

Home is Where the Pipeline Ends: Characterization of Volatile Organic Compounds Present in Natural Gas at the Point of the Residential End User

 pubs.acs.org/doi/full/10.1021/acs.est.1c08298

Synopsis

Natural gas delivered to homes contains trace amounts of nonmethane gases that can impact air quality and human health at any point where natural gas is leaked.

U.S. oil and natural gas (O&NG) production has grown substantially since 1990 and now makes up over 69% of the total U.S. energy consumption. Natural gas (NG) has become the dominant energy source for nearly all-consuming sectors in the U.S., including industrial (41%), commercial (38%), residential (42%), and electrical power generation (33%). (1) Unprocessed NG can vary in the methane (CH_4) fraction (typically 60–90%), (2) with the remaining fraction consisting of impurities including a suite of nonmethane volatile organic compounds (NMVOCs; e.g., alkanes, cycloalkanes, and aromatics) and nonorganic compounds (e.g., hydrogen sulfide, carbon dioxide, water vapor, nitrogen, and helium). (2,3) An estimated 2.49 million tons of VOCs are emitted annually from upstream oil and NG production processes alone, making it the largest anthropogenic source of VOCs in the U.S. (4) The presence of certain NMVOCs in NG, including the aromatic compounds benzene, toluene, ethylbenzene, and *ortho*-, *meta*- and *para*-xylenes (collectively BTEX), are particularly relevant given their toxicity, carcinogenicity, and/or atmospheric reactivity as precursors to both ozone and secondary organic aerosol (SOA) formation. (5) While the presence of BTEX in unprocessed NG is well documented, (5–7) the degree to which BTEX is present in distribution-grade NG when delivered to end users is largely uncharacterized due to lack of direct measurements; for example, a 1994 study noted the presence of benzene in end-use NG, but this was inferred through ambient air measurements rather than directly from the end use gas stream. (8) A more definitive characterization of NMVOCs in the end use NG stream is, therefore, warranted, as several NMVOCs could produce air quality impacts and human health risks at any point where NG is leaked or incompletely combusted. U.S. pipeline operators and local gas distribution companies employ standardized gas chromatography (GC) compositional analytical procedures (i.e., ASTM D1945 or ASTM D7833) on processed NG to determine the heating value and to satisfy tariff gas quality specifications consistent with the North American Energy Standards Board (NAESB) standards. These methods, however, typically focus only on the 16 most abundant constituents (e.g., [Supporting Information Table S3](#)); in addition, they generally do not differentiate C6+ organic compounds except for hexane where available. NG utilities do perform more extensive trace gas analyses on occasion; however, these data are typically not

publicly available. (3) In 1992, the Gas Research Institute (now the Gas Technology Institute) conducted a large-scale NG characterization campaign collecting 6800 samples in 26 major U.S. cities to better determine fuel efficiency related to NG-powered vehicles. While the study found substantial spatial and temporal variability of C₁–C₃ and inert gases among and within cities, constituent analyses were not differentiated for C₄+ hydrocarbons (i.e., “butanes plus”); however, a national mean C₄+ concentration of 0.4 (mol %) was observed with a maximum of 2.1 (10th–90th percentile: 0.1–0.6) indicating the widespread presence of heavier hydrocarbons. (9) More recently, a study by the National Institute of Standards and Technology identified BTEX and other NMVOCs in distribution-grade NG; however, only two samples were analyzed from the laboratory gas valve supplied by the local NG distribution system in Colorado. (3) Two other NG compositional analyses found that underground storage and transmission pipeline NG consisted of approximately 93% methane and ~1% VOC; (7) however, it is undetermined whether the NG composition in these midstream systems is fully representative of downstream systems.

As federal vehicle emissions requirements continue to regulate NMVOC emissions, nontransportation sources of NMVOCs have become increasingly important and have garnered significant attention in recent years. (10,11) Given the limited characterization of trace gas-phase compounds in local NG distribution systems, the widespread use of NG, and the numerous studies that have now confirmed NG distribution systems as the largest source of urban methane, (12–23) we performed a 16 month hazard identification sampling campaign in the Greater Boston region to characterize NMVOC constituents and odorant content in distribution-grade NG. As sampling from distribution-NG infrastructure (e.g., pipelines or gas processing plants) is not possible without operator coordination, we instead accessed the many endpoints of the distribution system via residential NG stoves and building risers. The methods provide a safe and reliable approach for sampling whole NG at the point of end users using commercially available the United States Environmental Protection Agency (USEPA) methods. The results demonstrate methane, ethane, mercaptan odorant, and NMVOC whole NG characterization throughout Greater Boston, MA, with implications for air quality at any point where NG is leaked or incompletely combusted.

