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 1 y in the urban region of Boston, 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% confidence interval) g $CH_4 \cdot m^{-2} \cdot y^{-1}$. 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 | methane

Atmospheric methane (CH_4) is an important greenhouse gas (1) and major contributor to elevated surface ozone concentrations worldwide (2). Current atmospheric CH_4 concentrations are 2.5 times greater than preindustrial levels due to anthropogenic emissions from both biological and fossil fuel sources. The growth rate of CH_4 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).

Losses of natural gas (NG) to the atmosphere are a significant component of anthropogenic CH_4 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 US Climate Action Plan is reduction of CH_4 emissions (5), but implementation requires identification of dominant source types, locations, and magnitudes. A recent review and synthesis of CH_4 emission measurements in North America, spanning scales of individual components to the continent, found that inventory methods consistently underestimate CH_4 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 CH_4 fluxes from NG in the urbanized region centered on Boston. Elevated CH_4 concentrations in urban environments have been documented around the world for decades (7) (*SI Appendix*, 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 CH_4 emission rates were larger than those estimated by regional bottom-up inventories (8–12). In Boston, elevated CH_4 concentrations have been observed at street level and attributed to >3,000 NG pipeline leaks from antiquated infrastructure (13), but associated CH_4 emission rates were not quantitatively assessed.

In this study, we combine four key quantities in an atmosphere-based analytical framework: (*i*) atmospheric CH_4 enhancements above background (ΔCH_4) were determined from measurements at a network of continuous monitoring stations, inside and upwind of the urban core (Fig. 1), for 12 mo in 2012–2013; (*ii*) the NG fraction of the observed ΔCH_4 was quantified for cool and warm seasons by measuring atmospheric ethane (C_2H_6), a tracer of thermogenic CH_4, and comparing ratios of C_2H_6 and CH_4 in the atmosphere and in the pipeline gas flowing through the region; (*iii*) total CH_4 emissions were derived from an atmospheric transport model, which quantitatively links surface fluxes with observed ΔCH_4 using assimilated meteorology; and (*iv*) the fraction of delivered NG lost to the atmosphere was estimated by comparing CH_4 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.

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. Emissions from natural gas are found to be two to three 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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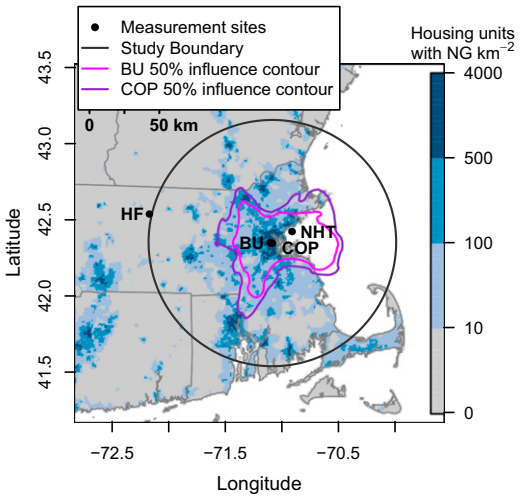

Fig. 1. Location of two city [Boston University (BU), 29-m height; Copley Square (COP), 215-m height] and two peripheral [Harvard Forest (HF); Nahant (NHT)] measurement stations (black points) in Boston, and the surrounding area, overlaid on a map of the number of housing units with NG per square kilometer (14). The 90-km radius circle delineates the ~18,000-km^2 land area for which CH_4 emissions and the NG loss rate were calculated. The magenta and purple contours enclose 50% of the average footprint (sensitivity area) of the BU and COP afternoon measurements, respectively. The two city sites are difficult to distinguish at this scale because the horizontal distance between them is ~2 km. The influence area is ~80% larger for COP than BU because the former station is higher. See *SI Appendix*, Table S2, for additional measurement site location information.

Methane Concentrations in the Boston Atmosphere

Atmospheric CH_4 concentrations were measured continuously from September 2012 through August 2013 at two locations near the urban center [Boston University (BU) and Copley Square (COP)] and two locations outside of Boston [Harvard Forest (HF) and Nahant (NHT)] (Fig. 1 and *SI Appendix*, Table S2 and section S1). Background concentrations in air flowing into the city were estimated by randomly sampling from a range (5th to 35th) of lower percentiles of CH_4 measurements from two upwind stations (HF or NHT, depending on the direction of simulated air trajectories; *SI Appendix*, section S3.1), averaged over a 48-h moving window, to capture synoptic-scale variability and remove possible influences of small nearby sources (*SI Appendix*, section S3.3). Values of ΔCH_4 were calculated by subtracting background from urban concentrations. Hourly average ΔCH_4 data were aggregated into daily afternoon (11–16 h EST, 16–21 h UTC) means to remove autocovariance and focus the analysis on periods of well-mixed atmospheric conditions.

