

## 1 Natural Gas Pipeline Replacement Programs Reduce Methane Leaks 2 and Improve Consumer Safety

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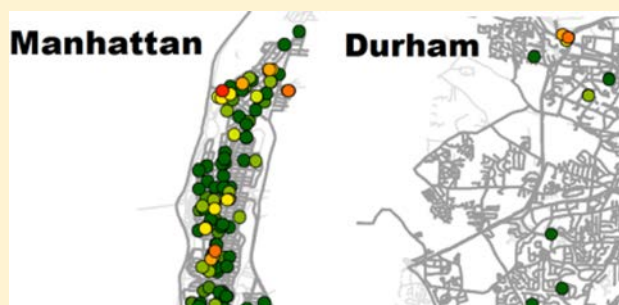
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### 13 **S** Supporting Information

14 **ABSTRACT:** From production through distribution, oil and gas  
15 infrastructure provides the largest source of anthropogenic  
16 methane in the United States and the second largest globally.  
17 Using a Picarro G2132i Cavity Ring-Down spectrometer, we  
18 mapped natural gas leaks across the streets of three United States  
19 cities—Durham, NC, Cincinnati, OH, and Manhattan, NY—at  
20 different stages of pipeline replacement of cast iron and other  
21 older materials. We identified 132, 351, and 1050 leaks in  
22 Durham, Cincinnati, and Manhattan, respectively, across 595,  
23 750, and 247 road miles driven. Leak densities were an order of  
24 magnitude lower for Durham and Cincinnati (0.22 and 0.47  
25 leaks/mi, respectively) than for Manhattan (4.25 leaks/mi) and two previously mapped cities, Boston (4.28 leaks/mi) and  
26 Washington, DC (3.93 leaks/mi). Cities with successful pipeline replacement programs have 90% fewer leaks per mile than cities  
27 without such programs. Similar programs around the world should provide additional environmental, economic, and consumer  
28 safety benefits.



### 29 **■** INTRODUCTION

30 Shale gas and other unconventional natural gas production can  
31 help reduce United States carbon dioxide (CO<sub>2</sub>) emissions if  
32 methane emissions from natural gas infrastructure are  
33 minimized.<sup>1</sup> Emissions during the production, processing,  
34 storage, transmission, and distribution of oil and gas were the  
35 second largest anthropogenic source of methane to the  
36 atmosphere globally in 2013.<sup>2</sup> Such emissions are important  
37 because methane's global warming potential (GWP) is 87 times  
38 greater than that of CO<sub>2</sub> over 20 years and 36 times larger over  
39 100 years.<sup>3</sup>

40 Reducing natural gas emissions during extraction, processing,  
41 and pipeline delivery has additional environmental, economic,  
42 and human health benefits.<sup>4–11</sup> Methane, ethane, and other  
43 hydrocarbons react with nitrogen oxides (NO<sub>x</sub>) and can lead to  
44 tropospheric ozone pollution.<sup>4</sup> The average economic loss of  
45 natural gas leaked or emitted from pipelines in the United  
46 States in 2013 was estimated to be \$2.1 billion.<sup>7</sup>

47 Natural gas pipeline safety in the United States has improved  
48 over recent decades,<sup>6</sup> but rare accidents still occur associated

with aging infrastructure and from excavations and human  
error. In 2014, there were 65 reported gas distribution pipeline  
incidents in the United States, with 18 fatalities, 93 injuries, and  
more than \$73 million in property damage, surpassing the five  
year average (2010–2014) in each category.<sup>6</sup> Such risks and  
impacts to the environment, economy, and human health led  
the U.S. Department of Transportation's (USDOT) Pipeline  
and Hazardous Materials Safety Administration (PHMSA) to  
issue a Call to Action in 2011 to “accelerate the repair,  
rehabilitation, and replacement of the highest-risk pipeline  
infrastructure.”<sup>9</sup> Pipeline age and material (specifically wrought  
and cast iron and bare steel pipelines) are indicators of higher  
risk pipelines frequently targeted for replacement.

