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Large Fugitive Methane Emissions From Urban Centers Along the U.S. East Coast

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Key Points:
- Aircraft observations downwind of six major cities along the U.S. East Coast are used to estimate urban methane emissions
- Observed urban methane estimates are about twice that reported in the Gridded EPA inventory
- Methane emissions from natural gas (including end use) in five cities combined exceeds nationwide emissions estimate from local distribution

Supporting Information:
- Supporting Information SI

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Abstract Urban emissions remain an underexamined part of the methane budget. Here we present and interpret aircraft observations of six old and leak-prone major cities along the East Coast of the United States. We use direct observations of methane (CH_4), carbon dioxide (CO_2), carbon monoxide (CO), ethane (C_2H_6), and their correlations to quantify CH_4 emissions and attribute to natural gas. We find the five largest cities emit 0.85 (0.63, 1.12) Tg CH_4/year, of which 0.75 (0.49, 1.10) Tg CH_4/year is attributed to natural gas. Our estimates, which include all thermogenic methane sources including end use, are more than twice that reported in the most recent gridded EPA inventory, which does not include end-use emissions. These results highlight that current urban inventory estimates of natural gas emissions are substantially low, either due to underestimates of leakage, lack of inclusion of end-use emissions, or some combination thereof.

Plain Language Summary Recent efforts to quantify fugitive methane associated with the oil and gas sector, with a particular focus on production, have resulted in significant revisions upward of emission estimates. In comparison, however, there has been limited focus on urban methane emissions. Given the volume of gas distributed and used in cities, urban losses can impact national-level emissions. In this study we use aircraft observations of methane, carbon dioxide, carbon monoxide, and ethane to determine characteristic correlation slopes, enabling quantification of urban methane emissions and attribution to natural gas. We sample nearly 12% of the U.S. population and 4 of the 10 most populous cities, focusing on older, leak-prone urban centers. Emission estimates are more than twice the total in the U.S. EPA inventory for these regions and are predominantly attributed to fugitive natural gas losses. Current estimates for methane emissions from the natural gas supply chain appear to require revision upward, in part possibly by including end-use emissions, to account for these urban losses.

1. Introduction

Atmospheric methane is a powerful greenhouse gas and a precursor to tropospheric ozone production (Fiore et al., 2002; National Academies of Sciences, E., 2018; West et al., 2006). Quantification of methane emissions and the resultant atmospheric burden is required to assess its current and future impact on climate and air quality. In the past decade, there has been intense focus on improving our understanding of methane emissions in North America, with many atmospheric measurements suggesting larger emissions than estimated in bottom-up inventories (Brandt et al., 2014; Miller et al., 2013). Since methane is the main component of natural gas (NG), a concerted effort has focused on emissions from the oil/NG sectors with an emphasis on production emissions. Synthesizing these recent studies to assess the current best estimate of oil/NG supply chain emissions, Alvarez et al. (2018) found emissions were ~60% higher than those estimated by the EPA. This work investigated multiple segments of the oil/NG industry (production, processing, transmission, etc.), but it specifically did not reexamine emissions from distribution or end use, instead using the current best estimate for local distribution loss by Lamb et al. (2015). Urban emissions, including end-use sources not captured in inventory estimates, remain poorly constrained in part due to lack of data. Only a small number of city-specific studies have quantified CH_4 emissions from urban domains (Boston, McKain et al., 2015; Indianapolis, Lamb et al., 2016; Los Angeles, Wennberg et al., 2012; DC/Baltimore, Ren et al., 2018), while others have examined the frequency of methane leaks with mobile platforms (Fischer et al., 2018; Jackson et al., 2014; Phillips et al., 2013; von Fischer et al., 2017). The general conclusion from these works is that urban CH_4 emissions are significant and the frequency and location of leaks and/or venting in these regions supports the inference that NG is a large contributor of excess urban CH_4.

Collectively, these works suggest urban emissions may exceed estimates by the EPA and Alvarez et al., but we have no good estimate of the total magnitude of this contribution.