Methane concentrations in Boston were consistently elevated over background (Fig. 2 and *SI Appendix*, Figs. S1 and S2) and followed a distinct daily pattern (Fig. 3 *A* and *C*, and *SI Appendix*, Fig. S16), associated with growth and decay of the planetary boundary layer. Concentrations fluctuated over short timescales (*SI Appendix*, Fig. S1) due to small-scale atmospheric circulations and heterogeneous sources in the urban environment. Methane concentrations were higher in winter than the other seasons at both sites, but ΔCH_4 varied less with season (Fig. 2). The average annual afternoon values of ΔCH_4 at BU and COP were 45.9 (37.3, 58.5) ppb and 30.5 (23.6, 39.3) ppb, respectively (Fig. 2), reflecting different sampling altitudes (30 and 215 m, respectively; *SI Appendix*, Table S2). All errors reported throughout the paper are 95% confidence intervals. Uncertainties in ΔCH_4 (Fig. 2) were calculated through a bootstrap analysis that included background concentrations and afternoon hourly, daily, and seasonally averaged CH_4 measurements (*SI Appendix*, section S3.3).

Contribution of NG to Elevated CH_4 Concentrations

To quantify the fraction of the observed ΔCH_4 that was due to NG emissions, we compared ratios of C_2H_6 and CH_4 measured in the atmosphere and NG pipelines serving the region. Ethane is a significant component of NG, whereas microbial CH_4 sources, such as landfills, sewage, and wetlands, produce little or no C_2H_6 (15). Because Boston has no geologic CH_4 seeps, no oil and gas production or refining, and low rates of biomass burning, there are no known significant sources of C_2H_6 in the region other than NG.

Ethane concentrations were measured with a laser spectrometer (15) at BU for 3 mo in the fall and winter of 2012–13 and 1 mo in the late spring of 2014 (*SI Appendix*, Fig. S6). Covariances between atmospheric C_2H_6 and CH_4 observations were determined from the daily slopes of a linear model that minimizes χ^2 (16) of 5-min median afternoon data (Fig. 4 and *SI Appendix*, section S2.1). The median of the daily slopes of atmospheric C_2H_6 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_2H_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 2012–2013 and 2.4 ± 0.1% in the spring of 2014, determined from hourly gas quality data from the three main pipelines that serve the region (17, 18) (*SI Appendix*, Figs. S7 and S8, and section S2.2). The quotient of the C_2H_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

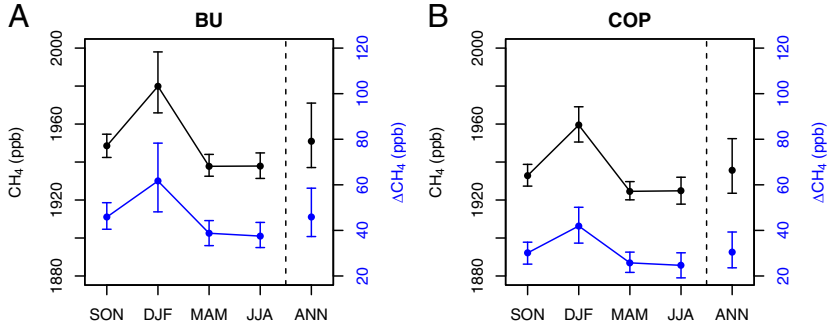

Fig. 2. Average (±95% confidence intervals) afternoon (11–16 h EST) CH_4 (black; left *y* axis) and ΔCH_4 (blue; right *y* axis) by season and for the whole year at (*A*) BU and (*B*) COP.