A number of studies have shown that age and material type  
of distribution pipelines correlates with leak frequency.<sup>10–14</sup>

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64 Phillips et al.<sup>10</sup> found 3356 methane leaks over 785 road miles  
65 surveyed in Boston, MA, and showed a strong relationship  
66 between the number of leaks per neighborhood and the  
67 number of miles of cast iron mains per neighborhood ( $R^2 =$   
68 0.79). Jackson et al.<sup>11</sup> found 5893 leaks over 1500 road miles in  
69 Washington, DC, that contained about 406 miles of cast iron  
70 mains. Aging natural gas infrastructure, particularly cast iron  
71 and unprotected steel pipelines, presents an opportunity for  
72 economic and environmental benefits if leaks and emissions can  
73 be identified easily and pipelines repaired or replaced  
74 economically.

75 For this study, we mapped natural gas leaks across the streets  
76 of three United States cities—Durham, NC, Cincinnati, OH,  
77 and Manhattan, NY—with different replacement plans and at  
78 different stages of completion (completed replacement,  
79 accelerated replacement, and general replacement, respec-  
80 tively). We compare these leak densities with previously  
81 mapped systems in Boston<sup>10</sup> and Washington, DC,<sup>11</sup> to  
82 examine the efficacy of accelerated pipeline repair and  
83 replacement programs.

## 84 ■ METHODS

85 Between February and July 2014, we surveyed three cities  
86 (Durham, NC, Cincinnati, OH, and Manhattan, NY) for  
87 methane concentration [ $\text{CH}_4$ ] on city streets using a mobile  
88 Picarro G2132i Cavity Ring-Down spectrometer (CRDS)/  
89 surveyor for Natural Gas Module 2 – Investigator unit  
90 (Picarro, Inc., Santa Clara, CA). The methods employed follow  
91 those described in Phillips et al.<sup>10</sup> and Jackson et al.<sup>11</sup> and are  
92 described briefly here. An individual leak or source was defined  
93 conservatively as a spatially contiguous set of [ $\text{CH}_4$ ]  
94 observations greater than 2.5 ppm (i.e., >20% above back-  
95 ground [ $\text{CH}_4$ ] of 1.8–2.0 ppm of  $\text{CH}_4$ ) with a distance  
96 threshold radius greater than 5 m from any other elevated  
97 [ $\text{CH}_4$ ] observation.<sup>10,11</sup> To detect leaks, the methane analyzer  
98 was installed in the back of a vehicle, with a sample line running  
99 from the front bumper of the vehicle to the instrument's sample  
100 inlet. Atmospheric air was sampled about 0.3 m above the road  
101 surface and continuously recorded every approximately 1 s.  
102 Inlet ports were covered with a gas-permeable membrane to  
103 prevent water from entering the system. A GPS and two-  
104 dimensional sonic anemometer (WindSonic; Ultrasonic Wind  
105 Sensor; Gill Instruments, Ltd., Hampshire, U.K.) were installed  
106 on the roof of the vehicle to give real time location (latitude/  
107 longitude) and wind speed and direction data (Tables S1–S3).  
108 The wind data were supplemented with additional wind and  
109 weather data from nearby National Oceanic and Atmospheric  
110 Administration (NOAA) weather stations supplying Quality  
111 Controlled Local Climatological Data (QCLCD), also available  
112 through NOAA's National Climatic Data Center (NCDC)  
113 (Tables S1–S3). The time stamp of the [ $\text{CH}_4$ ] observation was  
114 corrected for the short time lag between sampling at the  
115 bumper inlet and instrument measurement attributable to the  
116 length of the inlet tube. Some New York observations had large  
117 GPS positioning errors attributable to interference of the GPS  
118 signals by tall buildings. Any points that deviated by 10 m or  
119 more from the road observation were removed from our  
120 analysis. Leaks were expressed per city road mile to compare  
121 leak densities. Although the EPA<sup>2</sup> estimates that most methane  
122 losses are from mains rather than service lines (430 Gg  
123 compared to 190 Gg, respectively), we also compared observed  
124 leaks to the number of service lines per mile of main to examine  
125 any effects of higher service line densities in dense urban areas.

