assumptions about the relative contribution to emissions of specific NG consuming sectors or emission processes (SI Appendix 3.2.1), which could individually have very different loss rates than the aggregate estimate generated by this study. Our finding that the regional average NG emission rate was seasonally invariant may indicate that it does not strongly depend on the seasonally-varying components of the NG system, or could result from multiple compensating processes.

Comparison with Atmospheric Studies and Inventories

Two recent studies in Los Angeles covering ∼2 months provide the only previous atmosphere-based ("top-down") estimates of emissions from NG in an urban area, 1-2% (0.7-3% when accounting for the error ranges) of total NG consumed in the basin (10-11). However, attribution of CH₄ emissions to pipeline...
Fig. 4. Five-minute median atmospheric C$_2$H$_6$ and CH$_4$ measurement points at BU in fall and winter of 2012-13 (black) and spring of 2014 (blue), χ$^2$ optimization lines fit to each day (light lines), average fit lines for both seasons from all days with R$^2$ > 0.75 (bold solid lines), and lines with slopes of pipeline C$_2$H$_6$/CH$_4$ (dashed lines).

Fig. 5. Seasonal and annual average (±95% confidence intervals) (A) optimized CH$_4$ emissions in total and from NG, (B) NG consumption by sector, and (C) NG loss rates, derived from CH$_4$ concentration observations from the BU and COP sites together. Emission rates from each site individually are given in Fig. S15.

gas in Los Angeles is complicated by the presence of current and abandoned oil and gas wells, refinery operations, and natural CH$_4$ seeps, in addition to NG consumption. Other studies have estimated total CH$_4$ emission fluxes from a number of urban
areas around the world (Table S1), using atmospheric data-model frameworks of varying sophistication, but have not quantitatively attributed fluxes to NG. Our value for total CH₄ emissions in Boston is at the low end of the overall range of fluxes reported for other urban areas (Table S1), suggesting that total CH₄ emission rates in Boston are not anomalous.

The U.S. greenhouse gas (GHG) inventory (23) attributes 3302 Gg of CH₄ emissions to NG transmission, storage, and distribution, equal to ~0.8% of the NG delivered to consumers (24). The key input data for NG distribution systems in the national inventory are emissions factors developed from industry measurements (25) and activity data on miles of pipeline by material and counts of metering and regulating stations, customer meters, and pipeline maintenance events and mishaps (23). Emissions of NG in our study area are equal to ~9% of U.S. emissions attributed to distribution, transport, and storage, and ~24% of national emissions from distribution alone, a notably higher fraction than the ~3% of U.S. residential and commercial gas consumed in the study region. More detailed comparison of our results for the Boston urban region to the U.S. GHG inventory is not possible because the inventory is not spatially disaggregated.

Massachusetts has compiled a state GHG inventory (26, Table S4) using the same methods as the national inventory with state-level data, where available, and reports CH₄ emissions from NG systems equal to ~1.1% of NG consumed in the state. The larger loss fraction implied by the Massachusetts (~1.1%) versus the national (~0.7%) inventory is likely due to larger proportions of cast iron and bare steel pipelines (27), which have higher emission factors (23). Since most (68%) of our study region lies in Massachusetts, and most (88%) of the NG delivered in Massachusetts is consumed in the region, this value approximates the result that would be obtained by downscaling the national inventory to the study region. Our result for the NG loss fraction is ~2-3 times larger than that implied by the state inventory (although no uncertainty range is reported for the latter).

NG companies also report their GHG emissions and NG losses to public agencies. Methane emission and NG delivery data reported to both the U.S. Environmental Protection Agency (28) and Massachusetts GHG Reporting Programs (29) show NG loss rates of 0.4 to 1.6% among individual NG distribution companies in Massachusetts in 2012, with an average of 0.6%, weighted by delivered NG volumes. In contrast, data reported to the U.S. Energy Information Administration (30) for "losses from leaks, damage, accidents, migration and/or blow down" indicate a very small, nearly uniform loss rate of 0.4% in 2012 among all Massachusetts NG distribution companies.

Policy analyses of NG distribution emissions (31-32) sometimes use reported quantities of "lost and unaccounted-for" (LAUF) gas, an accounting term and cost-recovery mechanism reported by utilities to public utility commissions. LAUF fractions reported by individual distribution companies in Massachusetts in 2012 ranged from 0.1 to 10%, with a weighted average of 2.7% (30, 33). However, LAUF encompasses leaks, metering and accounting inaccuracies, and theft (34), and hence the relationship between LAUF and NG emissions is unknown.

Deficiencies in Existing Estimates

Several possible reasons may explain why existing methodologies predict lower CH₄ emissions from NG than we observe in the Boston urban region.

- Not all emission sources are inventoried. Namely, the U.S. and Massachusetts inventories (23, 26) do not include NG losses occurring downstream of customer meters, neither at large industrial facilities, nor in residential and commercial settings.

- Leak surveys are not comprehensive. Leak surveys (e.g. 13, 35-36) are based on detection of discrete, highly elevated atmospheric signals, expressed at accessible locations. Numerous small leaks can occur without posing a safety hazard while still contributing significantly to the total CH₄ source, and would require sensitive and accurate measurements for detection and quantification. Some NG leaks may be emerging in locations that are difficult to access (e.g. indoors, on private-property, through sewers or subway tunnels) with conventional surveys.

- Sampling protocols used to calculate emission factors have significant limitations. Due to practical constraints, NG emission factors are calculated from very small samples relative to the population they are intended to represent, and measurements are obtained from short-duration, non-repeated campaigns in a limited number of locations (25, 35). These limitations can lead to under-sampling of infrequent, high-emission events (6). Measurement of emissions from individual components requires access to restricted, privately-owned facilities, which could lead to sample bias (6), whether intentional or not. Inaccurate device and activity counts (6), and incomplete understanding of controlling variables, may lead to inappropriate extrapolation of emission factors in space and time. Data collected through new reporting requirements (37) may help address some of these limitations for specific devices and processes.