Sample Collection and Processing

All sampling procedures and safety protocols were IRB approved with an additional approval from Harvard’s chief research compliance officer. This project received IRB approval on October 29, 2019, from the Harvard T.H. Chan School of Public Health, ref: IRB19-1587 titled “Contaminants in the Kitchen.”

Whole NG samples were collected from indoor NG-fired stovetop appliances and outdoor gas appliance lines across three local NG distribution territories. VOCs were analyzed by GC/mass spectrometry via grab samples of whole NG collected in evacuated 1.4L Entech Silonite-lined canisters prepared by a commercial environmental testing lab (Phoenix Environmental Laboratories, Manchester, CT) according to the USEPA Method TO-15. To

verify sufficient sample capture, a subset of samples was tested for CH₄ and C₂H₆ using the USEPA Method 3C performed by New England Testing Laboratory, West Warwick, RI (managed internally by Phoenix Environmental Laboratories). Analytical methods for ASTM D1945, ASTM D1946, and EPA 3C are identical, though differ by QC criteria and compounds specified for each method. Tentatively identified compounds (TICs) were also reported for nontarget compounds including thiol-group compounds, such as common NG odorants (e.g., *tert*-butyl mercaptan [TBM; CAS no. 75-66-1], and isopropyl mercaptan [IPM; CAS no. 75-33-2]). TIC identification and estimation generally followed USEPA's National Functional Guidelines for Superfund Organic Methods Data Review. (24) As per this guidance, a compound must meet a set of 10 analytical criteria and 9 QA/QC evaluation checks to be considered a TIC. Most notably, the spectral peak for a TIC must have an area or height >10% of the area or height of the nearest internal standard and must receive a spectral library search match of 80% or higher to meet the definition of a "probable match." The concentration of a TIC can then be calculated normally by using total ion areas for both the TIC peak and the internal standard with the closest chromatographic retention time. Therefore, tentatively identified sulfur compounds herein are considered "NJ" flagged in that results are qualitatively identified and reported as estimated concentrations based upon the nearest internal standard. (24) While ASTM D5504—the standard test method for determining sulfur compounds in NG was not employed—fused-silica lined canisters (i.e., silonite) have been well validated for their suitability for volatile sulfur compound collection. (25)

Stovetop sample collection entailed a direct in-line connection between the stove's NG outlet and the sample canister via flexible Teflon-lined tubing ([Figure S1](#)). For all stoves sampled, the tubing diameter fully encompassed the gas outlet orifice. The in-line connection effectively bypassed the stovetop ignition source (where present) and ensured that the sample was not diluted by ambient air and that very minimal unburned NG was released. Building NG appliance line sampling was identical to stovetop sampling, except samples were collected from building risers that were typically used to connect to outdoor NG grills or firepits. Canisters were sampled within 5 days of arrival and returned within 3 days of sample collection. All lab analyses were completed within 3 weeks of sampling, and chain of custody forms were maintained for all samples. Samples were donated by homeowners since they were collected after NG was paid for and delivered to the home (i.e., after or "behind" the residential gas meter). See the [Supporting Information](#) Section 1.1 for details on sample collection methodology, including additional sampling safety precautions.