In this study we investigate CH_4 emissions with aircraft observations collected downwind of six urban centers along the East Coast of the United States (Washington, DC; Baltimore, MD; Philadelphia, PA; New York, NY; Providence, RI; and Boston, MA). The metropolitan regions range in size, from New York, NY, with a population of over 8.6 million to Providence, RI, at just over 180,000 (U.S. Census Bureau, P. D., 2018). While the region of study represents some of the oldest cities in the country in terms of infrastructure, these metropolitan regions represent 12% of the entire U.S. population and 4 of the 10 most populous urban areas in the United States. Simultaneous measurements of methane (CH_4), carbon dioxide (CO_2), carbon monoxide (CO), and ethane (C_2H_6) in the downwind plumes provide characteristic correlation slopes for each urban center that can be directly compared to current inventories. Multiple measurements across several days and along different tracks showed modest variability of the correlation slopes derived from the integrated downwind plumes, suggesting that the slopes are a simple and robust observational metric to characterize urban emissions. Examination of the latest emission inventories for CH_4, CO_2, and CO in combination with our observations shows that CH_4 emissions are underrepresented in the Gridded EPA inventory (Maasakkers et al., 2016) by more than a factor of 2 across the majority of the cities investigated. Measurements of ethane (C_2H_6) enable attribution of a significant portion of the excess CH_4 to NG. The observed emission correlation slopes are used in conjunction with the independent CO_2 and CO inventories to generate estimates of CH_4 for each of the urban domains. This aircraft-based methodology provides an important observational verification tool to better understand urban emissions.

2. Aircraft Observations

We flew a suite of instrumentation measuring CH_4, CO_2, CO, C_2H_6, and H_2O onboard a National Oceanic and Atmospheric Administration (NOAA) twin otter aircraft (De Havilland DHC-6-300) along the East Coast of the United States for 20 research flights from 8 April 2018 to 12 May 2018 totaling 120 flight hours. Flights covered major metropolitan areas from Washington, DC, to Boston, MA (shown in Figure 1a). Urban plumes were identified using the aircraft wind measurements in conjunction with tracer observations to select the portion of the data downwind of each region corresponding to the integrated city signal. An example of an urban plume is shown in Figure 1 for the New York City region. Emission sources within the urban region are not necessarily collocated within the city; however, measurement sufficiently downwind of the region allows for mixing to homogenize the measured plumes. To allow for sufficient vertical mixing to bring surface emissions to an altitude at which the aircraft could transit, flights were conducted during the afternoon hours (11:00–18:00 EST, 16:00–23:00 UTC). To directly measure air emitted from each urban area, the majority of the measurements were performed at 300–800 m above sea level within the planetary boundary layer. More information about the instrument payload and further flight details is provided in supporting information (SI) Text S1.

Tracer:tracer correlation slopes (CH_4:CO, CO:CO_2, CH_4:CO, and C_2H_6:CH_4) for each urban plume are determined using a Model II regression to allow for different measurement precisions of each trace gas (see SI Text S2) and directly compared to inventory-derived tracer:tracer ratios. Used in conjunction with established CO_2 and CO emissions inventories for each urban region, the observed slopes are used to estimate the CH_4 fluxes, which are compared to the latest CH_4 emission inventories.

3. Urban Emissions Inventories

The emissions inventories used in this study are summarized in Table 1. To be spatially and temporally consistent with our observations, the inventories are partitioned to isolate urban emissions, as detailed in SI Text S3, and used to generate inventory-based anthropogenic emissions tracer:tracer ratios. To date, the Gridded EPA CH_4 inventory by Maasakkers et al. represents our best understanding of the spatial distribution of anthropogenic CH_4 emitters across the United States. Developed to be consistent with the 2016 Inventory of U.S. Greenhouse Gas Emissions and Sinks (GHGI) for 2012, the Gridded EPA inventory spatially disaggregates the national CH_4 estimates provided in the GHGI using source information provided in a variety of sector specific databases. Pipeline material is considered in this approach which theoretically should capture higher leak rates associated with the large percentage of cast iron pipes used throughout the