These issues arise from our fundamental lack of knowledge about the specific sources and processes responsible for the

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discrepancies found in this and other studies (6), and about the requirements for designing and testing a statistically rigorous inventory of emissions from the NG supply chain. Both high-emITTER into diffuse low-emission sources need to be sampled continuously or repeatedly to gain understanding of the true distribution of NG emissions. In addition to emission data, improved quantification of the fractional NG loss rate requires the compilation and availability of more rigorous, standardized, and detailed data on NG flows. Datasets should be spatially explicit to facilitate collation of disparate datasets and analysis of specific areas. Closer cooperation in data-sharing and synthesis and wide data dissemination are needed to better constrain CH4 emissions from NG and to provide the information needed to reduce those emissions.

Significance of Natural Gas Emissions

This study used one year of atmospheric CH4 measurements from a network of observing stations, a high-resolution modeling framework, atmospheric measurements of a NG tracer, and statistics on NG composition and consumption to quantify the NG emission rate for the Boston urban area as 2.7 ± 0.6 % (95% CI) Cambridge, MA, higher than that given by the most applicable (state) GHG inventory. The total volume of emitted gas in the study area over one year was ~15 billion standard cubic feet (scf), valued conservatively at ~$363,959 million (using 2012 city gate prices in Massachusetts and Rhode Island, 38), equal to ~6 scf person−1 day−1 (using the study area population of ~7.2 million, 39).

The U.S. President’s Methane Strategy (5) for reducing downstream NG emissions describes state and utility programs to accelerate infrastructure replacement, but offers no new federal initiatives for the distribution sector (40). A new Massachusetts law (41) is intended to improve the classification, reporting, and repair of NG leaks. The current study provides an example of a measurement-model framework that can be used to evaluate the effectiveness of programs aimed at reducing NG distribution emissions. More detailed measurements and inventories, following a more rigorous statistical design, are needed to fully characterize and prioritize the components, geographic areas, and supply-chain sectors that are contributing the most emissions. The full environmental benefits of using NG in place of other fossil fuels will only be realized through active measures to decrease direct losses to the atmosphere, including in receiving areas such as the Boston urbanized region.

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Supporting Information for:

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**S1 Measurement Methods**

**S1.1 Methane Measurements**

Methane was measured at four sites with four different models of Picarro Cavity Ring Down Spectrometers (1, Table S2). All models measured the same spectroscopic feature of CH₄. Carbon dioxide (CO₂) and water vapor (H₂O) (and carbon monoxide at COP only) were also measured at each site. A small fraction of CH₄ data points were removed when the instrument’s optical cell pressure and temperature were outside 0.1 torr and 0.005 °C of their set-points to ensure compatibility with the instrument’s spectroscopic fit parameters. An empirical H₂O correction equation (2) was used to calculate dry molar fractions of CH₄. Methane was measured continuously at intervals of 2-7 seconds, depending on the instrument model.

A two-point linear calibration equation was calculated for each instrument in the field. Compressed air cylinders with known, approximately ambient CH₄ concentrations (“surveillance standards”) were measured by each instrument for four minutes every eight hours (Fig. S3) to quantify the intercept (null value) of the calibration equation and to track long-term drift (Table S3). Data from the first ~90 seconds of each surveillance measurement period were disregarded to ensure the surveillance measurement had equilibrated. Measured offsets were smoothed over a multi-day moving window before being applied to correct ambient CH₄ measurements (Fig. S3).

All calibration and surveillance standards were manufactured by Scott-Marin (Riverside, CA) using natural air (3) and tied to scales defined by the National Oceanic and Atmospheric Administration (4) and the World Meteorological Organization (5). Surveillance standards were calibrated against NOAA primary standards before and after field deployment and were all found to have changed ≤ 0.2 ppb for CH₄ (Table S2). One surveillance tank was used at each site for the entire year of measurements.

Analytical uncertainty is approximated as the sum of measurement precision, uncertainty in calibration and surveillance tank values (Table S3), and uncertainty in the H₂O correction. Long-term drift was not included in the calculation of total analytical uncertainty because it was captured and corrected for in the data processing. Uncertainty in the H₂O correction equation was estimated by Rella et al. (6) as ±2 ppb at water vapor concentrations up to 3.4%, which sufficiently captures the maximum ambient measured water vapor concentration. Therefore total analytical uncertainty for one year of average hourly CH₄ measurements among the four sites was ≤ ~3 ppb (95% CI), < 0.2% of ambient concentrations.

**S1.2 Measurement Site Considerations**

Atmospheric measurements made in urban environments are vulnerable to disproportionate influence from nearby sources because large sources may occur at high densities in such environments. Furthermore, for practical reasons, urban measurements are often made from rooftops, which may not be optimally positioned to sample free-stream flow (7). The two urban measurement sites in this study (BU and COP) were located on the tops of buildings, so special precautions were taken to ensure that the data were not contaminated with signals from very-near sources, in particular rooftop building vents. The finding that the two urban measurement sites yielded the same total emission estimate for the region (main text) is a strong indicator that the average enhancements measured at both sites were not dominated by signals from very near sources.

The BU measurement was made from a 2-meter tower mounted in the center of the flat rooftop of a 6-story building. The building is located in a neighborhood comprised of both shorter and taller buildings, and a few small bathroom vents are located on the same roof. The measured C₃H₆/CH₄ in one of these vents was similar to that of pipeline gas, suggesting that the excess CH₄ in the vent derived largely from
NG leaks into the sewer system. Signals from the sewer vents on the BU roof were characterized by very short periods (1-3 s) of clearly elevated concentrations (“spikes”). During the afternoon hours used in the analysis, these spikes accounted for < 0.2% of the BU observations. These signals were eliminated from the BU data by trimming the lower and upper (to ensure no bias was imposed) 5% of the data in each hour prior to calculating hourly averages.