Sampling Design

A spatially-stratified sample design was employed to represent the three major local distribution companies (Columbia Gas, Eversource, and National Grid—see [Figure 1](#)) that operate across the Greater Boston region. The sampling campaign was originally designed to capture 90 unique sampling locations—sampled twice in each of the cooling and heating seasons—in addition to 10 unique locations (100 total sampling sites) sampled approximately

monthly (“monthly”) for 12 months to resolve temporal variability; however, the COVID-19 pandemic required an alteration to this sample design. Sampling was suspended on March 12, 2020, in response to the COVID-19 pandemic. By that time, our research team had collected 109 samples from 81 unique locations across our six sampling areas and approximately 4 months of sampling at the 10 “monthly” locations. Abbreviated sampling activities were reinstated on June 26, 2020; however, our original sampling plan was altered as follows. First, additional sampling was limited to “monthly” stovetop sampling for pre-existing and willing participants that were repeated monthly for 11 months. We also recruited 20 new participants within our target areas who had an accessible outdoor NG service line typically connected to an outdoor NG-fired grill (i.e., not propane). In addition, two stove participants switched to sampling from their outdoor NG grill. Each of these NG grill locations were sampled at least twice.

Figure 1

Figure 1. Unique sample locations (black markers) by municipality (red outline) across the three major Greater Boston NG utility providers. Note, some locations were sampled multiple times. *Utility A was acquired by Utility B on October 13, 2020.

Data Filtering Methodology

The final analysis-ready data set (L3) contained 234 individual samples collected from 69 unique residential NG grills and NG stoves. The original raw data set (L1) contained 312 samples and underwent two iterations of quality control and filtering to ensure that all final data were representative of end use whole NG. While all post-COVID pause samples included methane and ethane analyses, only the last 19 of the 109 pre-COVID pause samples were tested for ethane and methane. Therefore, in the absence of methane and ethane measurements, we combined the pre- and post-COVID pause samples in an analytical framework to maximize our confidence in sample capture sufficiency without unnecessarily rejecting valid samples lacking methane content measurements. Correlation matrices indicated NG capture was highly positively correlated with the extent to which an evacuated sample canister was filled and highly negatively correlated with the length of time to fill a canister (See the [Supporting Information](#) Figures S4 and S5). See the [Supporting Information](#) Section 1.2 for additional details on the data filtering results and methodology.

Sample Collection QA/QC

A total of four sampling blanks were collected (two field blanks, two sample tubing blanks); all blanks were returned clean (i.e., criteria compounds were nondetects) and were removed from the main analysis data set. In addition, seven sample duplicates were collected (four from stoves, three from grills) and were retained as additional samples in the main analysis. Using BTEX as a replicate precision indicator, the median percent error aggregated across

each set of sample duplicates was 1.7, 2.6, 5.8, and 8.1 for each of the four BTEX constituents, respectively. As per USEPA method guidance, these precisions indicate acceptable repeatability and are also within one standard deviation of the BTEX means from the full data set (benzene: $\pm 9.7\%$; toluene: $\pm 12\%$; ethylbenzene: $\pm 14\%$; xylenes: $\pm 13\%$; see [Table 1](#)).
Table 1. Top 15 VOCs (Standard TO-15 Suite) with Statistical Measures and Confidence Intervals Derived from a 1000-Sample Bootstrap of Data from 234 Total Whole NG Samples^a

VOC [CAS] (units)	<i>n</i> (% detect)	Mean	SD	95% LCI	95% UCI
methane [74-82-8] (%) _{b,d}	184 (79) _c	101	1.0	99.2	103
ethane [74-04-0] (%) _b	184 (79) _c	2.07	0.05	1.98	2.17
hexane [110-54-3] (ppbv)	229 (98)	567	61	460	696
benzene [71-43-2] (ppbv)	223 (95)	165	16	136	195
toluene [108-88-3] (ppbv)	220 (94)	151	17	119	197
heptane [142-82-5] (ppbv)	219 (94)	267	26	220	324
cyclohexane [110-82-7] (ppbv)	208 (89)	215	20	176	254
<i>m,p</i> -xylene [108-38-3; 106-42-3] (ppbv)	175 (75)	58	7.2	44.9	73
ethanol [64-17-5] (ppbv)	153 (65)	126	18	92.4	164
<i>o</i> -Xylene [95-47-6] (ppbv)	148 (63)	17.5	2.4	13.1	22.4
ethylbenzene [100-41-4] (ppbv)	131 (56)	12.8	1.7	9.64	16.2
1,2,4-trimethylbenzene [95-63-6] (ppbv)	108 (46)	7.86	1.2	5.62	10.3
4-ethyltoluene [622-96-8] (ppbv)	95 (41)	6.02	0.88	4.35	7.68
1,3,5-trimethylbenzene [108-67-8] (ppbv)	66 (28)	3.04	0.51	2.08	4.06
acetone [67-64-1] (ppbv)	37 (16)	10.3	5.4	2.71	22.1
isopropylbenzene [98-82-8] (ppbv)	31 (13)	0.764	0.2	0.386	1.17
1,2-dichloroethane [107-06-2] (ppbv)	24 (10)	0.0652	0.039	0.0129	0.159