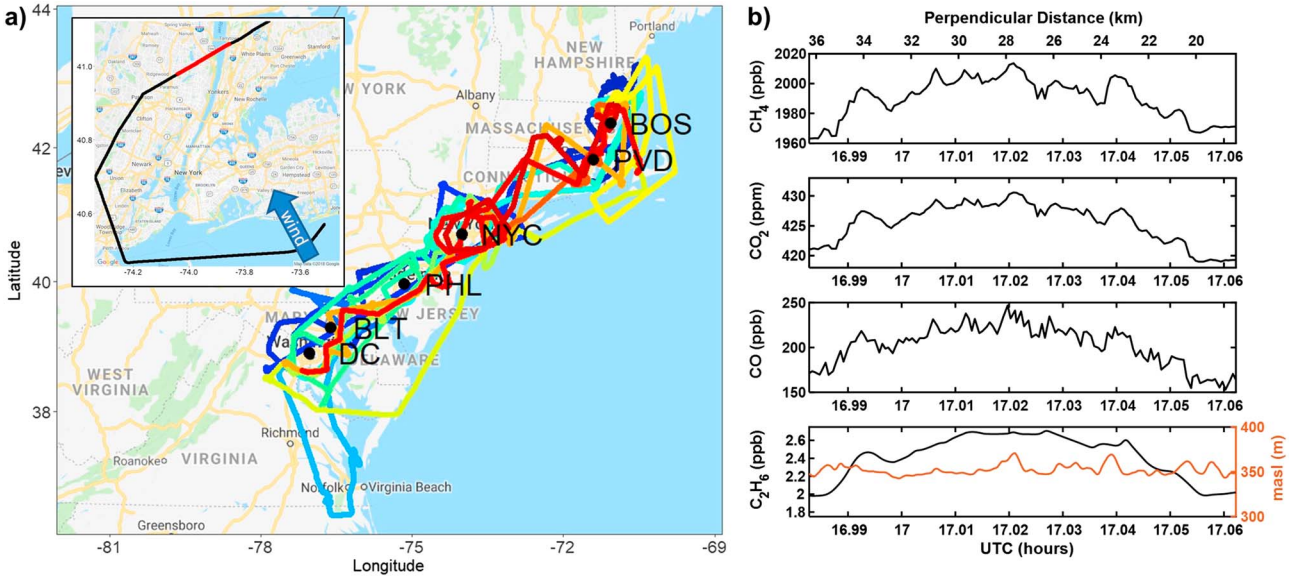

Figure 1. (a) Flight coverage by the ECO campaign around the major urban regions of Washington, DC (DC); Baltimore, MD (BLT); Philadelphia, PA (PHL); New York, NY (NYC); Providence, RI (PVD); and Boston, MA (BOS). Each flight is represented by a different color. The inset shows the flight path (black) and the region representing the downwind plume (red) for NYC on 9 May 2018. Map source: *Google Maps*, Accessed: 18 September 2018 (b) The tracer concentration time series of the NYC plume corresponding to the inset of (a).

region of study. This new inventory attributes a much higher proportion of total emissions to oil/NG production and processing facilities, a result that is consistent with recent top-down studies of major production sites (Maasakkers et al., 2016).

The Emission Database for Global Atmospheric Research (EDGAR) inventory reports CH_4 emission grid-maps by aggregating emissions based on country and sector specific activity data in conjunction with technology, abatement measures, and emission factors (Janssens-Maenhout et al., 2017). The most recent version of the EDGAR inventory (v4.3.2) shows a similar spatial distribution of CH_4 to that of the Gridded

Table 1
Emission Inventories Used in This Study

Inventory	Gases	Year used	Temporal resolution	TIMES scaling (CO_2)	Spatial resolution	Coverage	Reference
EDGAR v4.2FT2010	CH_4 and CO_2	2010	Annual	x	0.1° × 0.1°	Global	(European Commission, Joint Research Centre (JRC)/ Netherlands Environmental Assessment Agency (PBL), 2013)
EGDAR v4.3.2	CH_4, CO_2, and CO	2010	Annual and monthly	x	0.1° × 0.1°	Global	(Crippa et al., 2018; European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL), 2018; Janssens-Maenhout et al., 2017)
Gridded EPA	CH_4	2012	Annual, monthly, and daily		0.1° × 0.1°	United States	(Maasakkers et al., 2016)
ODIAC 2017	CO_2	2016	Monthly	x	1 km × 1 km	Global	(Oda & Maksyutov, 2015)
FFDAS 2014b	CO_2	2014	Hourly		0.1° × 0.1°	Global	(Asefi-Najafabady et al., 2014)
ACES	CO_2	2014	Hourly		1 km × 1 km	Northeast United States	(Gately & Hutyra, 2018)
Gridded NEI	CO	2011	Monthly		0.1° × 0.1°	United States	(Simon et al., 2010; Travis, 2017)

Note. The "TIMES scaling" column indicates the CO_2 inventories that are scaled using the factors detailed in Nassar et al. (2013) to account for the diurnal cycle of CO_2. Links for external datasets used in this study are provided in SI Text S9.