The COP site was located on a much taller building that stands well above surrounding structures, so contamination from nearby building sources was less of a concern. However, the building itself has many large vents on its rooftop for bathrooms, air heating and cooling systems, and a restaurant. Airflow over a building leads to the formation of small-scale turbulence and a zone of low pressure at the top of the building, which can entrain or aspirate contaminated air emitted from the building (7).

To avoid sampling air emitted from building vents at COP, four sample inlets were placed at the corners of the building, two stories below the top of the building, and each corner was sampled sequentially for five minutes each. The concept of the COP corner-sampling method is that, at any given time, at least one corner will represent uncontaminated, upwind conditions. The upwind corner(s) was selected as that with the lowest average concentration of CH$_4$, CO$_2$, or CO in each 20-minute, 4-corner sequence (Fig. S4). Equal lengths of tubing were used for the four sample lines to ensure equal delay times among them and a bypass pump continuously flushed all four sample lines.

Harvard Forest measurements were made from a tower in a mixed-deciduous forest in Petersham, Massachusetts (8). Methane was measured sequentially at eight heights (0.3, 0.8, 4.5, 7.5, 12.7, 18.3, 24.1, and 29 m) on the tower, for 4 minutes each and for 8 minutes at the highest (29 m) level. Methane concentrations were typically slightly depleted at the lowest measurement heights due to an oxidative soil sink (9). Only measurements from 29 m were used in this study.

To test whether the non-continuous sampling at COP and HF reduced the representativeness of the datasets, we simulated the COP sub-sampling routine with the continuous BU dataset by randomly sub-selecting one 5-minute period in each 20-minute interval. We found that hourly averages generated from the sub-selected dataset were not significantly different than hourly averages generated from the full dataset. Total data coverage for the year at the four sites was > 94%, with gaps due to maintenance, power outages, etc.

### S2 Ethane-Methane Ratio Analysis

**S2.1 Details of Atmospheric Analysis**

To quantify the relationship between the atmospheric C$_2$H$_6$ and CH$_4$ measurements, we used $\chi^2$ minimization (10, equation given below) of a straight-line fit (b=slope, a=intercept) to 5-minute medians of 1 hertz data points (Fig. 4, x and y), with errors in each variable at each time point (i=1:N) characterized by the standard error of the mean ($\sigma$).

$$\chi^2 (a, b) = \sum_{i=1}^{N} \frac{(y_i - a - b x_i)^2}{\sigma_{yi}^2 + b^2 \sigma_{xi}^2}$$

For the 2012-13 period, data from afternoon (11-17 h EST) hours only were used, but for the 2014 period, data from all hours were used because the C$_2$H$_6$ signal-to-noise ratio was smaller for the spring measurements (Fig. S5). Quantification of covariance (slope of the regression line) between C$_2$H$_6$ and CH$_4$ measurements from each day using short, 5-minute median, intervals was adopted to eliminate any
potential influence of building emissions (section 1.2) and because C$_2$H$_6$ measurements were not available at the background stations. If background C$_2$H$_6$ measurements had been available, then quantification of the atmospheric C$_2$H$_6$ to CH$_4$ covariance using longer intervals, and optimization of C$_2$H$_6$ emissions in an inversion framework, may have been possible. The current framework assumes that background concentrations did not vary substantially during individual days, supported by the tight correlation ($R^2 > 0.75$) between observed C$_2$H$_6$ and CH$_4$ on approximately half of the days. Days with large shifts in wind direction sometimes did not have consistent C$_2$H$_6$/CH$_4$ and were rejected by the $R^2 > 0.75$ criterion. We used daily fits to 5-minute intervals to calculate the average ratio in order to limit the influence of possible autocorrelation between points from individual hours or days. 95% confidence intervals were calculated by bootstrapping daily fit slopes with 1,000 iterations and sampling with replacement.

S2.2 Details of Pipeline Analysis

Three major pipelines, Tennessee (TGP), Algonquin (ALG), and Maritimes and Northeast (MNE), and a liquefied natural gas (LNG) import terminal, supply NG to the Boston region (Fig. S6). Hourly gas quality data were collected from the informational postings for each pipeline (11-12). Daily median C$_2$H$_6$ and CH$_4$ ratios were calculated for each pipeline using hourly data (Fig. S7) from the gas quality measurement stations closest to Boston (Fig. S6). The three pipelines delivered the following fractions of NG consumed in Massachusetts in 2012: 65% TGP, 30% ALG, and 5% MNE (13). Seasonal average pipeline C$_2$H$_6$ and CH$_4$ ratios were calculated for the same time period as the atmospheric ratios using the daily median pipeline ratios, weighted by the fractional contributions of each pipeline. 95% confidence intervals were estimated by bootstrapping daily weighted median ratios with 1,000 iterations and sampling with replacement.

Gas composition in the pipelines is measured using industry standard methods (14-15), but uncertainties due to sampling and measurement error are not reported. Additionally, the representativeness of the measured relative to the lost gas is not known. The approach described above to estimate the mean pipeline C$_2$H$_6$ and CH$_4$ ratio is intended to yield an aggregate estimate that is robust to sporadic erroneous and/or unrepresentative measurements.

S2.3 Robustness of Results to Assumptions

Central C$_2$H$_6$/CH$_4$ values calculated from both the atmospheric and pipeline data were insensitive to the underlying assumptions. Slopes from $\chi^2$ optimization (10) were 10-15% larger than those obtained from an ordinary least squared regression because variance in the CH$_4$ measurements cannot be neglected. The atmospheric ratios reported in the main text were based on daily fits to data with $R^2 > 0.75$ and from afternoon hours only (for the 2012-13 period only). The same calculation using five-minute means instead of medians, a less stringent $R^2$ cutoff, and data from all hours instead of afternoon hours yielded both larger and smaller ratios that varied by < 10% and were not significantly different from those reported in the main text.