^a

VOCs reported as parts per billion by volume (ppbv). A subset of samples was analyzed for methane and ethane using the EPA Method 3C and are reported as vol/vol %. The sample size and frequency of detection are noted by *n* and the associated %.

Methane and ethane tested according to the EPA method 3C/modified 3C via aliquot from the original sample canister.

^c

Methane and ethane were only regularly tested in samples after the COVID sampling pause. 77% detection reflects the lack of testing rather than implying sampling nondetects.

^d

At high concentrations, reported Methane results can slightly exceed 100% but are within the margin of error for the EPA method 3C. The noise associated with translating GC peak areas to concentrations based on a calibration response curve (area/ppmv) can lead to calculated concentrations that are >100% when sample concentrations are close to 100%.

Ethane: Methane Ratio

Ethane (C₂H₆) is typically used as a thermogenic CH₄ tracer for attributing CH₄ measurements to NG sources in the absence of other appreciable sources of thermogenic methane (McKain et al., 2015). We obtained C₂H₆/CH₄ ratios for 184 samples as an additional indicator for sufficient NG sample collection.

VOC Emissions from NG in Great Boston Region

To estimate emissions of VOCs from NG systems throughout the Greater Boston region, we bootstrapped mean ratios of VOCs to whole NG-CH₄ from our data set in combination with previous regional CH₄ emissions apportioned to NG systems over the approximately 90 km Greater Boston radius. (17,20,22) Following similar methods to Marrero, Townsend-Small, (26) and Deighton, Townsend-Small, (27) emission rates (kg yr⁻¹) of BTEX associated with CH₄ from NG leaks were calculated according to eq 1, where the CH₄ flux (kg yr⁻¹) is directly taken from each of McKain, Down, (17) Plant, Kort, (20) and Sargent, Floerchinger; (22) MW is the molecular mass of CH₄ and each BTEX constituent in g mol⁻¹; and [BTEX]/[CH₄] is the bootstrapped mean ratios of each BTEX constituent (ppbv) to CH₄ (ppbv) across our field sample data set. See the [Supporting Information](#) Section 2.9 for additional details on this calculation.

(1)

Results and Discussion

Overview of Final Analysis Data Set

We initially collected 312 NG samples from 99 unique households throughout the Greater Boston region. After removal of flagged samples and blanks, along with two layers of quality control, 234 whole NG samples across 69 locations were retained, reflecting a 77% sample retention (see the [Supporting Information](#) Section 1.2 for details on QC data processing).

Unique sample locations were spatially representative across the three major local distribution companies (utility A, $n = 20$; utility B, $n = 20$; utility C, $n = 29$; see [Figure 1](#)). Two separate winter (heating) seasons were sampled, spanning December 2019 to May 2021. In addition, locations were sampled throughout the December 2019 to May 2021 period to produce an 18 month time series (barring a 4 month sampling pause from April to July 2020 due to restrictions during the COVID-19 pandemic).

