



Majority of US urban natural gas emissions unaccounted for in inventories

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Across many cities, estimates of methane emissions from natural gas (NG) distribution and end use based on atmospheric measurements have generally been more than double bottom-up estimates. We present a top-down study of NG methane emissions from the Boston urban region spanning 8 y (2012 to 2020) to assess total emissions, their seasonality, and trends. We used methane and ethane observations from five sites in and around Boston, combined with a high-resolution transport model, to calculate methane emissions of 76 ± 18 Gg/yr, with 49 ± 9 Gg/yr attributed to NG losses. We found no significant trend in the NG loss rate over 8 y, despite efforts from the city and state to increase the rate of repairing NG pipeline leaks. We estimate that $2.5 \pm 0.5\%$ of the gas entering the urban region is lost, approximately three times higher than bottom-up estimates. We saw a strong correlation between top-down NG emissions and NG consumed on a seasonal basis. This suggests that consumption-driven losses, such as in transmission or end-use, may be a large component of emissions that is missing from inventories, and require future policy action. We also compared top-down NG emission estimates from six US cities, all of which indicate significant missing sources in bottom-up inventories. Across these cities, we estimate NG losses from distribution and end use amount to 20 to 36% of all losses from the US NG supply chain, with a total loss rate of 3.3 to 4.7% of NG from well pad to urban consumer, notably larger than the current Environmental Protection Agency estimate of 1.4% [R. A. Alvarez *et al.*, *Science* 361, 186–188 (2018)].

based on tower, aircraft, and remote sensing measurements (e.g., refs. 4–8). Top-down studies consistently estimate distribution and end use NG emissions to be significantly (two to 10 times) larger than the bottom-up estimates. The large gap between bottom-up and top-down analyses indicates that there are likely large missing sources in inventories, and it is unclear from which sector those emissions originate. In order to implement effective GHG mitigation policies, it is necessary to understand the dominant sources of CH₄ emissions from NG distribution and end use.

We present an 8-y top-down study of NG emissions from the Boston urban region to assess the NG loss rate over time and investigate the potential missing sources of NG emissions in inventories. We also assess the impact of the COVID-19 shutdown on CH₄ emissions during April 2020. This is one of only a few long-term, top-down studies of urban CH₄ emissions and is in an east-coast city with older, leak-prone infrastructure. Previous studies in other cities have not found any statistically significant trend in emissions over time (7, 9–11). Boston has set a target of becoming carbon neutral by 2050 (12), and Massachusetts has been working to reduce NG leaks, with several laws and regulations implemented between 2014 and 2019 requiring timelines for utilities to report and repair large leaks

urban | methane | natural gas | greenhouse gas emissions

Atmospheric methane (CH₄) is the second-most important greenhouse gas (GHG) after carbon dioxide (CO₂); the Intergovernmental Panel on Climate Change (IPCC) estimates that it was responsible for ~20% of global anthropogenic direct radiative forcing from 2000 to 2010 (1). Oil and natural gas (NG) systems are estimated to account for 31% of US anthropogenic methane emissions (2). NG emissions have increased over the last decade, as it has become an increasingly important energy source in the United States due to advances in extraction technology, reduction in cost, and its promotion as having lower CO₂-equivalent emissions relative to other fossil fuels.

Densely populated urban areas are well positioned to effect change in GHG emissions, as they have concentrated population, infrastructure, and emissions along with, in many cases, political will to implement emission reductions policies (3). Pipelines, transmission infrastructure, household and commercial appliances, meters, stationary combustion, and service leaks are thought to be the most significant source types for urban NG emissions (2). Bottom-up inventories estimate that distribution and end use contribute 6% of US emissions from the NG supply chain (2), but that estimate is highly uncertain. Urban NG emissions have been reported for several cities including Boston, Indianapolis, Washington, DC, Los Angeles, New York City, and Philadelphia using top-down methods

Significance

Methane emissions from distribution and end use of natural gas (NG) are not well known. We analyzed atmospheric methane measurements to quantify NG emissions in the Boston area over ~8 y, finding NG emissions approximately three times larger than calculated by usage-based inventories. We observed no change in emissions over 8 y despite efforts from the state to address NG pipeline leaks. Seasonal emissions are directly related to consumption of NG, implying that sources other than pipelines, such as transmission and appliances, are important and may require future policy action. We estimate total supply chain losses of 3.3 to 4.7% for NG consumed in urban areas, which significantly increases the climate impacts of NG compared to Environmental Protection Agency estimates.

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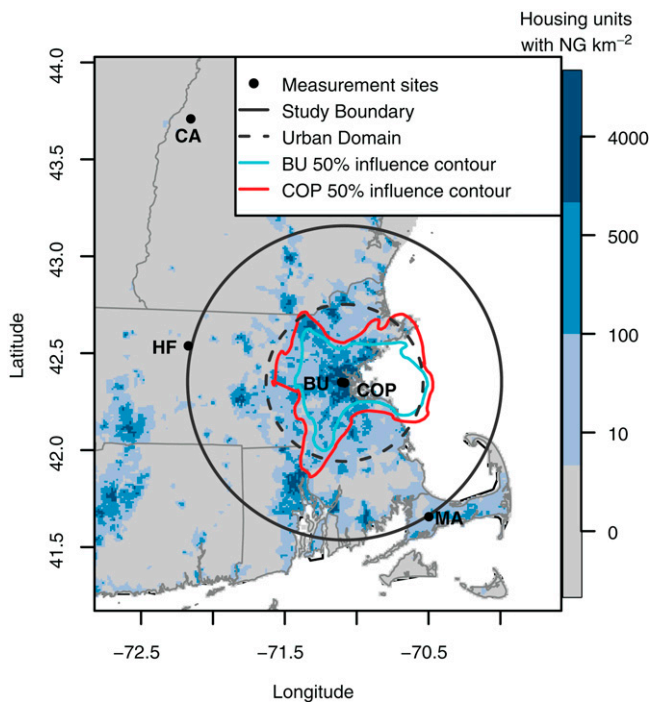


Fig. 1. Map of measurement stations in the Boston network. Study Boundary (black line) bounds the 90-km radius circle in which emissions were optimized. Urban Domain (dashed line) bounds the area for which optimized emissions were reported. The blue shading represents the number of housing units with NG per square kilometer (4). The red (blue) contour encloses 50% of the average footprint (sensitivity area) initiated at the COP (BU) site.

based on their size (13, 14); our study will assess whether these efforts have produced a measurable change in NG emissions.

To put this study in context with other cities and evaluate current knowledge of CH₄ emissions from NG distribution and end use, we then compare top-down NG emission estimates from four US cities with new studies estimating bottom-up emissions from pipeline leaks and in-house losses. By updating top-down versus bottom-up comparisons with the latest available data, we assess the current understanding of the total carbon footprint of NG, the state of the NG budget, and how well we can constrain urban end-use emissions.