We believe the relative contribution of NG to CH$_4$ emissions observed in the late spring of 2014 can be applied to the spring and summer months of the year prior because the average air temperature and observed ΔCH$_4$ in May-June were very similar between 2013 and 2014 and because the observation interval approximates the midpoint of the March-August period for which we adopted the observed NG fraction.

The pipeline C$_2$H$_6$/CH$_4$ reported in the main text was based on a weighted average between the three pipeline companies with contributions of 65% for TGP, 35% for ALG, 5% for MNE. However, these fractional contributions are only valid for the entire state of Massachusetts in 2012, and we have no
information about the relative sensitivity of the atmospheric measurements to the three pipelines and individual gas quality measurement locations. Therefore, we also calculated the pipeline ratio using equal contributions from TGP and ALG, and no contribution from MNE. This result was not significantly different than the ratio given in the main text. The high correlation between atmospheric C\textsubscript{3}H\textsubscript{6} and CH\textsubscript{4}, and the close correspondence of the atmospheric and pipeline gas ratios, support the hypothesis that NG was the major source of enhancements for both gases.

S3 Modeling Framework Description

S3.1 WRF-STILT Configuration

The Stochastic Time-Inverted Lagrangian Transport (STILT v656, 16) model was driven with customized meteorological fields from the advanced research version of the Weather Research and Forecasting Model (WRF v3.4.1, 17-18). Meteorological fields were generated at four gridded horizontal resolutions (1, 3, 9, and 27 km) in a two-way nested arrangement centered on Boston (Fig. S9). All WRF domains had 42 vertical levels. Initial and lateral boundary conditions were provided by the North American Regional Reanalysis (19). Overlapping 30-hour forecasts were initialized every 24 hours, at 00 UTC, and the first 6 hours of each forecast were discarded to allow for spinup. Grid nudging was used in the outer-most domain only and not within the planetary boundary layer (PBL). Additional details of the WRF configuration used in this study are given in Nehrkorn et al. (20, case “Turb-U”). Section 4.1 describes comparisons between WRF-simulated and measured meteorological parameters.

STILT was run in time-reverse mode in which an ensemble of 500 particles was released every hour from each of the urban measurement sites and transported backward in time for 10 days according to the WRF meteorology. The majority of particles reached the study boundary (Fig. 1, Fig. S8) in < 8 hours and the median travel time was < 3 hours. Background values generated from NHT measurements were assigned to particles that exited the coastal portion (at 20-140°) of circular boundary (~22% of particles) and background values from HF were assigned to all other particles.

S3.2 Prior Inventories

S3.2.1 Natural Gas Consumption

In order to understand emissions results as a fractional loss rate of NG delivered within the modeling framework, it was necessary to create a spatially explicit map of NG consumption. Consumption is an appropriate estimator of net gas flows through the study area because all of the pipelines entering the study area terminate inside or very near the study area boundary (Fig. S6). Reports of NG consumption by state, month, and sector were obtained from the Energy Information Administration (EIA, 21) for the study area and time period. Missing numbers were estimated as the average of the 2008-2011 data for the same state, month and sector, accounting for 20% of the gas consumed in the study region. Volumes of NG were converted to masses of CH\textsubscript{4} using the ideal gas law by assuming industry standard temperature and pressure (60 °F and 1 atm) and 97% CH\textsubscript{4} content, as reported in the gas quality measurements (section 2.2, 11-12), giving 1 scf NG = 1.16 moles CH\textsubscript{4} = 18.6 grams CH\textsubscript{4}.

Natural gas consumption is reported by the EIA for four main sectors: power production, industrial, commercial, and residential. All sectors were included in the consumption estimate because we were not able to distinguish emissions from individual NG sectors with the atmospheric data. Monthly consumption by the electric power sector was spatially allocated to individual power production facilities using the EIA data on monthly fuel consumption by plant (22) to identify and locate each facility (Fig. S10). Consumption by the residential and commercial sectors was spatially allocated using a parcel-level
database for Massachusetts of residential and commercial building square footage and the fuel type used in each building for space and water heating (23). This dataset was constructed from multiple state and local government data sources such as Registry of Deeds, Land Court data, Town Clerk data and tax assessor information. Industrial sector NG consumption was spatially allocated using commercial building square footage data because no spatial information was available for the industrial sector. For the study area outside of Massachusetts, census data on the number of housing units with NG (24) were used to allocate residential, commercial, and industrial sector NG consumption at the blockgroup scale. Within Massachusetts, the R^2 value for the 1 km^2 gridded census and square footage datasets was 0.8 for residential buildings and 0.7 for residential and commercial buildings together, demonstrating that the census data were a reasonable substitute for the square footage data where the former was not available. Figure S11 gives monthly average NG consumption by sector for Massachusetts and for the 90-km radius study area, as spatially allocated using the methods described above.

Uncertainty estimates for state monthly NG consumption are supplied by EIA (25). Monthly errors for Massachusetts are available for 6 of the 12 months in the study period, with NAs reported for the remaining months, and range from ± 0.4 to 1.1 %. Using the largest monthly error value and summing them in quadrature leads to a conservative error estimate of ± 1.9 % for three-month seasonal totals and ± 3.8 % for the annual total. We did not estimate uncertainties in spatial allocation of state total consumption to the study area, nor uncertainties in spatial distribution within the study area, because no independent dataset is available for comparison and because it is unknown how well the spatial distribution of consumption approximated the spatial distribution of emissions.