Verifying Sample Representation of the NG Stream

Whole NG sample collection was verified using multiple analytical markers. Of the 234 L3 samples, 184 were analyzed for CH_4 and C_2H_6 . 92% of the samples measured for CH_4 and C_2H_6 exhibited a sample CH_4 concentration of 90% or greater. The bootstrapped mean CH_4 concentration of the L3 data set was 101% (95% CI: 99.2–103%). Note that at concentrations that approach 100% CH_4 , reported CH_4 results can slightly exceed 100% due to measurement and calculation noise; however, these concentrations are within the margin of error for the EPA method 3C (see explanation in [Table 1](#)). Importantly, unlike many of the accompanying NMVOC compounds that exhibited a high skew, the raw $\text{C}_2\text{H}_6/\text{CH}_4$ ratios approach a normal distribution with a median ratio of 1.89 (median absolute deviation: 0.34%), indicating that regardless of the individual sample CH_4 content, the CH_4 itself was NG in origin ([Figure S7](#)). Our C_2H_6 and CH_4 results agree with posted pipeline hourly data and previous research showing Boston NG pipeline $\text{C}_2\text{H}_6/\text{CH}_4$ ratios of 2.04% (2.01, 2.07%) with a corresponding NG percentage of 110 (68,170). ([20](#)) Additionally, two of the primary NG odorant compounds were frequently detected: TBM was detected in 97% of samples, and IPM was detected in 81% of samples (see below). Finally, the iso-pentane to *n*-pentane ($i\text{C}_5/n\text{C}_5$) ratio has been utilized elsewhere as a source signature of O&NG. ([28](#)) Using reduced major axis (RMA) regression, we found an $i\text{C}_5/n\text{C}_5$ ratio of 1.48 (95% CI: 1.44–1.52; $R^2 = 0.95$) that approximates enhancement ratios of raw NG downwind of O&NG operations (0.82–1.10) and is distinctly different than the $i\text{C}_5/n\text{C}_5$ ratio of 2.96 consistently reported for sources related to fresh gasoline. ([29](#)) Collectively, these analytical markers indicate a high fidelity of whole NG sample capture of the L3 data set with minimal ambient air intrusion and residential NG as the source of NMVOCs.

NMVOC Profile of End Use NG Samples and Implications for Exposure to Hazardous Air Pollutants

From the L3 data set, 296 unique constituents were detected in end use NG, of which 21 (or approximately 7%) are designated as hazardous air pollutants (HAPs) (see accompanying analytical results). The top 15 VOCs and TICs by abundance are shown in [Table 1](#) and [Supporting Information Table S2](#), respectively. Given the long-tailed distribution, statistics were derived from bootstrapping (1000 resamplings). We found that benzene and toluene are persistent in distribution-grade NG at the point of the end user ([Table 1](#)). Benzene (mean = 165 ppbv; SD = 16; 95% CI 136–195) and toluene (mean = 151 ppbv; SD = 17; 95% CI 119–197) were detected in 95 and 94% of samples, respectively. Hexane, heptane, and

cyclohexane were also commonly detected at 98, 94, and 89%, respectively (Table 1). Overall, the mean total NMVOCs in distribution-grade NG throughout the study area was ~6.0 ppmv (95% CI 5.5–6.6; max 21.4 ppmv); the NMVOC total for each sample is the sum of all TO-15 and TICS NMVOCs, excluding ethane. While the concentrations of BTEX observed in NG herein are likely lower compared to other source types, the proximity of these emissions to populations—and the widespread use of NG indoors warrants further exposure assessment. Moreover, given benzene’s genotoxicity, no safe level of benzene exposure can be recommended, which is often reflected in its relatively low reference exposure levels. For example, the Commonwealth of Massachusetts has a threshold effect exposure level of 0.2 ppbv (24 h average) and an annual average allowable ambient level of 0.03 ppbv (0.1 $\mu\text{g}/\text{m}^3$) for benzene. (30) Therefore, addressing NG-methane leakage for climate reasons may produce additional health co-benefits through this inhalation risk pathway.

Recently, there has been an acknowledgment of the increasing contribution of indoor emissions of certain VOCs from volatile chemical products impacting air quality (11,31) both as direct human health toxicants and indirectly as ozone precursors and precursors of SOA that make up a fraction of $\text{PM}_{2.5}$. Sourcing indoor emissions of VOCs largely to household chemical products (e.g., pesticides, coatings, adhesives, cleaning agents, and personal care products), McDonald et al. (11) notably did not consider indoor sources of VOCs coemitted with NG, noting that “NG is a clean-burning fuel, so combustion emissions of VOCs located at the point of use are small and negligible.” This assumption likely misclassified any VOC emissions associated with both fugitive emissions and combustion-related emissions from NG-fired appliances such as cook-top burners, ovens, and ventless heaters that are not automatically vented to the outdoors. To date, three studies (12,18,23) and one critical review (21) have quantified unburned NG emissions (CH_4) indoors, indicating that cooking appliances (stovetops and ovens) exhibit the highest emissions rates per unit fuel consumption compared to other appliances. (18) Steady-state-off emissions from cooking appliances have been shown to contribute disproportionately to total cooking appliance-use emissions, indicating the presence of a persistent leakage source of NG and associated VOC emissions. (23) Moreover, the long-tailed distribution of steady-state-off leakage rates observed by Lebel et al. (23) carries implications for understanding the range of potential VOC exposures from leaking NG appliances, particularly from cooking appliances that are likely centrally located within the home and are generally not required to be externally ventilated. There is also evidence showing the formation of gas-phase aromatics and polycyclic aromatic hydrocarbons from CH_4 combustion itself through multiple kinetic pathways that coincidentally are not removed by coformation of soot particles due to methane’s relatively clean burning nature. (32–34)