Methods

Atmospheric CH₄ concentrations were measured continuously using Picarro cavity ring down spectrometers at two sites in Boston near the urban center, Boston University (BU) and Copley Square (COP), and three locations 90 to 175 km outside of Boston at Harvard Forest in Petersham, MA (HF), Canaan, NH (CA), and Mashpee, MA (MA) (Fig. 1 and *SI Appendix, Table S1*) from September 2012 to May 2020. BU, COP, and HF are operated by Harvard while CA and MA are operated by Earth Networks Inc. and the National Institute of Standards and Technology. The urban sites at BU and COP are 1.7 km apart and sample at 29 m and 215 m above ground level, respectively, providing a direct observation of the surface-layer vertical gradient.

The model-measurement approach used in our inverse analysis has been described by McKain et al. (4) who analyzed CH₄ observations from 2012 to 2013 and Sargent et al. (15) who analyzed CO₂ observations from 2013 to 2014. Briefly, we modeled changes in the CH₄ concentration as air traveled from the boundary of our study region, a 90-km radius circle centered on Boston, to our urban measurement sites at BU or COP. The modeled CH₄ enhancement (ΔCH_4) above the concentration at the region boundary was determined using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (16) run in the recently upgraded "Stochastic Time-Inverted Lagrangian Transport (STILT) mode" coupled with a spatially resolved bottom-up inventory of CH₄ emissions (4).

The HYSPLIT model released an ensemble of 500 particles at each hour at the urban measurement sites (receptors) and followed their trajectories backward in time based on wind fields and turbulence. We used two meteorological drivers, the North American Mesoscale Forecast System (NAM) at 12-km resolution and the High-Resolution Rapid Refresh (HRRR) at 3-km resolution (available 2017 through 2020). HYSPLIT generates an influence function ("footprint" units: parts per million CH₄ per unit flux in $\mu\text{mole}\cdot\text{meter}^{-2}\cdot\text{second}^{-1}$), which quantitatively links upwind surface fluxes to changes in CH₄ concentration at the receptor. In the near-field, the mixing layer height in HYSPLIT was adjusted based on the particle heights as described in ref. 15 to better account for the particles' interaction with the surface before they are fully mixed through the planetary boundary layer.

The footprints were convolved with a 1-km resolution prior model of anthropogenic and biogenic CH₄ emissions previously described by McKain et al. (4), which was augmented with updated landfill and NG point source fluxes (17–19), residential (20–22) and commercial (23) building losses, and seasonally varying wetland emissions (24) (*SI Appendix, section S2*). The inventory also included newly reported pipeline losses from Weller et al. (25), who assessed pipeline leaks with methane analyzers driven through four cities, covering ~10,000 km of pipelines. The convolution of the HYSPLIT footprint within our study region and gridded prior emissions enabled us to compute ΔCH_4 , the expected increase in CH₄ concentration between our study boundary and urban measurement site based on the bottom-up emissions estimate.

Ethane (C₂H₆) measurements from the two urban sites as well as aircraft measurements were used to quantify the fraction of the observed ΔCH_4 that was due to NG emissions. Ethane is a significant component of NG, whereas microbial CH₄ sources, such as landfills, sewage, and wetlands, produce little or no ethane (26). Because Boston has no geologic CH₄ seeps, no oil and gas production or refining, and low rates of biomass burning, there are no known significant sources of ethane in the region other than NG. Ethane concentrations were measured using a laser spectrometer (26) at BU for 3 mo in the fall and winter of 2012 to 13 and 1 mo in the late spring of 2014 (5); they were measured via aircraft in August/September 2017 and March 2018 (27); they were also measured at COP for 5 mo in the fall and winter of 2019 to 2020. Following the method of McKain et al. (4), the atmospheric C₂H₆:CH₄ ratio was determined from the slopes of colocated C₂H₆ versus CH₄ measurements and compared to the C₂H₆:CH₄ ratio of NG flowing into the region during the measurement period (*SI Appendix, section S3 and Fig. S1*). The percentages of observed CH₄ due to NG from these three studies are shown in Table 1, with average values of 91% NG in the dormant season and 76% in the growing season. The aircraft study shows a much smaller change in NG fraction with season than the tower study, likely because the aircraft sampled a smaller, more urban domain with less biogenic emissions. Due to large uncertainties in prior NG emissions estimates, the NG emissions layer in our prior inventory was scaled to be consistent with the attribution results from ethane data. Without adjusting the biogenic component of prior CH₄ emissions, the prior NG emissions were scaled such that they contributed on average 91%, 84%, and 76% of the footprint-weighted ΔCH_4 at the urban sites in the dormant, transitional, and growing seasons, respectively. Unscaled priors were also tested, along with NG fractions spanning the range of observed values and a range of wetland emissions from different inventories.

The CH₄ concentrations at the boundary of the study region were calculated as the lower 20th percentile, 48-h running average of CH₄ measurements from the HF, CA, or MA sites, with the upstream site chosen based on the average azimuth at which the HYSPLIT particles exited the 90-km radius circle around Boston. The model selected the MA site for exit angles from 120 to 180°, the HF site for 180 to 300°, and the CA site for 300 to 360°.

Observations corresponding to easterly wind directions (0 to 120° exit azimuth) were discarded due to lack of a suitable boundary site and uncertainty in modeling sea breezes that account for a significant fraction of the easterly inflow conditions. As westerly winds are the most common, this criterion excluded 25% of days in the spring and fall and 11% of days in the winter. During sea breeze conditions, air can recirculate over the city, picking up large amounts of pollution. This is very difficult for the HYSPLIT model to capture accurately using regional scale meteorology (28); we therefore excluded these angles.

The observed ΔCH_4 was calculated as the difference between the observed CH₄ at each urban site and the background concentration, with a time delay between the upwind and downwind sites equal to the average travel time from the receptor to the study region boundary. Hourly average ΔCH_4 values were aggregated into daily afternoon averages (11 to 16 h EST) to focus on periods when the atmosphere is well-mixed.