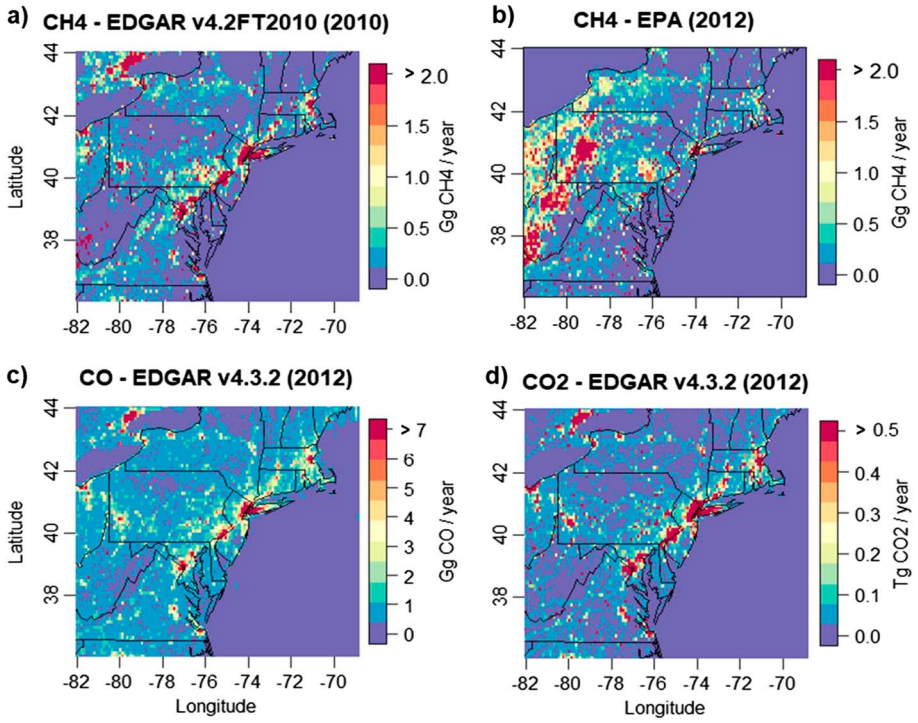

Figure 2. Anthropogenic emissions of CH_4 from (a) EDGAR v4.2FT2010, (b) gridded EPA, emissions of CO from (c) EDGAR v4.3.2, and emissions of CO_2 from (d) EDGAR v4.3.2. EDGAR = Emission Database for Global Atmospheric Research.

EPA for the urban regions of interest in this study. This is in contrast to older versions of EDGAR (v4.2 and v4.2FT2010) that exhibited substantially different spatial emission patterns, specifically with emissions concentrated in cities instead of production basins (Figures 2a and 2b). The spatial pattern provided in EDGAR v4.2 (and v4.2FT2010) has not been thought to be representative and instead a consequence of distributing emissions that should be attributed to oil/NG production and processing with a population proxy, thus underestimating or missing emissions associated with those sectors (Maasakkers et al., 2016). Some recent top-down studies of several large-scale NG facilities have shown consistency with the Gridded EPA inventory (Barkley et al., 2017; Maasakkers et al., 2016; Smith et al., 2017), while other studies of urban domains reported significantly higher CH_4 emissions than those reported by the EPA (McKain et al., 2015; Ren et al., 2018). No CH_4 inventories currently account for end-use emissions that are captured in atmospheric observations of thermogenic emissions, which may partially explain the observed discrepancies.