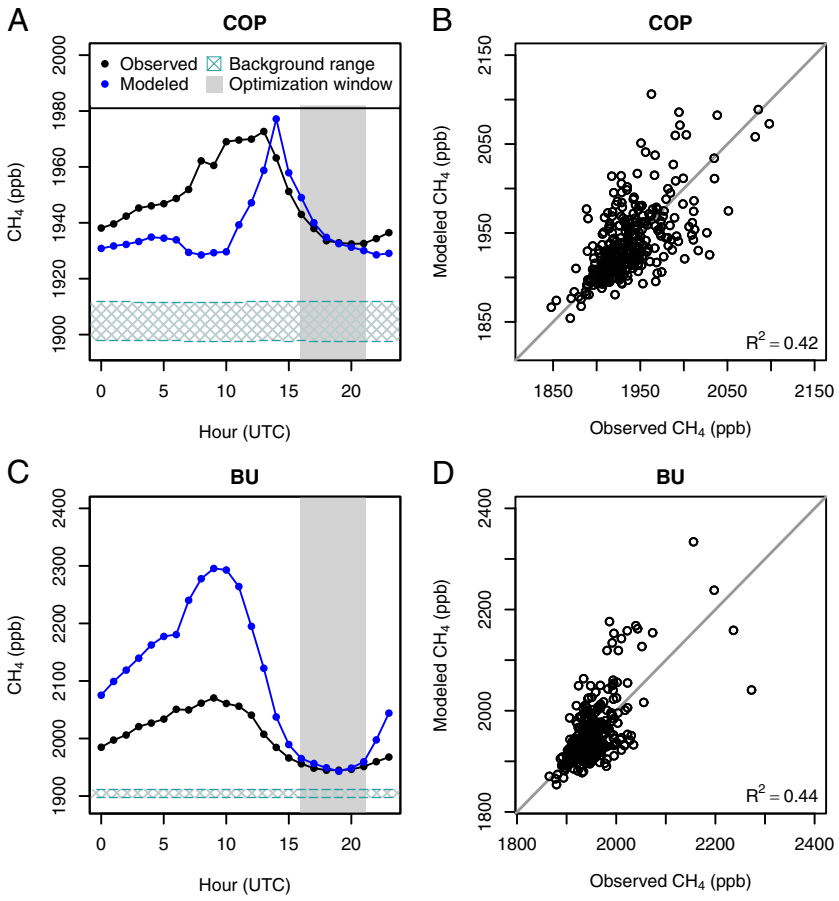

Fig. 3. Observed and optimized modeled CH_4 at (*A* and *B*) COP and (*C* and *D*) BU for 1 y. (*A* and *C*) Observed, modeled, and background CH_4 concentrations averaged by hour of the day. The horizontal hatched area shows the average range of possible background concentrations, derived from 5th to 35th percentiles of the background station data. The gray vertical shaded area indicates the afternoon model optimization period, 11–16 h EST (16-21 h UTC). (*B* and *D*) Modeled versus observed daily average afternoon CH_4 concentrations. The gray line is the one-to-one line.

to the filtering criteria for the atmospheric data (*SI Appendix*, section S2.3).

Methane and Natural Gas Emissions in Greater Boston

Methane enhancements were modeled at BU and COP with the Stochastic Time-Inverted Lagrangian Transport (STILT) model (19), coupled to the Weather Research and Forecasting (WRF) meso-scale meteorological model run at 1-km^2 grid resolution (WRF-STILT; ref. 20; *SI Appendix*, section S3.1). WRF-STILT generates footprints (with units ΔCH_4 per unit surface flux), which represent the sensitivity of each measurement point in space and time to upwind 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 (Fig. 1 and *SI Appendix*, Table S2).

A spatially resolved prior model of CH_4 emissions was constructed for the study region (*SI Appendix*, section S3.2.2, Fig. S12, and Table S4) 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 *SI Appendix*, sections S3 and S4. Observation-model comparisons are shown in Fig. 3 and *SI Appendix*, Figs. S13 and S14.

The mean annual optimized emission rate for the study area was 18.5 ± 3.7 g $CH_4 \cdot m^{-2} \cdot y^{-1}$ from all sources (Fig. 5*A*). 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. 5*A*). 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 not significantly different (*SI Appendix*, Fig. S4), despite the large differences in ΔCH_4 (Fig. 2) and modeled footprints (Fig. 1) 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*, sections S4.2–S4.3).