The number of service lines per mile of main ranged by a factor  
of two across the five cities: 49, 72, 86, 61, and 103 service lines  
per mile for Durham, Cincinnati, Manhattan, Boston, and  
Washington, DC, respectively.<sup>15</sup> We did not have access to data  
for underground regulator stations, but there are only 560  
across all of New York City and only about 100 in  
Manhattan;<sup>16</sup> as such, they are unlikely to affect our results  
substantively.

To confirm the accuracy and consistency of the concen-  
trations measurements, a 5 ppm [ $\text{CH}_4$ ] standard was measured  
on the instrument periodically throughout the survey, with  
concentration values always within 0.3 ppm of  $\text{CH}_4$ . Addition-  
ally, we measured independent standard sample bags [(1) 5  
ppm,  $-38.0\%$ , (2) 20 ppm,  $-36.8\%$ , (3) zero air (Airgas, Inc.,  
Durham, NC)] periodically to confirm concentration measure-  
ments. Values were always within 0.2, 0.7, and 0.1 ppm of the  
known values for the 5 ppm, 20 ppm, and zero gas standards,  
respectively.

We used the Picarro G2132i Investigator to capture  $\delta^{13}\text{CH}_4$   
of a subset of street sources to differentiate between biogenic  
and thermogenic methane sources. Signatures of  $\delta^{13}\text{CH}_4 >$   
 $-40\%$  (reference to Vienna Pee Dee Belemnite standard)  
generally suggest a thermogenic source for methane, whereas  
 $\delta^{13}\text{CH}_4$  values  $< -60\%$  suggest a biogenically derived  
source.<sup>17,18</sup> During the original surveys, isotopic capture  
measurements were made at seven to eight sites in each city,  
with three and four repeated captures in Cincinnati and  
Durham, respectively, to confirm repeatability ( $1.6 \pm 0.8\%$ ; all  
values mean  $\pm$  s.d. unless otherwise noted). We took additional  
isotopic captures in July for both Durham (eight captures) and  
Manhattan (six captures) several months after the original  
surveys to assess potential changes in isotopic signature. For  
Durham, the average isotopic signature was within 2.5% of the  
original survey ( $-41.3 \pm 2.2\%$  for the March survey and  $-38.8$   
 $\pm 1.7\%$  in July) and similar to a value of  $-41.6\%$  for a direct  
pipeline sample measured on a Picarro G2132i CRDS at the  
Duke Environmental Stable Isotope Laboratory (DEVIL).  
Captures made in Manhattan also showed less than 3.2%  
difference on average ( $-24.3 \pm 2.6\%$  for the May survey and  
 $-27.5 \pm 4.5\%$  in July). Both sets of measurements confirmed  
the sustained presence of the leaks and their thermogenic  
nature. In addition, evacuated cylinders or sample bags were  
collected using a hand pump at a subset of the Durham and  
Manhattan sites visited in July; these samples were then  
analyzed for [ $\text{CH}_4$ ] and  $\delta^{13}\text{CH}_4$  on the Picarro G2132i CRDS  
at the DEVIL within 2 days of sampling to compare field  
isotopic measurements with laboratory measurements. Labo-  
ratory analyses of bag and cylinder samples were  $2.7 \pm 1.1\%$   
(mean  $\pm$  s.e.;  $n = 6$ ) lighter than car field measurements,  
suggesting a slight bias in the driving instrument (likely  
attributable to ethane interference) but not large enough to  
alter determinations of thermogenic versus biogenic sources.