4. Tracer:Tracer Slopes: Comparison Between Inventories and Observations

4.1. CH_4:CO_2

The direct comparison of the observed CH_4:CO_2 correlations and inventory-derived emission ratios for each city is shown in Figure 3a. In each method, the CH_4:CO_2 relationship characterizes the anthropogenic emissions due to the urban core. Natural sources of CH_4 and CO in these urban regions are excluded in the inventories and are assumed to be negligible in our observations, while ecosystem CO_2 respiration and uptake (i.e., photosynthesis) have the potential to skew the observed plumes due to gradients between the respective fluxes within the urban core and the adjacent regions. We examine this potential source of error in SI Text S4 and find limited photosynthesis activity during the campaign. Ecosystem respiration is considered in our uncertainty analysis based on estimates in the literature. In addition, we also perform an independent methane emissions estimate using the observed CH_4:CO slopes, which are not sensitive to these biogenic influences during the time of the observations. This analysis is discussed further in the next section. The variation in the derived emission slopes across multiple flights with differing wind directions provides statistical constraints on our observed slope estimates. Uncertainty in the inventory ratios are inferred by the spread of

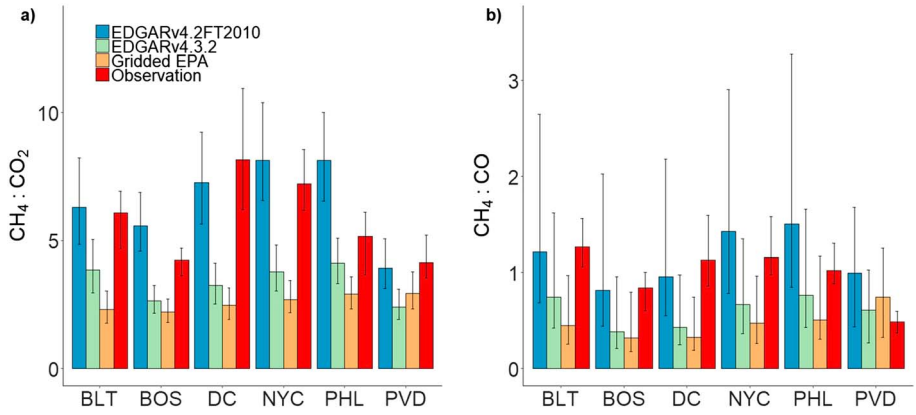

Figure 3. Observed correlation slopes (a) CH_4:CO_2 and (b) CH_4:CO, shown in red, compared to the corresponding inventory-derived ratios for each city. The inventory tracer:tracer values are labeled with the corresponding CH_4 inventory used in the analysis (EDGAR v4.2FT2010 [2010, blue], EDGAR v4.3.2 [April 2010, green], and Gridded EPA [2012, orange]), while the average of the five CO_2 inventories is used to generate a single CH_4:CO_2 inventory estimate. Error bars represent 95% confidence intervals determined using the bootstrap methodology detailed in SI Text S5. EDGAR = Emission Database for Global Atmospheric Research.

the CO_2 inventories. More information about bootstrap methodology used to generate confidence intervals on our estimates is provided in SI Text S5.

For all the cities, the observed CH_4:CO_2 slopes are larger than those generated with the Gridded EPA inventory. The discrepancy for each city, with the exception of Providence and Philadelphia, is more than a factor of 2–3. The Gridded EPA inventory ratios are relatively constant over all the cities, while the observations and inventory estimates of the two EDGAR inventories vary more significantly. The observations show the most agreement with the EDGAR v4.2FT2010 CH_4 inventory. It is important to note, however, that this inventory has been shown to misattribute oil/NG production and processing emissions to urban regions (Maasakkers et al., 2016). Overall distribution by population in EDGARv4.2 is thought to be the cause of the low emissions in oil/NG processing regions. This is evident in our results where for Boston, Philadelphia, and the most populous city, NYC, EDGAR v4.2FT2010 produces a CH_4:CO_2 in excess of that observed (Figure 3). We include this older inventory not to validate it but to rather highlight the discrepancy between the magnitudes of urban CH_4 emissions between the various inventories. While the Gridded EPA inventory agrees well with top-down studies of oil/NG production emissions, our results suggest that urban emissions are considerably higher than those provided in both the Gridded EPA and the EDGAR v4.3.2 inventories. These results suggest the spatial distribution of emissions along the East Coast may qualitatively look more like a combination of the EDGARv4.2 inventory (which captures urban emissions) with the Gridded EPA inventory (which captures emissions in production basins).