Odorant Detection Thresholds: Implications for Chronic CH_4 Leaks and HAP Exposure

From the Supporting Information Table S2, the most abundant TIC observed was the NG odorant TBM (97% detection) (μ = 579 ppbv; SD = 41; 95% CI 499–661). The other

commonly detected odorant, IPM, was also detected in 81% of samples ($\mu = 22.2$ ppbv; SD = 3; 95% CI 16.8–28.3). Notably, no differences were observed between gas companies for either TBM ($X^2 = 4.9$; $p = 0.2$) or IPM ($X^2 = 2.8$; $p = 0.09$); however, both TBM and IPM exhibited substantial variability with 3 and 2 orders of magnitude, respectively, yet were highly correlated (Pearson's $r = 0.94$, $p = <0.00001$). This variability was partly explained by seasonal variability with significantly lower odorant concentrations in the winter season for both TBM ($X^2 = 20.9$; $p = 0.007$) and IPM ($X^2 = 25.2$; $p = 0.001$). This variability is also partly explained by the fact that TBM and IPM were outside the target compound list as per the USEPA Method TO-15 and, therefore, are considered “NJ” flagged results that exhibit a high degree of uncertainty in terms of compound detection and concentrations. However, given the high frequencies of detection and the suitability to identify sulfurous compounds using fused silica-lined canisters, quantitative results are reported but should be viewed with extra caution. Furthermore, odorants are reported to be added to NG at concentrations ranging from 1–4 ppmv, (35,36) which agree with odorant data observations, particularly, after considering the likelihood of decreased odorant concentrations at the point of the end user due to some expected odor fade or other forms of degradation or removal. (37)

According to federal regulations (49 CFR 192.625), distribution-grade NG must contain odorants at a detectable concentration in air that is one-fifth of the lower explosive limit (LEL) for NG—that is, equal to detectable odor at $\sim 1\%$ NG in air by volume. While general human odor detection thresholds can differ by up to 1000-fold for the least and most sensitive population subgroups, a recently updated odor detection threshold for the NG odorant TBM was estimated at 6.26×10^{-3} ppbv (37,38) and is similar to the odor detection threshold for IPM. (39) Using these criteria, all samples that reported TBM ($n = 235$) met the 6.26×10^{-3} ppbv odor threshold (not including IPM or other thiol compounds) at one-fifth the LEL. The lowest TBM concentration at this level was 0.09 ppbv which also meets less conservative TBM odorization detection thresholds of 0.08 (39) or 0.029 ppbv. (40)

Relatedly, after scaling CH_4 content to 100% minus $\text{C}_2\text{H}_6\%$ in all samples, we found that 1% CH_4 —or approximately 1/5th the LEL—would coincide with a mean benzene concentration of 1.66 ppbv.

Using the sampled NG odorant content, we can also determine an NG leakage CH_4 concentration that coincides with the odor detection threshold—or the CH_4 level in air at which the odorant in NG cannot be detected by 50% of persons with a normal sense of smell. Given the similarity in odor detection thresholds for both TBM and IPM, we added the odorant concentrations together and used a 6.26×10^{-3} ppbv odorant detection threshold taken from the literature. (38–40) Using the 184 NG samples that were analyzed for CH_4 , TBM, and IPM, we conserved the mean molar ratios and found that the odor detection threshold of 6.26×10^{-3} ppbv coincided with a mean concentration of 21.3 ppmv CH_4 (95% CI: 16.7–25.9) and median of 8.8 ppmv (see the [Supporting Information](#) Section 2.10 for details). This implies that a steady-state ambient NG- CH_4 concentration of approximately 21.3 ppmv could persist undetected by persons with a normal sense of smell, and perhaps greater concentrations for those with anosmia (41) and those who may be unable to smell NG odorants either chronically and/or as a result of COVID-19 illness. (42) However, this NG-