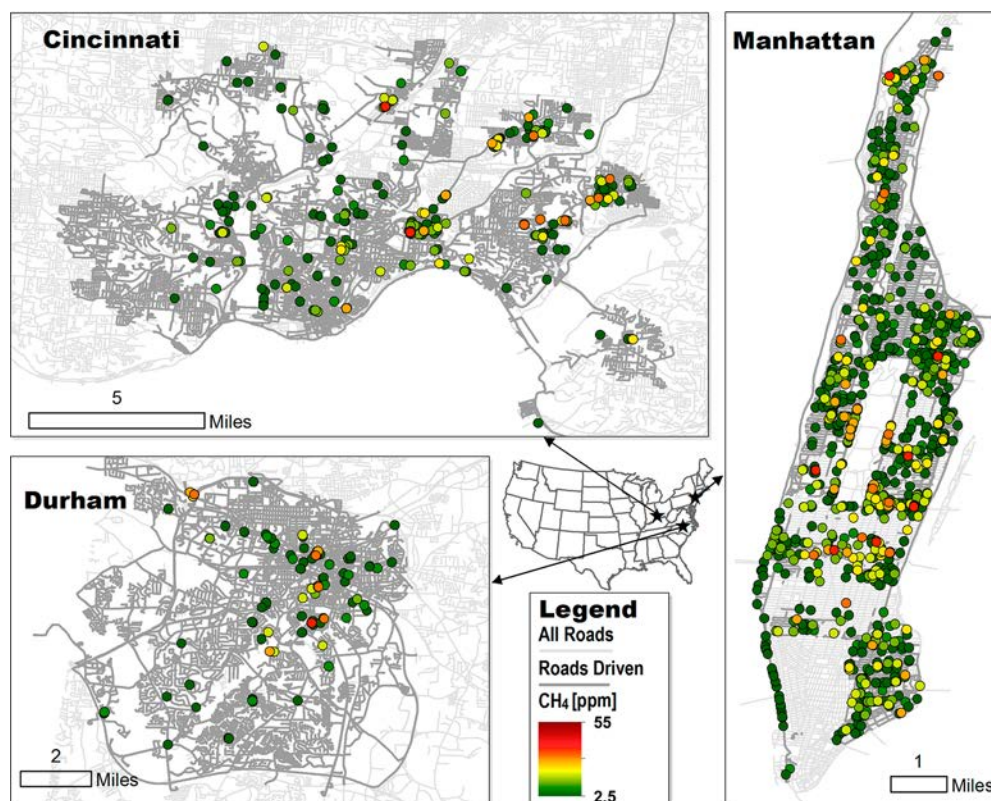
Standard sample bags filled with either 100 ppm,  $-36.8\%$   
(Airgas, Inc., Durham, NC) or 2500 ppm,  $-66.5\%$  (Airgas,  
Inc., Durham, NC)  $\delta^{13}\text{CH}_4$  standard gases were used to release  
small puffs of gas standard near the bumper inlet to simulate a  
plume. On average, the isotope capture was  $-1.0\%$  heavier  
than the known delta for the 100 ppm standard and  $-1.5\%$   
heavier than the known delta for the 2500 ppm standard  
( $-35.84 \pm 0.86\%$  ( $n = 5$ ) and  $-65.03 \pm 0.78\%$  ( $n = 4$ ),  
respectively).

In June and July of 2015, we carried out two follow-up field  
campaigns to gather additional data. To confirm that most of

Table 1. Street Leak Comparison of Five Major United States Cities<sup>a</sup>

city	road miles driven	total # of leaks	leaks/mile	leaks >5ppm	leaks >10ppm	leaks >25ppm	mean (ppm)	median (ppm)	max (ppm)	% rep. can. (mains)	% rep. can. (service lines)	service lines/mi of main (#/mi)	leaks/mi normalized by service lines/mile
Washington, DC	1,500	5,893	3.93	1122	334	67	4.6	3.1	88.6	43%	25%	103	0.0381
Boston, MA	785	3,356	4.28	435	97	1	3.7	2.9	28.6	37%	23%	61	0.0706
Manhattan, NY	247	1,050	4.25	186	53	11	4.5	3.1	60.0	52%	23%	86	0.0493
Cincinnati, OH	750	351	0.47	66	19	5	4.7	3.1	54.3	2%	12%	72	0.0065
Durham, NC	595	132	0.22	24	10	4	4.7	3.0	33.1	0%	0%	49	0.0045
Durham, NC, 2015	145	46	0.33	5	4	0	3.8	2.9	12.7	0%	0%	49	0.0045