Optimized CH₄ emissions were calculated for groups of 2mo with similar NG consumption (January/February, December/March, November/April, May/October, June/September, and July/August) in each year from September 2012

Table 1. Fraction of atmospheric CH₄ attributed to NG sources based on three measurement campaigns

	Time period	Location	Dormant	Growing
Ref. 4	2012 through 2013	BU	98%	67%
Ref. 27	2017 through 2018	Aircraft	87%	85%
This work	2019 through 2020	COP	88%	

to May 2020 (or for single months for which a corresponding month was not available). For each 2-mo group, a single scaling factor (SF) was determined by dividing the mean observed CH₄ enhancement by the mean modeled CH₄ enhancement:

$$SF = \text{mean}(\Delta\text{CH}_4\text{obs}) / \text{mean}(\Delta\text{CH}_4\text{model}).$$

Optimized ΔCH_4 and ΔNG emissions were calculated as the product of the prior emissions and the SF for each period:

$$\text{CH}_4\text{optimized} = \text{CH}_4\text{boundary} + SF * \Delta\text{CH}_4\text{model}.$$

We compared optimized emissions from the portion of MA within our 90-km circle to state-level monthly NG consumption (29) multiplied by 0.88, the fraction of state NG use estimated to be within our study area based on the spatially resolved map of NG consumption from McKain et al. (4). We did not include the emissions or consumption from other states because the vast majority of the HYSPLIT footprint falls within MA. Fractional loss rates to the atmosphere were obtained by dividing optimized NG emissions by total NG consumption in MA and within the study region. Uncertainties in optimized emissions were calculated through a bootstrap analysis that accounted for variations of hourly and daily observed and model enhancements and boundary CH₄, as well as uncertainties in prior emissions and atmospheric transport.

Results

We quantified average methane emissions within a 45-km radius of Boston to be $37.5 \pm 9 \text{ g}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ (95% CI) from 9/2012 to 5/2020, with $24.1 \pm 4.6 \text{ g}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ of that total originating from NG, corresponding to total emission of 49 Gg/yr of NG methane from this region. Average NG emissions in the 90-km radius study domain were $14.0 \pm 2.7 \text{ g}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$, which is comparable to the value of $15.3 \pm 3.5 \text{ g}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ calculated by McKain et al. for the same domain (4). The 45-km radius study area corresponds to $\sim 50\%$ of the HYSPLIT footprint influence (Fig. 1) and approximates the Boston urban area defined by the US Census Bureau Topologically Integrated Geographic Encoding and Referencing (TIGER) database (30).

We found an average NG loss rate of $2.5 \pm 0.5\%$ from 2012 to 2020 by comparing NG emissions to consumption in our

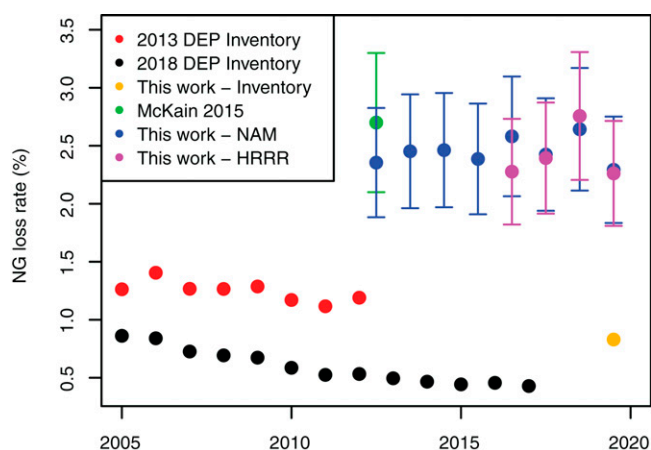


Fig. 2. Yearly NG loss rates based on inventories released by MassDEP in 2013 (red) and 2018 (black), calculated with a top-down method by McKain et al. (4) (green), by this work from the inventory (orange), and top-down analysis using NAM12 (blue) or HRRR (purple) meteorology. Error bars were calculated using a bootstrap method and represent 95% CI.

study area. There was no statistically significant trend in loss rate over our 8-y study period (Fig. 2), despite new regulations in Massachusetts requiring utilities to repair large leaks (14). We also saw no trend in ΔCH_4 between our Boston and background sites over that time period (*SI Appendix, Fig. S3*). Our calculated loss rate is three times higher than the 0.8% loss rate indicated by our prior inventory for Boston and six times higher than the Massachusetts Department of Environmental Protection (MassDEP) estimate and the Gridded Environmental Protection Agency (EPA) inventory (31), neither of which include end-use emissions. The variability among the three inventories shown in Fig. 2 is mainly due to using pipeline emission factors based on different studies.

From 2017 to 2020, we calculated a loss rate of $2.4 \pm 0.5\%$ using the HYSPLIT model; the loss rates calculated using the NAM 12-km resolution and HRRR 3-km resolution meteorologies were indistinguishable. These rates and the $2.5 \pm 0.5\%$ loss rate based on NAM for 2012 to 2020 are comparable to the $2.7 \pm 0.6\%$ loss rate calculated by McKain et al. (4) from 2012 to 2013 using the Weather Research and Forecasting (WRF) model meteorology and the STILT model. The excellent agreement among models using three different meteorology products and two transport models provides confidence that wind errors are not the cause of discrepancies between top-down and bottom-up studies. There was also excellent agreement in annual average emissions between analyses based on the COP and BU sites (Fig. 3), providing confidence in the total emissions and temporal trends.

As a check, we also calculated methane emissions for December 2013 through February 2014 using the observed atmospheric CO₂:CH₄ ratio and optimized CO₂ emissions calculated by Sargent et al. (15) and calculated a loss rate of 1.7%, slightly lower than our CI. The ratio method uses the Anthropogenic Carbon Emissions System (ACES) CO₂ inventory, which is entirely independent from the methane inventory. This method implicitly assumes that methane emissions are collocated with CO₂ emissions and focused on winter when biological emissions are low and emissions of both species are dominated by anthropogenic sources. This assumption is imperfect given that CO₂ emissions from traffic and CH₄ emissions from landfills and wastewater may not be collocated with the other species, but the method is useful as an independent order of magnitude check. This estimate also gives a loss rate more than double that of bottom-up methods.

Our model reproduced daily and weekly variability in atmospheric CH₄ well (Fig. 4). Both the observed methane concentrations and the enhancement between the urban and background sites were highest in the winter and lowest in the summer (*SI Appendix, Figs. S3 and S4*). Fig. 3 shows a strong seasonal cycle in optimized NG emissions, with a larger amplitude at the COP site than at the BU site. There is no strong seasonal variability in optimized total CH₄ emissions (*SI Appendix, Fig. S7*). The seasonal variability in NG emissions is larger than the variability in the observed ΔCH_4 , because the C₂H₆:CH₄ ratio indicates a larger fraction of CH₄ from NG in the winter than in the summer, while wetland CH₄ emissions peak in the summer.