4.2. CH_4:CO

CH_4:CO correlations provide an independent estimate of methane emissions as well as a check on the CH_4:CO_2 approach. The benefit of using CO is that prior to spring leaf-out (when our observations were collected), the direct urban plume is primarily a product of anthropogenic activity with little impact from the biosphere. Although CO provides a good confirmation that the biosphere is not dramatically biasing our results, the CO emissions inventories are more uncertain than CO_2 inventories, which are directly tied to fuel usage. We examine two CO inventories (EDGAR v4.3.2 and NEI 2011) and found they differ by a factor of 2 in their emissions estimates for the urban regions (SI Text S3). Multiple previous studies have suggested the National Emissions Inventory (NEI) overestimates urban CO emissions by up to a factor of 2 (Brioude et al., 2013; Miller et al., 2008; Salmon et al., 2018). Our results are consistent with this, where observed CO:CO_2 slopes suggest the EDGAR v4.3.2 inventory matches more closely, whereas the NEI inventory is overestimated by 2×. We thus use the EDGAR v4.3.2 inventory as the CO inventory in our subsequent analysis. We assume an uncertainty of 30% (1σ), estimated from the total national uncertainty in Crippa et al. (2018). Comparing our observed CH_4:CO correlations with those in the inventories, Figure 3b yields a

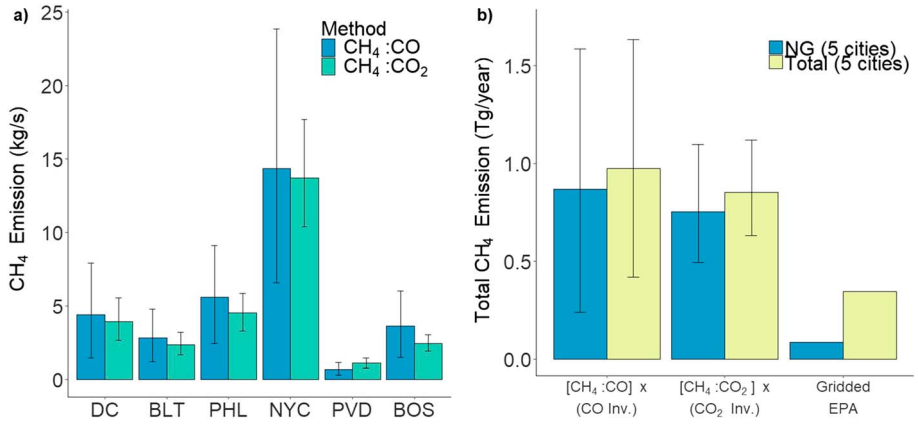

Figure 4. (a) Methane emissions (kg/s) for the six urban regions calculated by using CH_4:CO_2 and CH_4:CO analyses and (b) summed total emissions (Tg/year) for the five largest cities (Providence excluded) compared to Gridded EPA inventory. Uncertainty on the emission estimates is determined using a bootstrap analysis of the observed slopes and inventories to calculate 95% confidence intervals. For the NG emission estimates in (b), the uncertainty in C_2H_6:CH_4 slope and pipeline C_2H_6:CH_4 is also considered in the bootstrap analysis. A more detailed description of the uncertainty propagation is provided in SI Text S5. NG = natural gas.

result consistent with the CH_4:CO_2 slopes, providing independent and robust estimates of methane emissions and suggesting biospheric influence on CO_2 was likely small during our campaign. More detailed consideration of biospheric influence on CO_2 is discussed in SI Text S4.