<sup>a</sup>Percent replacement candidate for mains and service lines calculated from PHMSA data<sup>15</sup> for the year of each study (2014 for Manhattan, Durham, and Cincinnati; 2013 for Washington, DC;<sup>11</sup> 2011 for Boston<sup>10</sup>). The second Durham entry reflects the results of 145 miles of the city re-driven at night in 2015.

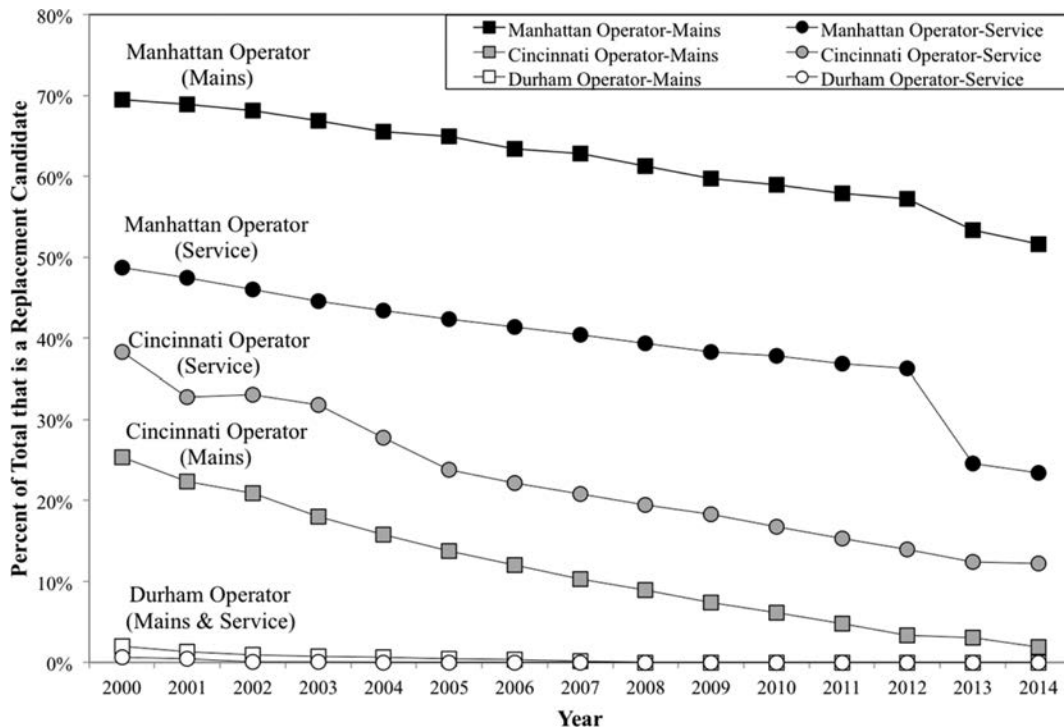


**Figure 1.** Maps of methane leaks surveyed in Cincinnati, OH (top left), Durham, NC (bottom left), and Manhattan, NY (right). Roads driven are outlined in darker gray, with leak locations marked by colored circles for CH<sub>4</sub> concentration. Note that the map scales vary for the three cities. See Table 1 for information on road miles driven, total leaks, and leaks per mile.