S3.2.2 Methane Emissions

Two spatially-resolved inventories of CH_4 emissions, EDGAR v4.2 FT2010 (26) and one created for this study (Table S4), were tested as a priori inputs to the modeling framework. EDGAR is a global product that uses simplified methods (e.g. scaling by population density) to spatially disaggregate emissions. To take advantage of locally available data and knowledge, a customized inventory with five anthropogenic and biogenic source categories (described below) and 1 km^2 spatial resolution was developed for the study domain (Fig. S12). The custom inventory was not meant to be exhaustive, but rather was created to provide detailed emission estimates for key sectors with improved spatial resolution and accuracy. Emission results using the customized inventory are given in the main text. Emission results using EDGAR are discussed in section 4.3 and shown in Fig. S18. Both inventories were adopted as temporally invariant.

Wetlands. Data on wetlands’ location, size, and type were obtained from the National Wetlands Inventory (27) for the four states in the study region. These wetland inventories are based on aerial photography and have a mapping unit of 0.4 – 1.2 hectares. Average CH_4 emission rates for each type were taken from Bridgham et al. (28), which calculated mean emission rates from > 100 studies. An average emission rate of 7.6 g CH_4 m^{-2} year^{-1} was applied to the freshwater wetlands (emergent and forested/shrub) and an emission rate of 1.3 g CH_4 m^{-2} year^{-1} was applied to saltwater wetlands (estuarine and marine). Areas of open water (rivers, lakes, deepwater marine) were not included in the wetlands emission layer due to a lack of data on emission rates from these areas. The total wetland area in our domain is 1,900 km^2, ~11% of the land area.

Enteric Fermentation. Methane emissions from ruminant livestock were spatially allocated to counties according to county-level headcounts of cattle and calves from the USDA 2007 Agricultural Census (29). An emission factor of 117 kg CH_4 head^{-1} year^{-1}, the EPA emission factor for mature dairy cattle in the North Atlantic Region (30), was multiplied by the cattle count in each county to yield a total average emission rate. Emissions from animals other than cattle were not included because cattle accounted for the majority of livestock emissions in the study area.
Transportation. Methane emissions from transportation were estimated using per-mile emission factors by vehicle type and model year (31-32), state-level data on vehicle fleet composition (33), and a database of vehicle miles traveled per road (34). Gately et al. (34) provide a complete methodological description for an analogous emissions model for CO₂. This layer does not include emission estimates from CNG vehicles, of which there are ~2,000 in Massachusetts (35).

Point-sources. Facility-level data reported to the EPA GHG Reporting Program (36) for 2011 were used to represent CH₄ emissions from the largest point sources, including landfills and industrial facilities. Reported locations of landfills were visually checked using aerial photographs and adjusted if necessary to ensure that emission points were located at landfills rather than at associated administrative offices.

Natural Gas Leaks. In the prior inventory, we estimated CH₄ emissions from NG losses to be 2% of the NG consumption prior (section 3.2.1). The prior emissions inventory was not used to determine the fractional contribution of the NG, nor any other, source sector to total CH₄ emissions because the C₂H₆ measurements provided definitive attribution of CH₄ emissions from NG.

S3.3 Error Estimation for Optimized Methane Emissions

Data points with model-data residuals > 3-σ of the residual distribution (< 5% of points for any individual site and season) were excluded from the emission scaling factor calculations (Fig. S14). Exclusion of outliers had no impact on the mean emission result, but led to slightly smaller confidence intervals (Fig. S18, pt. 0 vs. pt. 1) and larger R² values for the optimized data-model fits (Fig. S14).

Means and 95% confidence intervals on all reported estimates of ΔCH₄ and of CH₄ fluxes were generated through an end-to-end bootstrap analysis with the following steps. Distributions of possible background CH₄ concentrations at the two exterior sites (HF and NHT) were generated at 1-percentile increments between the 5th and 35th percentiles, over a 48-hour moving window. The lower percentile and moving window approach was employed to capture synoptic-scale variability and because near-surface nighttime observations are often affected by small nearby sources due to stratification of the nocturnal boundary layer which traps emissions near the ground. In the bootstrap, background distributions were randomly sampled each day to calculate ΔCH₄,obs. Average hourly afternoon CH₄,obs and ΔCH₄,mod values were randomly sampled separately for each day to generate average daily values of observed and modeled ΔCH₄. Daily average ΔCH₄ values were randomly sampled to generate seasonal average ΔCH₄ and inventory scaling factors to derive average CH₄ emissions. Lastly, seasonal average ΔCH₄ and CH₄ emissions were randomly sampled to generate annual averages of each. Each of these steps was performed 1,000 times and means and confidence intervals were calculated from the resulting distributions.

S4 Robustness of Emission Results

S4.1 WRF Validation

WRF-simulated meteorological fields were compared against available meteorological measurements at National Weather Service observing sites (37) using the Model Evaluation Tools verification software (38) from the National Center for Atmospheric Research. Figure S16 shows summary statistics of average near-surface temperature and wind speed biases and root mean square errors for the year of simulations and each surface station used in the verification. For most land-based stations, WRF wind speeds were biased slightly high and temperatures were biased slightly low. Examination of the average diurnal cycle (Fig. S17) reveals that the wind speed bias is present at all hours of the day, whereas the temperature bias...
is due largely to stronger than observed nocturnal cooling. The latter finding is consistent with overestimated nocturnal low-level stability, which likely contributed to the over-prediction of nighttime CH₄ concentrations at BU (Figs. 3C, S13), typically located below the nocturnal boundary layer, and the modest under-prediction of nighttime concentrations at COP (Figs. 3A, S13), which was often above the nocturnal boundary layer. The emissions estimate was derived from afternoon data only and thus not affected by this model bias.