5. Methane Emissions

The observed correlation slopes are used in conjunction with established CO_2 and CO emissions to estimate the CH_4 fluxes for each city, the results of which are shown in Figure 4a. The total CH_4 emission flux for the six cities in aggregate totals to 0.89 (0.65, 1.16) Tg CH_4/year (95% confidence interval) and 0.99 (0.43, 1.67) Tg CH_4/year (95% confidence interval), using the CH_4:CO_2 and CH_4:CO scaling, respectively. In comparison, the Gridded EPA CH_4 inventory estimates only 0.37 Tg CH_4/year over the same urban domains, more than a factor of 2 lower than our estimates. For context, our results indicate that these six cities combined are a larger emitter than NG production regions such as the Four Corners region (0.54 Tg CH_4/year, Smith et al., 2017; 0.59 Tg CH_4/year, Kort et al., 2014) and the Bakken shale (0.25 Tg CH_4/year, Peischl et al., 2016). Our total CH_4 estimates are broadly consistent with previous studies of Boston (McKain et al., 2015) and the Washington, DC, and Baltimore region (Ren et al., 2018) in showing CH_4 emissions significantly larger than inventory estimates. Considering actual emissions rates, we find some differences with prior studies; for DC/Baltimore, this can be attributed to domain definition, while for Boston, it is unclear what factors may explain the difference (SI Text S6).

Our extrapolation assumes our observations are representative of the full year. Present inventories for anthropogenic urban methane emissions predict little to no seasonality, although some recent observational work has suggested modest seasonality may exist (see SI Text S7 and McKain et al., 2015; Wong et al., 2016). Additional observations of urban regions covering multiple regions/cities and seasons would provide further valuable, direct observational assessment of possible seasonality.

6. Methane Attribution to NG

Previous city-specific studies in Boston (McKain et al., 2015), Los Angeles (Wunch et al., 2016), and Indianapolis (Lamb et al., 2016) attributed the majority of excess urban CH_4 to NG. In this work, the in situ measurement of ethane (C_2H_6) facilitates a similar analysis in which C_2H_6:CH_4 correlations slopes for each city and NG quality data are used to estimate the percentage of methane emissions associated with NG activity, as detailed in SI Text S8. Low ethane signals downwind of Providence result in estimates with large enough uncertainties that we do not include Providence in our NG emission estimates. The remaining five cities emissions total to 0.85 (0.63, 1.12) and 0.97 (0.42, 1.63) Tg CH_4/year, using the CH_4:CO_2 and CH_4:CO

analyses, respectively, and are shown in Figure 4b. Using the partitioning provided by the C_2H_6:CH_4 correlation analysis and the methane estimates discussed in the previous section, the total NG flux observed for the five urban regions (Providence excluded) is 0.75 (0.49, 1.10) and 0.87 (0.24, 1.58) Tg CH_4/year (95% confidence interval) based on the CH_4:CO_2 and CH_4:CO studies, respectively. This comparison is also shown in Figure 4b. The NG emission estimates for these five cities are about a factor of 10 larger than values provided in the Gridded EPA inventory (0.085 Tg CH_4/year) for NG sectors (transmission and distribution) in the study regions. Our observations include all sources of fugitive NG losses in the domain, so these results could indicate underestimates from the transmission and distribution sectors, that currently unaccounted for sources in inventories (such as end-use) are important, or some combination thereof. Our estimate of all NG emissions in these five cities alone is more than the national estimates for local distribution: Alvarez et al. (2018) used the 2015 GHGI estimate based on the results in Lamb et al. (2015) to estimate CH_4 losses from local distribution of 0.44 Tg CH_4/year (U.S. Environmental Protection Agency, 2017), and the Gridded EPA inventory, which is based on the 2016 GHGI for 2012, reports 0.46 Tg CH_4/year of NG distribution losses.

7. Implications

Here we present estimates of urban emissions from a large portion of the U.S. East Coast region, using a simple and robust aircraft-based observation technique showing that large urban CH_4 emissions are not anomalous and limited to just a few cities. According to the Gridded EPA inventory, these six cities (as defined by the population boundaries used in this work) account for ~14.4% of the NG distribution loss for the entire country. A future revision upward of emissions from urban located sectors of the NG supply chain would be necessary to accurately account for the magnitude of emissions directly observed in this study. Applying a similar approach to more strategically representative urban centers could address the question of representativeness and total fugitive losses from local distribution and end use, as the cities examined in this work some of the oldest in the country and may not exemplify other regions. Simplistic in execution, the analysis of emission correlation slopes of urban centers from downwind aircraft measurements presented here provides a robust observational metric to quantify, and potentially track, urban emissions.

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