189 the observed leaks came from underground infrastructure  
 190 instead of other city infrastructure, including buildings,  
 191 aboveground meters, and other sources, we identified the  
 192 source of all leaks identified in four sections of Manhattan  
 193 (Figure S1, Table S4). For each of the 42 leaks identified, we  
 194 used a flame ionization detector (Dafarol A-600; Dafarol  
 195 Associates, Hopedale, MA) and a Bascom–Turner combustible  
 196 gas analyzer to locate and attribute the source of each leak  
 197 detectable from the survey. The second field campaign was to  
 198 eliminate any possibility that the leak densities observed in  
 199 Durham were associated with higher wind speeds or any other  
 200 weather conditions. We re-drove 145 road miles of Durham  
 201 (24% of the original survey) in five areas from 10pm to 8am

(July 27–29, 2015) (Figure S2). During this nighttime window, 202  
 the air was still (Table S5), which would lead to a maximum 203  
 number of leak detections. Leak locations and densities were 204  
 then compared to results from the 2014 survey. 205

To provide context for the city observations, we analyzed 206  
 pipeline material data collected by the USDOT’s PHMSA 207  
 annually from gas distribution operators ([www.phmsa.dot.gov](http://www.phmsa.dot.gov) 208  
 – Distribution, Transmission & Gathering, LNG, and Liquid 209  
 Annual Data) for the years 2000 to 2014.<sup>15</sup> We analyzed 210  
 pipeline materials data from 2013 for all United States states 211  
 from 2000 to 2014 for distribution operators that service 212  
 Manhattan (Operator ID: 2704), Cincinnati (ID: 2364), and 213  
 Durham (ID: 15938)<sup>19–21</sup> and from 2013 for Washington, DC 214



**Figure 2.** Percent of total pipelines that are replacement candidates in different areas. The figure shows the percentage of the total miles of main or service lines from 2000 through 2014 that are replacement candidates (unprotected bare or coated steel, cast/wrought iron, ductile iron, copper, and other) for Manhattan, NY, Cincinnati, OH, and Durham, NC, as reported by company.<sup>15</sup>

(ID: 22182) and 2011 for Boston (ID: 2652),<sup>21</sup> the years of sampling for each city (Table 1). In addition to reporting total miles of mains and number of service lines, gas distribution companies report miles of main or number of service lines by material type (unprotected bare steel, unprotected coated steel, cathodically-protected bare steel, cathodically-protected coated steel, plastic, cast/wrought iron, ductile iron, copper, and other). Materials that are considered “replacement candidates” are defined as unprotected bare or coated steel, cast/wrought iron, ductile iron, copper, and other. We calculated the percent replacement candidate as the miles of mains that are replacement candidates per total miles of mains or the number of service lines that are replacement candidates per total number of service lines. Percent replacement candidate is calculated with data collected for the year of each study: 2014 for Manhattan, Cincinnati, and Durham; 2013 for Washington, DC; 2011 for Boston. PHMSA data are the only publicly available data source for such analyses but should be interpreted with caution. Changes in the amount of pipeline materials for a given operator can arise not just through replacement programs but through reclassification (e.g., when an operator realizes that a protective steel coating is no longer functioning) or when companies merge or sell assets.

## RESULTS AND DISCUSSION

We observed 132, 351, and 1050 street leaks for Durham, Cincinnati, and Manhattan, respectively, across the 595, 750, and 247 road miles surveyed in each city (Figure 1). Leak densities were an order of magnitude lower for Durham and Cincinnati (0.22 and 0.47 leaks/mi, respectively) than for Manhattan (4.25 leaks/mi) and for those observed previously in Boston (4.28 leaks/mi) and Washington, DC (3.93 leaks/mi) (Table 1). Manhattan also had 3 to 5 times more high-concentration leaks (>10 ppm) than Cincinnati or Durham

despite having less than half the road miles surveyed. 248  
Manhattan had 53 leaks with concentrations greater than 10 249  
ppm of CH<sub>4</sub> (Table 1). Cincinnati and Durham had only 19 250  
and 10 leaks greater than 10 ppm of CH<sub>4</sub>, respectively. 251  
Manhattan also had the highest CH<sub>4</sub> concentration observed 252  
across the three cities, 60 ppm, compared to maximum 253  
observed values of 54 and 33 ppm in Cincinnati and Durham, 254  
respectively (Table 1). When leak densities were normalized by the 255  
number of service lines per mile of main, Durham and 256  
Cincinnati still had 5- to 10-fold lower values than Manhattan, 257  
Boston, or Washington, DC (0.0045, 0.0065, 0.0493, 0.0706, 258  
and 0.0301 leaks per service line, respectively). 259