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 and *SI Appendix*, section S3.2.1). 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. 5 *A* and *B*). The inferred mean annual NG loss rate in the study area was 2.7 ± 0.6% of the total delivered gas in 2012–2013, with little seasonal dependence (Fig. 5*C*). Uncertainties in the average loss rates were calculated by summing in quadrature the relative

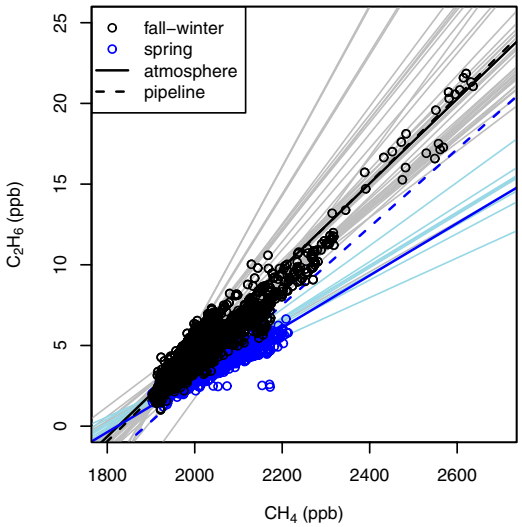

Fig. 4. Five-minute median atmospheric C_2H_6 and CH_4 measurement points at BU in fall and winter of 2012–2013 (black) and spring of 2014 (blue), χ^2 optimization lines fit to each day (light lines), average fit lines for both seasons from all days with $R^2 > 0.75$ (bold solid lines), and lines with slopes of pipeline C_2H_6/CH_4 (dashed lines).

errors for the average emissions, atmospheric NG fraction, and NG consumption terms (*SI Appendix*, section S3.2.1).

The modest seasonality of the inferred NG loss rate (Fig. 5*C*) is driven by the small seasonal variability in total NG consumption (Fig. 5*B*). Our analysis makes no assumptions about the relative contribution to emissions of specific NG-consuming sectors or emission processes (*SI Appendix*, section S3.2.1), 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 mo 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 CH_4 emissions to pipeline 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 (*SI Appendix*, Table S1), using atmospheric data-model frameworks of varying sophistication, but have not quantitatively attributed fluxes to NG. Our value for total CH_4 emissions in Boston is at the low end of the overall range of fluxes reported for other urban areas (*SI Appendix*, Table S1), suggesting that total CH_4 emission rates in Boston are not anomalous.

The US greenhouse gas (GHG) inventory (23) attributes 3,302 Gg of CH_4 emissions to NG transmission, storage, and distribution in 2012, equal to ∼0.7% 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 ∼8% of US emissions attributed to distribution, transport, and storage, and ∼23% of national emissions from distribution alone, a notably higher fraction than the ∼3% of US residential and commercial