**S4.2 Simple Mass Balance Model**

Methane enhancements in the urban core reflect the accumulation of emission inputs into the PBL during transit from the 90-km radius model boundary to the observation point (Fig. 1), less exchange by vertical or horizontal mixing with background air (e.g., from above the PBL, or via a sea breeze circulation). We checked our posterior emission fluxes for consistency with this basic mass balance concept using the HYSPLIT back-trajectory model (39) driven by meteorology from the North American Model mesoscale forecast system with 12 km resolution (40, NAM12).

The model equation for the mass balance model is:

\[ Z_{\text{PBL}} \Delta C_{\text{H}_4} \bar{N}_{\text{air}} = < F_{\text{CH}_4} > \tau_{\text{transit}} - K \Delta_n \text{CH}_4 \bar{N}_h \tau_{\text{transit}} \]

where \( Z_{\text{PBL}} \) is the depth of the mixed layer (m), \( \Delta C_{\text{H}_4} \) is the mean enhancement (mole fraction) in the PBL, \( \bar{N}_{\text{air}} \) is the average air number density in the PBL (m⁻³), \( < F_{\text{CH}_4} > \) is the mean surface flux (m² s⁻¹), and \( \tau_{\text{transit}} \) is the transit time (s) within the PBL. The last term in the equation represents column-integrated exchange with background air during transit, where \( K \) is the exchange velocity (m s⁻¹), \( \Delta_n \text{CH}_4 \) is the concentration difference (mole fraction), and \( N_h \) is the mean number density (m⁻³) for the exchange process.

The mass balance model was used to simulate \( \Delta \text{CH}_4 \) at COP for afternoon hours (17-21 UTC) in September and October, 2012 using values for \( Z_{\text{PBL}} \) and \( \tau_{\text{transit}} \) from HYSPLIT/NAM12. The boundary layer in most of these simulations was well-developed with transit times to the model boundary between 2 and 8 hours for > 80% of the trajectories. Unlike WRF-STILT, HYSPLIT back trajectories do not simulate exchange and the last term of the model equation was therefore neglected.

This mass balance model and our optimized fluxes yielded mean \( \Delta \text{CH}_4 \) values that were ~25% higher than both observed and simulated values from WRF-STILT. This small excess is consistent with the absence of an entrainment term in the box model. We conclude that the optimal fluxes from WRF-STILT reported in the main text are consistent with analysis using a simple mass balance approach based on independent meteorology and transport models.

**S4.3 Modeling Framework Variants**

The sensitivity of the emissions result was tested against several variants of the modeling framework (Fig. S18). Optimized emissions resulting from the use of EDGAR (26) and the custom inventory at a coarser spatial resolution were not significantly different than the main result which used the custom emission inventory (section 3.2.2, Fig. S18 pts. 2-3). We also tested the null hypothesis for spatial variation of sources, using an inventory of uniform, constant flux over land and zero flux over water. This variant resulted in similar model-data correlations (Fig. S14) as the spatially-varying inventories (section 3.2.2), but mean footprint-weighted, optimized emission fluxes differed by ~30% between the two sites. Therefore, we rejected the null hypothesis of no significant spatial variation in emissions and adopted the custom prior emission field (section 3.2.2) instead.
Measured CH₄ enhancements presented in the main text (Fig. 2) were aggregated into daily points by taking afternoon (11-16 h EST) averages. This aggregation method was tested by also averaging the four lowest hourly observations and model enhancements in the period of 9-18 h EST each day (Fig. S2, Fig. S18 pt. 4). Both of these approaches to data selection and aggregation aim to focus the analysis on periods when the atmosphere is well-mixed and when the data are less variable, which maximizes the areal representativeness of the results and minimizes the influence of a possible model biases in boundary layer height. Both aggregation methods yielded comparable results.

References

43. California Air Resources Board 2005 Almanac, Appendix D.
**Fig. S1.** Mean hourly CH$_4$ concentrations in the empirical background and measured at BU and COP from September 1, 2012 through August 31, 2013.

**Fig. S2.** Mean hourly measured CH$_4$ concentrations (black lines), the range of empirical background concentrations from upwind stations (red lines), and the daily afternoon average (green) and minimum (blue) points that represent mean enhancements at BU and COP, from an example period of three months in 2013.
**Fig. S3.** CH$_4$ calibration intercepts, calculated as the difference between known and measured surveillance standard values for the four instruments over one year. Black points are average measured values for each ~2.5 minute measurement period and the red line is the smoothed intercept used to correct the data. All plots were shifted to have zero mean offsets to aid visualization of the drifts.
Fig. S4. Example of the COP corner-sampling method for three days in October, 2012. The colored lines show average concentrations during each 5-min sampling interval from each corner. The black line shows the average hourly inferred upwind concentration. The legend describes the orientation of each corner.
Fig. S5. Five-minute median $\text{C}_2\text{H}_6$ and $\text{CH}_4$ measurements from BU.

Fig. S6. Approximate locations of the three interstate gas pipelines (Tennessee – TGP, Algonquin – ALG, and Maritimes and Northeast – MNE, 41) serving Boston and the surrounding area, the gas quality measurement stations used in this study, and the LNG import terminal. The gray circle is the study area boundary.
Fig. S7. Stacked histograms of (ab) hourly and (cd) daily median ratios of C$_2$H$_6$ and CH$_4$ in the pipeline gas during the two time periods of the atmospheric C$_2$H$_6$ measurements (Fig. S5). (ab) Hourly data were obtained from the three pipelines (Tennessee – TGP, Algonquin – ALG, and Mariti – MNE, 12-13) for the stations shown in Fig. S6. (cd) Daily median ratios for each pipeline were used to estimate a mean ratio for each of the two time periods, weighted by the volumes delivered by each pipeline to Massachusetts. Each of the three pipelines is equally represented in the daily median plots, but not the hourly plots because hourly data coverage varied between stations.
**Fig. S8.** Average afternoon (11-16 h EST) footprints for the BU and COP measurement sites and the study year.