The resurvey of Durham roads in 2015 during the still, 260  
nighttime conditions (Figure S2, Table S5) found a higher leak 261  
density than in 2014 but confirmed that Durham had the 262  
lowest leak densities of any city in the survey. Across 145 road 263  
miles driven (24% of the original dataset), we found 46 leaks at 264  
concentrations of 2.5 to 13 ppm of CH<sub>4</sub>, with only 5 leaks 265  
greater than 5 ppm. The observed leak density was 0.33 leaks 266  
per mile, 50% higher than in the daytime conditions of 2014 267  
when the leak density for Durham was estimated to be 0.22 268  
leaks/mi (for both the full city survey and the subset of roads 269  
re-driven in 2015). Manhattan’s leak density of 4.25 leaks/mi 270  
was still 13 times higher than the revised nighttime survey of 271  
Durham. 272

Real-time isotopic measurements showed that the observed 273  
CH<sub>4</sub> came from thermogenic rather than biogenic sources. 274  
Durham leaks had the lightest δ<sup>13</sup>CH<sub>4</sub> signature of the three 275  
cities surveyed (−41.3 ± 2.2‰) but were still considerably 276  
heavier than biogenic sources. Cincinnati and Manhattan CH<sub>4</sub> 277  
leak signatures were even heavier (−36.1 ± 2.6‰ and −24.3 ± 278  
2.6‰, respectively). In comparison, biogenic isotope values 279  
ranged from −53.1 to −64.5‰ for eight landfill, wetland, and 280  
sewage treatment sites in Boston, MA, sampled previously.<sup>10</sup> 281

282 A detailed sampling of leaks in four randomized regions of  
283 Manhattan showed that emission sources from under streets  
284 rather than from buildings or other aboveground sources were  
285 responsible for the leaks observed. Of the 42 leaks surveyed to  
286 isolate the source (Figure S1), 41 (98%) clearly originated from  
287 street infrastructure, including manhole covers, valve boxes, and  
288 other locations (Table S4). The source of only one leak was  
289 ambiguous, as both a building fan and a street repair showed  
290 elevated concentrations of methane.

291 Accelerated pipeline replacement programs help explain the  
292 order-of-magnitude lower densities of leaks observed in  
293 Durham and Cincinnati compared to Manhattan (Table 1;  
294 Figure 1) and to Boston, MA,<sup>10</sup> and Washington, DC.<sup>11</sup> The  
295 percentage of replacement candidate mains and service lines  
296 was strongly related to leak densities for the five cities overall  
297 ( $r^2 = 0.95$  and  $0.85$ , respectively). Durham, which had the  
298 lowest density of leaks observed here (0.22 leaks/mile; Table  
299 1), had replaced all of its cast iron and unprotected steel natural  
300 gas pipelines by 2008;<sup>22</sup> all mains in its distribution system are  
301 now either plastic (60%) or cathodically treated coated steel  
302 (40%). Similarly, an accelerated pipeline replacement program  
303 in Cincinnati,<sup>23</sup> a city with only 0.47 leaks/mi (Table 1), is  
304 almost complete, with only 3% of cast/wrought iron mains  
305 remaining across its network; the remaining 97% of its mains  
306 are comprised of plastic (50%) and cathodically protected  
307 coated steel (47%). Replacement candidate pipelines have  
308 steadily decreased in miles remaining for all three cities  
309 surveyed here, but they are much lower in Durham and  
310 Cincinnati than in Manhattan, Boston, or Washington, DC  
311 (Table 1; Figure 2). Continued replacements should help  
312 reduce CH<sub>4</sub> emissions from urban infrastructure.<sup>24</sup> A recent  
313 analysis in Boston, for instance, showed that the average  
314 regional CH<sub>4</sub> flux was  $18.5 \pm 3.7$  g CH<sub>4</sub> m<sup>-2</sup> y<sup>-1</sup>, with 60–  
315 100% attributable to natural gas losses; the average fractional  
316 loss to the atmosphere from all downstream components of the  
317 natural gas system was  $2.7 \pm 0.6\%$ , more than double the 1.1%  
318 estimate from the most comparable state inventory.<sup>25</sup>