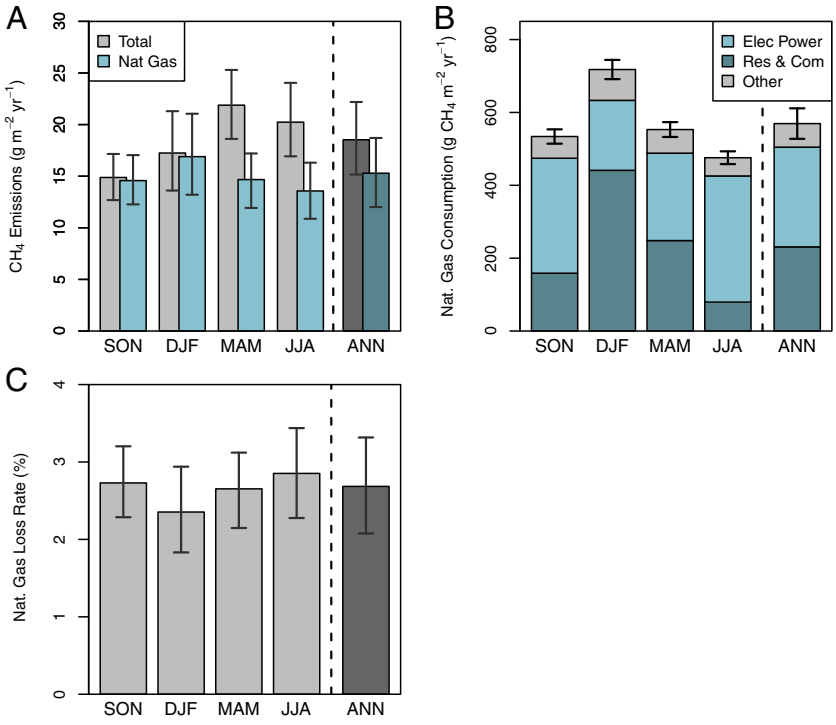

Fig. 5. Seasonal and annual average (±95% confidence intervals) (*A*) optimized CH_4 emissions in total and from NG, (*B*) NG consumption by sector, and (*C*) NG loss rates, derived from CH_4 concentration observations from the BU and COP sites together. (*B*) Consumption categories are electric power, residential and commercial, and other, which is comprised of industrial, vehicle fuel, and pipeline and distribution use (*SI Appendix*, section 3.2.1).

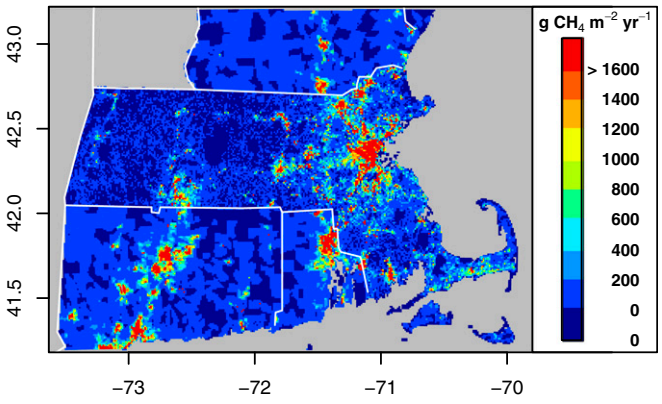

Fig. 6. Reconstructed geographical distribution of NG consumption, in units of CH_4 mass flux, during September 2012 through August 2013 for the four states included in the study region (*SI Appendix*, section S3.2.1).

gas consumed in the study region. More detailed comparison of our results for the Boston urban region to the US GHG inventory is not possible because the inventory is not spatially disaggregated.

Massachusetts has compiled a state GHG inventory (26) (*SI Appendix*, Table S4) using the same methods as the national inventory with state-level data, where available, and reports CH_4 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). Because 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 approximately two to three 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 US Environmental Protection Agency (28) and Massachusetts GHG Reporting Programs (29) show NG loss rates of 0.4–1.6% among individual NG distribution companies in Massachusetts in 2012 and 2013, with an average of 0.6%, weighted by delivered NG volumes. Data reported to the US Energy Information Administration (30) for "losses from leaks, damage, accidents, migration and/or blow down" indicate loss rates of 0–1.1%, with a weighted average of 0.4%, among Massachusetts NG distribution companies in 2012 and 2013.

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 and 2013 were 0–4.6%, with a weighted average of 2.7% (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 CH_4 emissions from NG than we observe in the Boston urban region.

i) Not all emission sources are inventoried. The US 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.

ii) Leak surveys are not comprehensive. Leak surveys (e.g., refs. 13 and 35) 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 CH_4 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.

iii) 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, nonrepeated campaigns in a limited number of locations (25). These limitations can lead to undersampling 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 (36) may help address some of these limitations for particular devices and processes.

These issues arise from our fundamental lack of knowledge about the specific sources and processes responsible for the discrepancies found in this and other studies (6), and about the requirements for designing and testing a statistically rigorous accounting of emissions from the NG supply chain. Both high-emission events and 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 CH_4 emissions from NG and to provide the information needed to reduce those emissions.

Significance of Natural Gas Emissions

This study used 1 y of atmospheric CH_4 measurements from a network of observing stations, a high-resolution modeling framework, atmospheric measurements of C_2H_6, a tracer for NG emissions, and statistics on NG composition and consumption to quantify the NG emission rate for the Boston urban area as 2.7 ± 0.6% (95% confidence interval) of consumed NG, approximately two to three times higher than that given by the most applicable (state) GHG inventory. The total volume of emitted gas in the study area over 1 y was ∼15 billion standard cubic feet (scf), valued conservatively at ∼$90 million [using 2012 and 2013 Massachusetts city gate prices (37)], equal to ∼6 scf·person^{-1}·d^{-1} [using the study area population of ∼7.2 million (38)].

The US 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 (39). A new Massachusetts law (40) 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 accounting,

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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