Summer NG emissions at COP and BU were 58% and 28% lower, respectively, than winter emissions. The difference in seasonal amplitude between the two sites is likely due to different emitters in the footprints of each site, with a larger fraction of NG emissions in the footprint of the BU site that are independent of season, such as restaurants and pipeline leaks. We expect the much taller COP site, which samples at 215 m, to be more representative of regional emissions, while the 29 m BU site is more sensitive to local emissions. Though the amplitude of the seasonal cycles differs at the two sites, the average

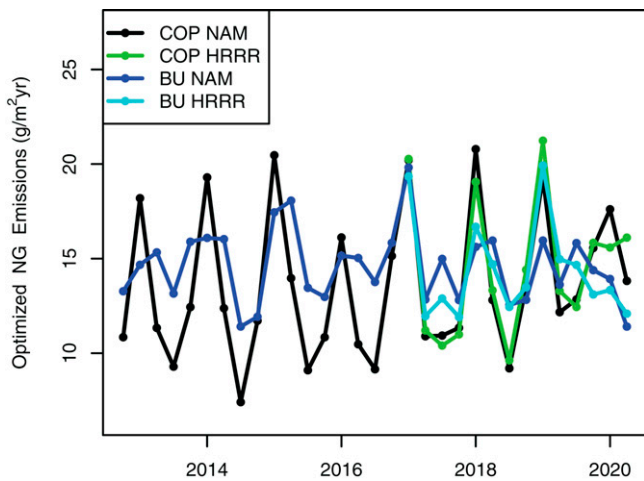


Fig. 3. Seasonal average optimized NG emissions (grams/meter²·year) based on observations at the COP and BU sites using NAM12 or HRRR meteorology.

annual emissions and temporal trends calculated from the two sites are in excellent agreement (Fig. 3).

We find that NG emissions and consumption are highly correlated. This is surprising, because distribution pipelines, thought to be a dominant source of NG losses, are at fairly constant pressure year-round, and thus, their emissions are not expected to vary with consumption. Posterior NG emissions from measurements at the COP site were compared with Massachusetts monthly NG consumption in the study area from the residential, commercial, and industrial sectors from the US Energy Information Administration (EIA) (29) (Fig. 5). As the COP site has a larger footprint than the BU site, we expect state-level consumption values to be more representative of consumption in its footprint than in the BU footprint (city-level NG consumption was not available) (*SI Appendix, Fig. S5*). Fig. 5 shows a weighted, linear least-squares fit between emissions and consumption. The intercept of this line implies a seasonally invariant loss of 6.4 g/m²/yr which is ~3 times higher than the pipeline emissions estimated by Weller et al. (25). The slope of this line (0.025) implies an additional loss of 2.5% of consumed NG, which is in good agreement with the 2.5% loss rate calculated by comparing total yearly emissions to consumption. The correlation also holds for total methane emissions during the dormant season (Fig. 5, *Right*, blue points). Note that NG

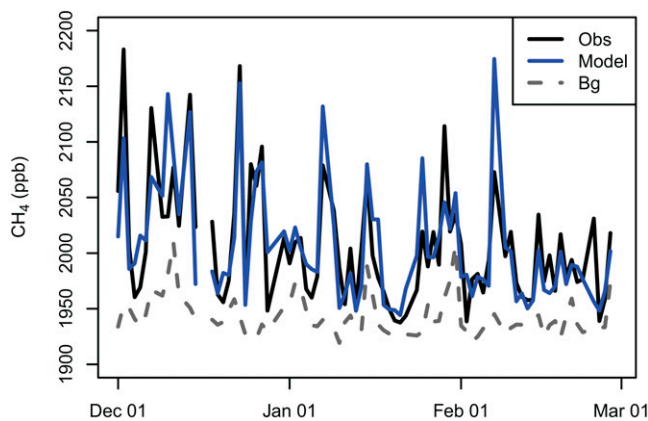


Fig. 4. Observed (Obs), scaled model (Model), and background (Bg) CH₄ at COP. Daily afternoon average (11 to 16 h EST) concentrations for December, January, and February 2014 to 2015.

emissions and consumption are fully independent datasets, as NG consumption was not used in the prior inventory.

In bottom-up studies of residential and commercial buildings, appliances and furnaces were reported to have loss rates of 0.1 to 0.3% of consumed gas (20, 21, 23); transmission (compressor stations, etc.) is estimated to have a loss rate of 0.2% (4). Though we expect the slope of 2.5% at COP (215-m sampling height) to be more representative of regional emissions than the slope of 1% at BU (which, sampling at 29 m, compares fairly local emissions to state-level consumption), even taking a loss of 1% of consumption as a lower bound is significantly higher than is expected from bottom-up inventories. Thus, while there is some uncertainty in the regional seasonality of NG losses based on the difference between our two urban sites, both sites indicate consumption-driven losses that are significantly higher than bottom-up estimates.

To test whether the correlation between emissions and consumption was an artifact of the seasonality of the prior inventory, we performed sensitivity studies with a range of NG fractions and wetland emissions as well as a seasonally invariant prior. These sensitivity studies produced NG emissions within $\pm 20\%$ of the main configuration and maintained the strong relationship between emissions and consumption (*SI Appendix, section S5*). We also ran a test which included angles of 0 to 120°, using MA as a background; it produced NG and CH₄ emissions which agreed with the main configuration to within 2%. Additionally, we compared results with and without MA exit angles, as they can sometimes be difficult to model due to sea breezes. We found no difference in results when MA angles were excluded. We found that the wetland methane source in our prior inventory is not significantly different for east versus west winds, so wind direction could not be used to separate out the wetland methane source.

Total methane emissions do not follow NG consumption in the summer when the ethane:methane ratios indicate that biological sources such as wetlands and landfills are a larger portion of the total methane enhancement in the city. Because they tend to be located farther from our urban sites than the NG emissions, wetland and landfill emissions lead to relatively small changes in concentrations at the receptor. Therefore, our network cannot strongly constrain emissions from biological sources, leading to larger uncertainty in total methane emissions, particularly in the summer, than in NG emissions (*SI Appendix, Fig. S7*). We have confidence in the NG component of methane emissions throughout the year, as these emissions are concentrated near the receptors and strongly influence observations. The optimized emissions of NG are not significantly influenced by adjusting wetland emissions in the prior by up to a factor of 6.

Seasonal changes in urban methane emissions have previously been found by Huang et al. (32) in Washington, DC (summer CH₄ emissions were 41% lower than winter emissions) and He et al. (10), Wong et al. (10), and Yadav et al. (33) in Los Angeles (summer emissions were 26%, 22%, and 40% lower than winter emissions, respectively). However, the fraction of NG was not determined in these studies, which assessed total methane emissions only. The associated consumption in the measurement footprint and relationship to consumption was calculated by He et al., who found a slope of 0.014 between CH₄ emissions and residential and commercial consumption, which is notably lower than the slope of 0.025 (versus NG) or 0.021 (versus CH₄ in dormant season) that we found in Boston. He et al. used a remote mapping spectrometer to measure the CO₂:CH₄ ratio, combined with a prior CO₂ inventory to calculate top-down CH₄ emissions. The difference in slopes could be influenced by the different ages of infrastructure, the types of emitters, and the NG heating demand due to different climates in the two cities.