319 In states such as Ohio, North Carolina, and Indiana,  
320 accelerated pipeline repair and replacement programs have  
321 resulted from partnerships among companies, states, municipal-  
322 ities, and public utility commissions. A partnership between  
323 distribution companies and the Ohio Public Utility Commis-  
324 sion, for instance, which sets cost recovery rates for natural gas  
325 pipeline repairs, is the reason that Cincinnati, OH, is on track  
326 to complete its replacement of pipeline mains by 2015 (the  
327 original goal) and of service lines before 2020, based on a linear  
328 projection of the data (Figure 2).<sup>23</sup> At the opposite end of the  
329 spectrum, replacement rates in Baltimore, MD, have been  
330 among the slowest in the United States, with about 140  
331 additional years projected to full replacement based on  
332 replacement rates between 2004 and 2013<sup>11</sup> (and acknowl-  
333 edging recent programs in Maryland to speed pipeline  
334 replacements). Manhattan falls somewhere in between. There,  
335 the New York distribution company maintained a fairly steady  
336 rate of 1–2% replacement for both mains and service lines from  
337 2000 to 2014 (with an unusually, and possibly unreasonably,  
338 high replacement rate reported in 2013; Figure 2). On the basis  
339 of an approximate linear projection of the data in Figure 2, it  
340 will take another 26–52 years for mains or 11–23 years for  
341 service lines for completion. Between 2000 and 2014 the New  
342 York distribution company decreased its portion of mains and  
343 service line replacement candidates by 26% and 52%,  
344 respectively, a substantial improvement.

Overall, natural gas pipeline safety is improving across the  
United States, and the miles of distribution replacement  
candidate pipelines are decreasing. The number of gas pipeline  
incidents causing death or major injury dropped by half  
between 1991 and 2011, from about 70 incidents per year on  
average to around 35.<sup>6</sup> Of the approximate 2,150,000 miles of  
gas distribution lines (mains and service) in the United States  
in 2013, 7% of mains and 9% of service lines were replacement  
candidates in 2013, down from 12% and 14%, respectively, in  
2000.<sup>21</sup> In fact for most states, less than 10% of main and  
service pipelines are now replacement candidates (41 and 35  
states, respectively). The greatest concentration of replacement  
candidate pipelines is in the northeastern United States, where  
infrastructures are generally older (Figure S3). Continued and  
sustained progress in natural gas pipeline replacements and  
repairs, implemented with an eye to detection and cost, will  
improve safety and air quality and reduce greenhouse gas  
emissions.

## ■ ASSOCIATED CONTENT

### 📄 Supporting Information

The Supporting Information is available free of charge on the  
ACS Publications website at DOI: 10.1021/acs.estlett.5b00213.

Figure S1 shows the 42 street locations in Manhattan  
selected for a survey to determine the source of natural  
gas sources. Figure S2 maps the 145 road miles re-driven  
at night in Durham, NC, in July of 2015 and the  
locations with concentrations >2.5 ppm of CH<sub>4</sub>. Figure  
S3 shows United States states with high concentrations  
of replacement candidate mains in 2013. Tables S1, S2,  
and S3 present wind speed and weather data during the  
2014 driving campaigns for Durham, NC, Cincinnati,  
OH, and Manhattan, NY, respectively. Table S4 shows  
results for the source identification survey of Manhattan  
in July 2015 (Figure S1). Table S5 presents wind speed  
and weather conditions during July 2015 for the Durham,  
NC, re-drive of 145 road miles at night (NOAA Quality  
Controlled Local Climatological Data, Durham, NC, and  
Raleigh-Durham International Airport, NC, stations).

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### Notes

The authors declare no competing financial interest.

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