**Fig. S9.** Location of the four nested WRF domains. The horizontal resolutions of the largest to smallest domains are 27, 9, 3, and 1 km.
**Fig. S10.** Location and 2012 gas consumption (units: $10^6$ scf yr$^{-1}$) of each natural-gas fueled utility power plant (22) in the four states included in the study area, as delineated by the gray circle.

**Fig. S11.** Average monthly NG consumption by sector (a) in Massachusetts (21) and (b) in the study area, spatially allocated as described in section 3.2.1. Commercial and industrial sectors are aggregated in panel b because spatial information on industrial sector emissions was not available.
Fig. S12. Emission (g CH$_4$ m$^2$ yr$^{-1}$) maps by source type and in total on a 1 km$^2$ grid from the inventory constructed for this study. Scale bars for individual sectors are not linear and were set to have an equal sample size in each bin in order to better render spatial patterns. The scale bar for total emissions is linear. The methods used to construct these maps are described in section 3.2.2.
Fig. S13. Observed, optimized modeled, and background CH₄ concentrations averaged by hour of the day for the two sites and four seasons. The horizontal hatched area shows the average range of sampled background concentrations, derived from 5-35th percentiles of the background station data. The gray vertical shaded area indicates the afternoon model optimization period, 11-16 h EST (UTC-5). Although both high and low biases exist in the nighttime model data, modeled and observed data from the afternoon optimization window are in good agreement.
Fig. S14. Optimized modeled daily versus observed average afternoon CH$_4$ concentrations for the two sites and four seasons. The gray line is the one-to-one line. Outlying points marked by crosses were excluded from model optimization.
Fig. S15. Optimized $\text{CH}_4$ emissions seasonally and annually, derived from $\text{CH}_4$ observations from BU and COP individually.
Fig. S16. Average (ac) bias and (bd) root mean square error (RMSE) of WRF-simulated (ab) near-surface temperature and (cd) wind speed for National Weather Service stations (37) in the innermost WRF domain.
Fig. S17. Observed (red) and WRF-simulated (blue) near-surface (ab) temperature (cd) and wind speed, averaged by hour (UTC) for one year and stations (ac) KBOS (at Boston Logan airport) and (bd) KBED (northwest of Boston).
Fig. S18. Mean annual emission results from the main configuration (pt. 0) and several variants of the modeling and analysis framework (pts. 1-4). The emission estimate presented in the main text (pt. 0) resulted from the custom emission inventory at 1 km² spatial resolution, data aggregation into daily afternoon (11-16 h EST) points, and removal of extreme outliers. Point 1 shows the emission results when outliers were not removed. Point 2 shows the emission result from EDGAR (26) instead of the custom emission inventory (section 3.2.2). Point 3 shows the result when the custom inventory was aggregated to a coarser spatial resolution. Point 4 shows the emission result from aggregating the four lowest hourly observed concentrations and model enhancements in the period of 9-18 h EST each day instead of afternoon hours (Fig. S2).
Table S1. Methane emissions in urban areas from atmosphere-based (“top-down”) studies. Only studies that reported emission rates averaged in time and space are listed.

<table>
<thead>
<tr>
<th>Study</th>
<th>Location</th>
<th>Measurement year</th>
<th>Emission Rate (g CH$_4$ m$^{-2}$ yr$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>44</td>
<td>Nagoya, Japan</td>
<td>1990-91</td>
<td>7</td>
</tr>
<tr>
<td>45</td>
<td>Midwest town, USA</td>
<td>1991</td>
<td>55</td>
</tr>
<tr>
<td>46</td>
<td>Two towns in East Germany</td>
<td>1992</td>
<td>12, 60</td>
</tr>
<tr>
<td>47</td>
<td>North Britain</td>
<td>1994</td>
<td>28 – 56</td>
</tr>
<tr>
<td>48</td>
<td>Heidelberg, Germany</td>
<td>1995-97</td>
<td>8 ± 2</td>
</tr>
<tr>
<td>49</td>
<td>Krakow, Poland</td>
<td>1996-97</td>
<td>20</td>
</tr>
<tr>
<td>50</td>
<td>St. Petersburg, Russia</td>
<td>1996-2000</td>
<td>32 ±9</td>
</tr>
<tr>
<td>51</td>
<td>Beijing, China</td>
<td>2000</td>
<td>50</td>
</tr>
<tr>
<td>52</td>
<td>Los Angeles County, CA, USA</td>
<td>2007-08</td>
<td>205 ± 6†</td>
</tr>
<tr>
<td>53</td>
<td>South Coast Air Basin, CA, USA</td>
<td>2007-08</td>
<td>228 ± 38*</td>
</tr>
<tr>
<td>54</td>
<td>Indianapolis, IN, USA</td>
<td>2008</td>
<td>71 ± 50</td>
</tr>
<tr>
<td>55</td>
<td>South Coast Air Basin, CA, USA</td>
<td>2010</td>
<td>167 ± 57*</td>
</tr>
<tr>
<td>56</td>
<td>South Coast Air Basin, CA, USA</td>
<td>2010</td>
<td>156 ± 14†</td>
</tr>
<tr>
<td>57</td>
<td>South Coast Air Basin, CA, USA</td>
<td>2010</td>
<td>127 ± 21*</td>
</tr>
<tr>
<td>58†</td>
<td>South Coast Air Basin, CA, USA</td>
<td>2010</td>
<td>160 ± 30†</td>
</tr>
<tr>
<td>58‡</td>
<td>South Coast Air Basin, CA, USA</td>
<td>2010</td>
<td>118 ± 30*</td>
</tr>
<tr>
<td>59</td>
<td>Florence, Italy</td>
<td>2011</td>
<td>58</td>
</tr>
<tr>
<td>60</td>
<td>London, UK</td>
<td>2012</td>
<td>66 ± 10</td>
</tr>
</tbody>
</table>

*Basin-total fluxes reported in the California studies (*) were converted to average area fluxes using areas from the California Air Resources Board (43).
†Aircraft observations
‡Satellite observations
Table S2. Locations and instrument models of the four measurement sites.