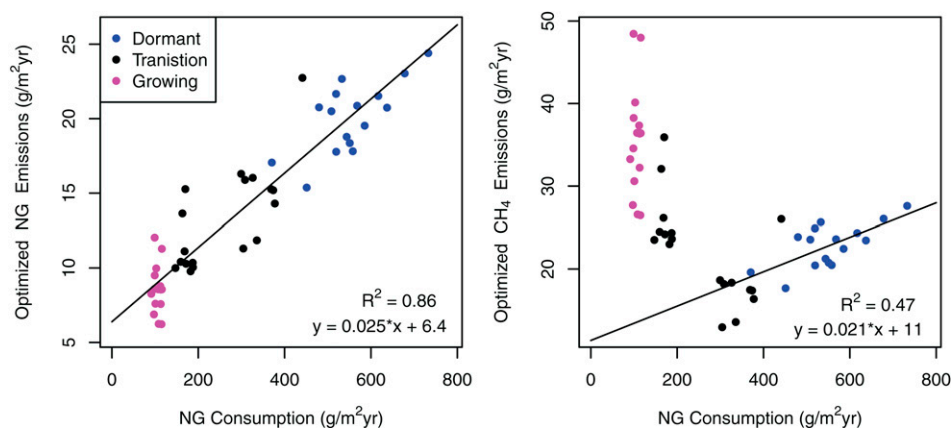


Fig. 5. Optimized NG emissions (*Left*) and total methane emissions (*Right*) compared to residential and commercial NG consumption in MA based on measurements at the COP site. The linear fit to all data (*Left*) and dormant season data (December through March) (*Right*) is shown. Each point represents a 2-mo average.

We were also able to investigate the impact of the 2020 Covid-19 shutdown on methane and NG emissions in Boston. April 2020 emissions of both methane and NG determined by inverse analysis at the BU site were 42% lower than the average of previous April emissions and 22% lower than the lowest April emissions from our study period in 2017. In contrast, April 2020 emissions based on inverse analysis at the COP site were approximately equal to the average of April emissions from 2013 to 2019. Fig. 6 shows that 2020 was the only year in which the average April methane concentration at the 215-m COP site was higher than that at the 29-m BU site (which is only 1.7 km away), demonstrating an inversion of the typical atmospheric concentration gradient.

Massachusetts' total NG consumption during April 2020 showed no change in the residential or industrial sectors compared to previous years, while the electrical sector showed a 35% decrease in consumption compared to 2018 and 2019, and there was a small decrease in commercial consumption (29) (*SI Appendix, Fig. S8*). However, state-level data are not necessarily representative of changes that could have happened locally near BU, as evidenced by the weaker correlation between emissions calculated from the BU site and state-level consumption compared to the correlation at the COP site. The marked decrease in methane emissions at BU could be due to reduced appliance use in office buildings, restaurants, and/or the BU campus surrounding the BU site. There is no evidence that BU buildings changed their heat consumption, though they did stop cooking and serving food. Like the strong correlation between NG emissions and consumption over the 8-y period, the significant change in methane emissions at BU during the Covid-19

shutdown indicates that changes in local consumption are driving methane enhancements and retrieved emissions at this station, reflecting the low sampling altitude (29 m agl). The significant decrease in CH₄ emissions locally around BU during April 2020, when residential NG consumption and pipeline losses were constant, points to the importance of other sources such as beyond-the-meter losses and the necessity of further studies to quantify these sources.

Fig. 7 compares this study's top-down and bottom-up per capita NG emissions for Boston with other cities that have been studied across the United States. We compared seven top-down studies that explicitly calculate NG emissions. In order to compare NG emissions from as many studies as possible, we also calculated the NG component of methane emissions for top-down studies which estimated total methane emissions only by multiplying methane emissions by the fraction of methane from NG determined by other studies of the same city (*SI Appendix, section S8*).

Across the six cities, we find remarkably similar levels of NG emissions when adjusted for population. The emissions per capita are significantly higher than bottom-up estimates in every study, irrespective of the infrastructure age, notwithstanding many differences among study designs. Tower-based, top-down estimates for Boston, Indianapolis, Washington, DC, and Los Angeles were three, six, 10, and two times greater, respectively, than bottom-up estimates. We find that while the updating the Boston inventory with the latest pipeline, appliance, and building losses increased estimated emissions and reduced the gap between top-down and bottom-up studies, large, unexplained gaps remain (Fig. 7B). Among these studies, loss rates from NG infrastructure were calculated in different ways; for comparison, we recalculated loss rates for some studies according to our method (*SI Appendix, Table S3*). These studies produce loss rates of 1.1 to 2.1% for Washington, DC (8, 32) and 2 to 2.3% for Los Angeles (9, 35), comparable to the 2.5% shown here for Boston.

Conclusions

Top-down studies which estimate methane emissions from atmospheric concentration measurements are a powerful tool that can be used to quantify the scale of NG emissions and assess how the total carbon footprint of NG compares to that of other fuels. We analyzed nearly 8 y of methane observations from five sites in and around Boston in an inverse model framework and found that NG emissions are three times higher than our bottom-up inventory estimates and six times higher than

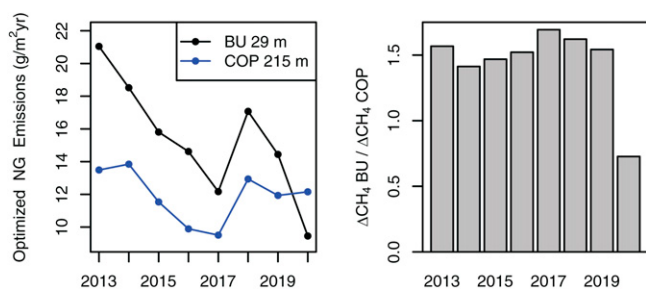


Fig. 6. (*Left*) Mean April optimized emissions from inverse model based on observations from BU or COP site. (*Right*) Ratio of mean April ΔCH_4 BU versus ΔCH_4 COP, in which ΔCH_4 is the enhancement between the urban and background CH₄.