<table>
<thead>
<tr>
<th>Site</th>
<th>Longitude (°)</th>
<th>Latitude (°)</th>
<th>Height (m above ground)</th>
<th>Picarro model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Boston University (BU)</td>
<td>-71.10</td>
<td>42.35</td>
<td>29</td>
<td>G2101-i</td>
</tr>
<tr>
<td>Copley Square (COP)</td>
<td>-71.08</td>
<td>42.35</td>
<td>215</td>
<td>G2401</td>
</tr>
<tr>
<td>Harvard Forest (HF)</td>
<td>-72.17</td>
<td>42.54</td>
<td>29</td>
<td>G2301</td>
</tr>
<tr>
<td>Nahant (NHT)</td>
<td>-70.91</td>
<td>42.42</td>
<td>16</td>
<td>ESP-1000</td>
</tr>
</tbody>
</table>

Table S3. Methane measurement performance statistics for the four sites and one year. Individual measurement precision is the average standard deviation of the raw 2-6 second surveillance measurements made throughout the year at each site (Fig. S3). Surveillance tank uncertainties derive from both uncertainties in the primary NOAA calibration tanks (4) against which they were calibrated and from the measurement precision of the instrument used to calibrate the tanks. The surveillance tank at BU is still in use and has not yet been returned for a second, post-deployment calibration.

<table>
<thead>
<tr>
<th>Site</th>
<th>Long-term Instrument Drift (ppb)</th>
<th>Individual Measurement Precision (1σ) (ppb)</th>
<th>Surveillance Tank Uncertainty (1σ) (ppb)</th>
<th>Surveillance Tank Long-term Drift (ppb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BU</td>
<td>10.5</td>
<td>0.9</td>
<td>0.1</td>
<td>--</td>
</tr>
<tr>
<td>COP</td>
<td>2.8</td>
<td>0.3</td>
<td>0.2</td>
<td>0.1</td>
</tr>
<tr>
<td>HF</td>
<td>6.4</td>
<td>0.3</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>NHT</td>
<td>1.7</td>
<td>0.3</td>
<td>0.1</td>
<td>0.2</td>
</tr>
</tbody>
</table>
Table S4. Average CH₄ emission in Massachusetts and in the 90-km radius study area (Fig. 1) by sector and in total from three emissions inventories – the custom inventory (section 3.2.2), EDGAR v4.2 FT2010 (23), and the Massachusetts GHG inventory (42). The Massachusetts inventory was not tested in the modeling framework because it is not spatially resolved.

<table>
<thead>
<tr>
<th>Inventory</th>
<th>Sector</th>
<th>Average Emissions (g CH₄ m⁻² yr⁻¹) (% of total)</th>
<th>Massachusetts</th>
<th>Study Area</th>
</tr>
</thead>
<tbody>
<tr>
<td>Custom*</td>
<td>Wetlands</td>
<td>0.93 (16%)</td>
<td>1.02 (14%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Enteric Fermentation</td>
<td>0.25 (4%)</td>
<td>0.22 (3%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Transportation</td>
<td>0.05 (1%)</td>
<td>0.06 (1%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Large Point Sources (Power plants, Industrial, Landfills, Wastewater)</td>
<td>1.13 (19%)</td>
<td>1.43 (20%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Natural Gas</td>
<td>3.56 (60%)</td>
<td>4.49 (62%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>5.92</td>
<td>7.22</td>
<td></td>
</tr>
<tr>
<td>EDGAR (2010)</td>
<td>Energy production (1A1_1A2)</td>
<td>0.17 (2%)</td>
<td>0.23 (2%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Non-road Transportation (1A3a_c_d_e)</td>
<td>0</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Road Transportation (1A3b)</td>
<td>0.05 (1%)</td>
<td>0.06 (1%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Stationary Combustion (1A4)</td>
<td>0.28 (4%)</td>
<td>0.38 (4%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Fugitive from Solid Fuels (1B1)</td>
<td>0</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Oil Production &amp; Refining (1B2a)</td>
<td>0.12 (2%)</td>
<td>0.15 (2%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Natural Gas Production &amp; Distribution (1B2b)</td>
<td>3.50 (46%)</td>
<td>4.76 (59%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Industrial (2)</td>
<td>0.03</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Enteric Fermentation (4A)</td>
<td>0.15 (2%)</td>
<td>0.14 (1%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Manure Management (4B)</td>
<td>0.05 (1%)</td>
<td>0.05</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Agricultural Soils (4C_4D)</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Agricultural Waste Burning (4F)</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Solid Waste Disposal (6A_6C)</td>
<td>2.13 (28%)</td>
<td>2.44 (35%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Waste Water (6B)</td>
<td>1.04 (13%)</td>
<td>1.44 (15%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Fossil Fuel Fires (7A)</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>7.52</td>
<td>9.69</td>
<td></td>
</tr>
<tr>
<td>Massachusetts State (2011)</td>
<td>Stationary Combustion</td>
<td>Residential</td>
<td>0.19 (2%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Commercial</td>
<td>0.05 (1%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Industrial</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Electric Power</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mobile</td>
<td></td>
<td>0.30 (3%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Natural Gas Transmission &amp; Distribution</td>
<td>3.73 (42%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Enteric Fermentation</td>
<td>0.18 (2%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Manure Management</td>
<td>0.05 (1%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Landfills &amp; Waste Combustion</td>
<td>3.49 (39%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Wastewater</td>
<td>0.95 (11%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>8.96</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*The custom inventory covers the majority, but not the entire state of Massachusetts (Fig. S12), so the average emissions rate for Massachusetts was calculated from the area of the state that is covered by the inventory.