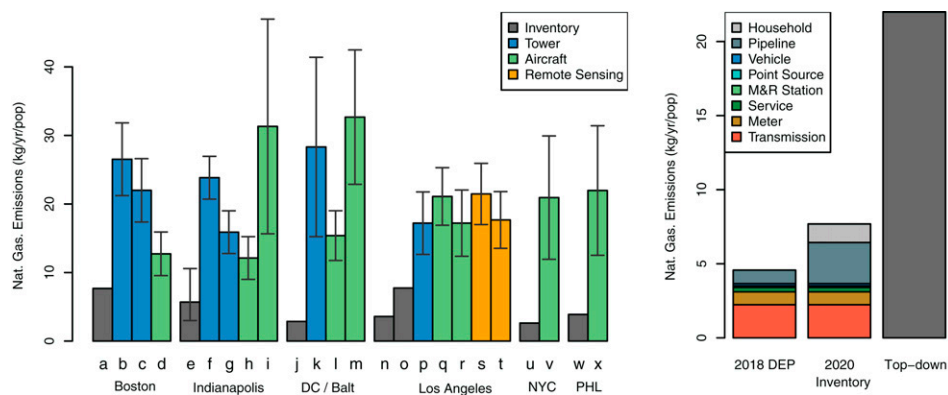


Fig. 7. (Left) Per capita NG emissions from top-down studies in six US cities. Boston: (a) This work, (b) McKain (4), (c) This work, (d) Plant (5), Indianapolis: (e) Lamb (6), (f) Lamb (6), (g) Balashov (11), (h) Lamb (6), (i) Cambaliza (34), DC/Baltimore: (j) EPA (8), (k) Huang (32), (l) Ren (8), (m) Plant (5), Los Angeles: (n) CARB (35), (o) EPA (33), (p) Yadav (33), (q) Cui (36), (r) Peischl (35), (s) Wunch (7), (t) Wong (10), New York City: (u) EPA (5), (v) Plant (5), Philadelphia: (w) EPA (5), and (x) Plant (5). (Right) Boston bottom-up inventory NG emissions by sector compared to top-down emissions. "2018 DEP": vehicle, point source emissions as described in *Methods*, no household emissions, and all other sources from 2018 DEP (37); "2020 inventory": same as "2018 DEP" except pipeline emissions from Weller et al. (25), household and building emissions from Fischer et al. (20), Lebel et al. (22), and California Energy Commission (23); "Top-down": emissions from this study.

the most recent MassDEP estimate, which does not include end use. Comparing NG emissions to consumption in the region, we find an average loss rate of $2.5 \pm 0.5\%$ from urban infrastructure or about 49 Gg/yr of NG methane for the metro area.

The city of Boston has set a goal of becoming carbon neutral by 2050 (12), and Massachusetts implemented new laws and regulations between 2014 and 2019 requiring utilities to report and repair large leaks based on their size (13, 14). A 2019 study by the Home Energy Efficiency Team (HEET) predicted that the 2018 law requiring the repair of leaks deemed "significant environmental impact" could reduce pipeline emissions by half, based on a finding that 7% of leaks emit half of all gas by volume (38). Our analysis finds that these efforts have not yet resulted in a measurable change in methane emissions, as we find no statistically significant trend over the last 8 y (Fig. 2; changes larger than 19% should be detectable given the model uncertainty).

Pipeline emissions account for 42% of total NG emissions in the bottom-up inventory; if they account for the same fraction of unknown emissions, a reduction of 50% due to policy action would be detectable by our model. All gas companies in Massachusetts have released the number of known leaks and repairs on their systems each year since 2014 (39). Notably, there has been no significant change in the number of grade 1 or 2 leaks on the pipeline system and only a slight reduction in the number of grade 3 leaks, with no sign of change after the 2018 law (*SI Appendix, Fig. S9*). Hence, from both a bottom-up leak count and our top-down analysis, it seems that new leaks are appearing in the aging Boston pipeline system as fast as old ones are being fixed. In contrast, the MassDEP bottom-up estimate indicated a 15% decrease in transmission and distribution emissions from 2012 to 2018, which was mainly due to changes in emission factors (estimated leaks per mile of pipeline, based on studies performed in other cities).

We found a strong, unexpected correlation between NG emissions and consumption in Boston (Fig. 5), a relationship which has also been demonstrated in Washington, DC (32) and Los Angeles (9, 10, 33). During the COVID-19 shutdown in April 2020, we calculated a 42% decrease in methane emissions retrieved at our lower sampling height compared to the average from April of previous years. As NG usage changed significantly in some sectors during this period, this is further evidence that NG emissions are significantly driven by consumption. This

correlation between emissions and consumption is surprising because distribution pipelines, thought to be a dominant source of NG losses, are at fairly constant pressure year-round, and thus, their emissions are not expected to vary very much with consumption. Our results therefore indicate that either pipeline emissions do vary with throughput, and/or a large fraction of emissions are from other sources in which emissions are directly linked to consumption, such as space heating and other appliances in residential, industrial, and commercial buildings, transmission intersection points, flow meters, boosting compressors, or step-down and regulation. Current efforts to reduce NG emissions often target pipeline leaks; however, if a significant portion of NG emissions are not from pipelines but from consumption-driven processes, it could require changing the scope of future policy. The results also imply that policies aimed at reducing NG consumption such as shifting away from NG use in buildings could substantially reduce GHG emissions if NG is replaced with green alternatives.

To put this research in context with other cities across the United States, we compared top-down methane emissions from 12 studies across six cities with bottom-up emissions estimates based on the latest studies of pipeline, appliance, home, and commercial building losses (Fig. 7). The longest of these studies were 4 y (10) and 9 y (7); both used tracer-tracer ratios with a CO₂ or CO prior inventory. Of the studies using a CH₄ prior, the longest was 3 y (11). Across the cities, we found top-down emissions to be two to 10 times greater than bottom-up emissions estimates. The cities studied, Boston, Indianapolis, Washington, DC, Los Angeles, New York City, and Philadelphia, represent cities with both older and newer infrastructure, warmer and colder climates, and a wide range of populations. We therefore expect the range of emissions from those cities to be fairly representative of the variability across US cities. Somewhat surprisingly, though the cities with newer infrastructure, Indianapolis and Los Angeles, had slightly lower estimated emissions per capita than the cities with older infrastructure, the difference was small and within the uncertainty of the studies. This result might also point to an important role of consumption-associated emissions.

In these cities, NG emissions were estimated to account for 43 to 88% of total methane emissions. Indianapolis was an outlier at only 43% of methane from NG, because the city has a large landfill within the urban area that accounts for a large fraction of the city's emissions. These studies include tower and

aircraft-based sampling as well as ground-based remote sensing methods and span cities with very different topography, wind patterns, and infrastructure. They employ model frameworks based on a variety of meteorological datasets and with methane emissions inventories, tracer–tracer methods based on CO₂ or CO emissions inventories combined with the observed CO₂:CH₄ or CO:CH₄ ratios, and aircraft mass balance. Each model may have bias due to wind speed errors, background calculations, or atmospheric mixing parameterizations, but given the diversity of approaches to the problem, it seems unlikely that such errors could account for the consistently larger emissions from top-down compared to bottom-up estimates across this heterogeneous group of urban studies. We conclude therefore that it is very likely that there are large missing sources of emissions in bottom-up methane inventories related to NG distribution and, in particular, end use.

The rate of urban NG emissions calculated by top-down studies also has important implications for the carbon footprint of NG as a fuel. An estimated 2.2% (40) of NG is lost to the atmosphere from production and transmission of the fuel before it arrives in the city. Adding to that loss rates of 1.1 to 2.5% from distribution and end use across the studies summarized here yields total loss rates of 3.3 to 4.7% associated with the full NG value chain supplying urban areas. Therefore, 30 to 50% of value chain losses for NG consumed in urban areas are from distribution and end use. For Boston, a city with older infrastructure, we calculate a total loss rate from the entire NG supply chain of 4.7%.

If the top-down emissions from these cities are representative of emissions across the country, we estimate that NG losses from distribution and end use amount to 20 to 36% of all losses from the US NG supply chain (including all end uses, not only urban; based on supply chain losses from ref. 2) and 6 to 11% of all anthropogenic methane emissions (including agriculture) (*SI Appendix, section S8*). These top-down studies thus indicate that the climate footprint of NG is larger than generally supposed, implying the need to identify and mitigate urban sources. Because methane has 86 to 125 times the global warming potential of CO₂ over 20 y and 25 to 36 times over 100 y, if 3 to 6% of consumed NG is lost directly to the atmosphere as CH₄, the greenhouse impact of NG is equivalent to that of coal (2). We note, however, that reductions in criteria pollutants emissions continues to be a benefit of NG use compared to coal or oil.

Determining the processes and source types responsible for NG emissions unaccounted for in bottom-up inventories remains a significant challenge. In Boston, the largest NG emitters from our bottom-up estimate are pipelines, transmission, and buildings, which account for 14%, 8%, and 6%, respectively, of our estimated top-down emissions; 67% of top-down emissions are unaccounted for in the bottom-up inventory. Appliance and building emissions including furnaces and boilers, which are expected to follow NG consumption consistent with this study, warrant further study to understand whether they could be a significant source of missing emissions; in particular, few studies have assessed commercial and industrial building emissions. However, as current bottom-up estimates for building losses are 0.1 to 0.3% of consumed gas (20, 21, 23), accounting for only 6% of top-down emissions in Boston (Fig. 7), these

estimates would need to be increased by fivefold or more to account for a significant fraction of missing emissions.

Transmission emissions have been fairly extensively studied at a facility level, with thousands of stations assessed (e.g., refs. 41 and 42) and an estimated loss rate of 0.2% (4). The most important unaccounted-for transmission losses are likely due to “super-emitters” representing operating anomalies or failed systems; facility-level studies have shown them to be very important, and they are difficult to statistically sample using bottom-up methodology. “Super-emitters” could also play a role in missing emissions across other sectors such as losses from appliances and NG pipes within buildings. Combining building and transmission emissions, which should both correlate with consumption, bottom-up estimates are approximately sixfold lower than the 2.5% of consumed gas that is indicated by the relationship between our top-down emissions and consumption.

Weller et al. (25) assessed pipeline leaks with methane analyzers driven through four cities, covering ~10,000 km of pipelines; their extensive coverage of roadways makes it unlikely that they missed a significant fraction of street-level emissions. One potential unaccounted-for source of emissions is pipeline gas that escapes away from streets, such as through sewage pipes or stacked vents in buildings. When we compare top-down emissions from this study to gas consumed, the intercept of the regression line implies a seasonally invariant loss of 6.4 g/m²/yr, which is approximately three times higher than the pipeline emissions estimated by Weller et al. The seasonally invariant emissions in excess of those expected from Weller et al. could be due to a combination of additional pipeline emissions and appliances used year-round such as stoves and water heaters.

Our Boston top-down analysis as well as a review of studies in other cities indicate that a majority of NG emissions in urban areas are not accounted for in bottom-up inventories. Bottom-up studies have assessed each portion of the NG supply chain, and none of these studies can account for the NG emissions that we observe from top-down methods. Therefore, bottom-up studies must be missing significant emissions. Fixing fugitive NG pipeline emissions in the streets is a policy priority in many cities; however, if beyond-the-meter, transmission station, or pipeline losses away from streets make up a significant fraction of the unknown emissions, they could necessitate greater focus in policy action. Targeted, neighborhood level studies from both top-down and bottom-up perspectives will be essential to identify the emissions not accounted for in bottom-up inventories so that stakeholders can effectively focus on mitigating the largest methane emissions sources.

Data Availability. Data have been deposited in Oak Ridge National Laboratory Distributed Active Archive Center (<https://doi.org/10.3334/ORNLDAAC/1982>).

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1. Intergovernmental Panel on Climate Change, *Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, R. Pachauri, L. Meyer, Eds. (Intergovernmental Panel on Climate Change, Geneva, 2014), p. 46.
2. P. M. B. Saint-Vincent, N. J. Pekney, Beyond-the-Meter: Unaccounted sources of methane emissions in the natural gas distribution sector. *Environ. Sci. Technol.* **54**, 39–49 (2020).
3. H. Tabuchi, H. Fountain, Bucking Trump, these cities, states and companies commit to Paris accord. *NY Times*, 2 June 2017, Section A, p. 12.

4. K. McKain et al., Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proc. Natl. Acad. Sci. U.S.A.* **112**, 1941–1946 (2015).
5. G. Plant et al., Large fugitive methane emissions from urban centers along the U.S. east coast. *Geophys. Res. Lett.* **46**, 8500–8507 (2019).
6. B. K. Lamb et al., Direct and indirect measurements and modeling of methane emissions in Indianapolis, Indiana. *Environ. Sci. Technol.* **50**, 8910–8917 (2016).
7. D. Wunch et al., Quantifying the loss of processed natural gas within California's South Coast Air Basin using long-term measurements of ethane and methane. *Atmos. Chem. Phys.* **16**, 14091–14105 (2016).

8. X. Ren *et al.*, Methane emissions from the Baltimore-Washington area based on air-borne observations: Comparison to emissions inventories. *J. Geophys. Res. Atmos.* **123**, 8869–8882 (2018).
9. L. He *et al.*, Atmospheric methane emissions correlate with natural gas consumption from residential and commercial sectors in Los Angeles. *Geophys. Res. Lett.* **46**, 8563–8571 (2019).
10. C. K. Wong *et al.*, Monthly trends of methane emission in Los Angeles from 2011 to 2015 inferred by CLARS-FTS observations. *Atmos. Chem. Phys.* **16**, 13121–13130 (2016).
11. N. V. Balashov *et al.*, Background heterogeneity and other uncertainties in estimating urban methane flux: Results from the Indianapolis Flux Experiment (INFLUX). *Atmos. Chem. Phys.* **20**, 4545–4559 (2020).
12. City of Boston, *Climate Action Plan* (City of Boston, Boston, MA, 2017). <https://www.boston.gov/departments/environment/climate-action-plan>. Accessed 11 June 2021.
13. B. Moran, New state regulation could cut emissions from natural gas leaks in half, report says (2019). <https://www.wbur.org/earthwhile/2019/04/01/natural-gas-methane-leaks-massachusetts-rule>. Accessed 15 July 2021.
14. Gas Leak Allies, Laws and regulations. <https://www.gasleakallies.org/laws-and-regulations>. Accessed 15 July 2021.
15. M. Sargent *et al.*, Anthropogenic and biogenic CO₂ fluxes in the Boston urban region. *Proc. Natl. Acad. Sci. U.S.A.* **115**, 7491–7496 (2018).
16. A. F. Stein *et al.*, NOAA's HYSPLIT atmospheric transport and dispersion modeling system. *Bull. Am. Meteorol. Soc.* **96**, 2059–2077 (2015).
17. Massachusetts Department of Environmental Protection, Active landfills, <https://www.mass.gov/doc/list-of-active-landfills-in-massachusetts-january-2020/download>. Accessed 15 July 2021.
18. Massachusetts Department of Environmental Protection, Inactive and closed landfills, <https://www.mass.gov/doc/list-of-inactive-closed-landfills-dumping-grounds-in-massachusetts-january-2020/download>. Accessed 15 July 2021.
19. Landfill Methane Outreach Program (LMOP), Project and landfill data by state, <https://www.epa.gov/lmop/project-and-landfill-data-state>. Accessed 15 July 2021.
20. M. L. Fischer *et al.*, An estimate of natural gas methane emissions from California homes. *Environ. Sci. Technol.* **52**, 10205–10213 (2018).
21. Z. Merrin, P. W. Francisco, Unburned methane emissions from residential natural gas appliances. *Environ. Sci. Technol.* **53**, 5473–5482 (2019).
22. E. D. Lebel, H. S. Lu, S. A. Speizer, C. J. Finnegan, R. B. Jackson, Quantifying methane emissions from natural gas water heaters. *Environ. Sci. Technol.* **54**, 5737–5745 (2020).
23. E. Johnston *et al.*, California Energy Commission, Characterization of fugitive methane emissions from commercial buildings in California (2020), <https://www.energy.ca.gov/publications/2020/characterization-fugitive-methane-emissions-commercial-buildings-california>. Accessed 15 July 2021.
24. A. A. Bloom *et al.*, *CMS: Global 0.5-deg Wetland Methane Emissions and Uncertainty (WetCHARTS v1.0)* (ORNL DAAC, Oak Ridge, TN, 2017).
25. Z. D. Weller, S. P. Hamburg, J. C. von Fischer, A national estimate of methane leakage from pipeline mains in natural gas local distribution systems. *Environ. Sci. Technol.* **54**, 8958–8967 (2020).
26. T. I. Yacovitch *et al.*, Demonstration of an ethane spectrometer for methane source identification. *Environ. Sci. Technol.* **48**, 8028–8034 (2014).
27. C. Floerchinger *et al.*, Relative flux measurements of biogenic and natural gas-derived methane for seven U.S. cities. *Elementa* (1):000119 (2021).
28. H. Flocas *et al.*, Synoptic and local scale atmospheric circulation associated with air pollution episodes in an urban Mediterranean area. *Theor. Appl. Climatol.* **95**, 265–277 (2009).
29. U.S. Energy Information Administration, Natural gas consumption by end use, https://www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_m.htm. Accessed 5 August 2020.
30. United States Census Bureau, TIGER/Line Shapefiles, technical documentation 2018 (2018). https://www2.census.gov/geo/pdfs/maps-data/data/tiger/tgrshp2018/TGRSHP2018_TechDoc.pdf. Accessed 21 July 2020.
31. J. D. Maasackers *et al.*, Gridded national inventory of U.S. methane emissions. *Environ. Sci. Technol.* **50**, 13123–13133 (2016).
32. Y. Huang *et al.*, Seasonally resolved excess urban methane emissions from the Baltimore/Washington, DC metropolitan region. *Environ. Sci. Technol.* **53**, 11285–11293 (2019).
33. V. Yadav *et al.*, Spatio-temporally resolved methane fluxes from the Los Angeles megacity. *J. Geophys. Res. Atmos.* **124**, 5131–5148 (2019).
34. M. O. L. Cambaliza *et al.*, Quantification and source apportionment of the methane emission flux from the city of Indianapolis. *Elementa* **3**, 000037 (2015).
35. J. Peischl *et al.*, Quantifying sources of methane using light alkanes in the Los Angeles basin, California. *J. Geophys. Res. Atmos.* **118**, 4974–4990. [10.1002/jgrd.50413](https://doi.org/10.1002/jgrd.50413). (2013).
36. Y. Y. Cui *et al.*, Top-down estimate of methane emissions in California using a meso-scale inverse modeling technique: The South Coast Air Basin. *J. Geophys. Res. Atmos.* **120**, 6698–6711 (2015).
37. D. E. P. Massachusetts, Greenhouse Gas Baseline, Inventory & Projection, Appendix C: Massachusetts Annual Greenhouse Gas Emissions Inventory: 1990–2017, with partial 2018 & 2019 data, <https://www.mass.gov/lists/massdep-emissions-inventories>. Accessed 10 March 2020.
38. Natural Gas Leaks of Significant Environmental Impact (SEI), Report of the 2018 SEI field trial utilities enacting the leak extent method (2019). <https://heet.org/wp-content/uploads/2019/04/HEET-Report-of-the-2018-SEI-Field-Trial.pdf>. Accessed 12 July 2021
39. Massachusetts Department of Public Utilities, Report on the prevalence of natural gas leaks in the natural gas system to the Joint Committee on Telecommunications, Utilities, and Energy, and the Joint Committee on Public Safety and Homeland Security, pursuant to an act relative to natural gas leaks (2014). <https://eeonline.eea.state.ma.us/DPU/Fileroom/dockets/bynumber>. Accessed 15 July 2021.
40. R. A. Alvarez *et al.*, Assessment of methane emissions from the U.S. oil and gas supply chain. *Science* **361**, 186–188 (2018).
41. R. Subramanian *et al.*, Methane emissions from natural gas compressor stations in the transmission and storage sector: Measurements and comparisons with the EPA greenhouse gas reporting program protocol. *Environ. Sci. Technol.* **49**, 3252–3261 (2015).
42. D. J. Zimmerle *et al.*, Methane emissions from the natural gas transmission and storage system in the United States. *Environ. Sci. Technol.* **49**, 9374–9383 (2015).