associated CH₄ odor threshold requires additional verification considering the unreliability of these TIC odorant data and is likely an overestimate, assuming that sulfurous compounds in NG went undetected or were underestimated. Additionally, NG leak detection would be mediated by other factors as well, such as the location of a leak within a home, considering that the heavier sulfur-odorant compounds tend to sink versus CH₄, which is lighter than air. Nonetheless, Sargent et al. (22) recently found that an estimated $2.5 \pm 0.5\%$ of NG entering the Boston region is lost, noting that emissions are correlated with seasonal end use consumption implying that emissions may predominate from consumption-driven source types that includes beyond-the-meter leaks and residential end use appliances. The odorant content observed in end use NG in combination with known odor detection thresholds supports this hypothesis in the sense that small NG leaks may persist undetected and, therefore, may be more prevalent and not be immediately mitigated as has been previously assumed.

The odorant-threshold CH₄ concentration of 21.3 ppmv coincides with a mean benzene concentration of 0.004 ppbv—a concentration below MA’s annual average allowable ambient level of 0.03 ppbv. Assuming a steady-state ambient concentration and ignoring any chemical transformation or other sinks, a CH₄ concentration of 21.3 ppmv would require the benzene content in NG to exceed 1162 ppbv to reach MA’s allowable ambient level of 0.03 ppbv, which was not met in any samples herein (maximum whole NG benzene = 1080 ppbv). In their annual air quality reports, the Massachusetts Department of Environmental Protection (MassDEP) provides annual averages of 24 h outdoor ambient benzene concentrations measured at two monitoring sites within the greater Boston area (Boston, MA and Lynn, MA). The average annual 24-h ambient benzene during 2010–2020 ranged from 0.11 to 0.19 ppbv (Boston) and 0.09–0.13 ppbv (Lynn). Therefore, assuming similar indoor benzene levels, benzene coemitted with the 21.3 ppmv NG-CH₄ odorant threshold represents a 3% (95% CI: 2–4) enhancement over ambient with a 20% enhancement using the maximum observed whole NG benzene concentration of 1080 ppbv. Alternatively, in scenarios where NG can be detected by odor, such as at some gas utility worksites, it’s possible that NG concentrations would be high enough to produce a benzene concentration that exceeds MA’s AAL but likely not result in scenarios where OSHA’s 8 h time-weighted average limit of 1 ppmv or the 15 min short-term exposure limit of 5 ppmv for benzene is exceeded (29 CFR 1910.1028).

Characterizing Variability in NMVOC Content of NG with a Focus on BTEX Compounds

We also observed strong temporal trends in BTEX across the 18 month sampling campaign. Generally, BTEX content in NG peaked during the late winter season, with the highest overall concentrations observed spanning the 2020–2021 winter season (i.e., February to March 2021). As shown in [Figure 2](#), the variability of BTEX in NG also increased substantially in the winter months compared to other seasons. In aggregate, winter BTEX concentrations were 3-fold greater on average compared to the spring season and nearly 8-fold greater than the

summer season. The maximum BTEX concentrations were observed at nearly the same time of the year in the two successive heating seasons (late February). Notably, the wintertime BTEX peak also corresponded with regional low ambient air temperatures potentially corresponding with an increased NG demand which can alter NG source and supply practices (e.g., upstream underground storage withdraws, LNG-peak shaving). (9).

Figure 2

Figure 2. BTEX content of NG in raw (markers) and monthly averages (blue). Monthly averaged regional air temperature (°F) from the National Oceanic and Atmospheric Administration (NOAA) is displayed in red. The COVID-related sampling pause from mid-March 2020 through mid-July 2020 is highlighted in the grey box.

Consistent with the observation that greater CH₄ emissions coincide with NG usage during winter months, NMVOCs enhancement in NG observed during the winter months suggests that seasonality is an important effect modifier in considering potential associated health risks of leaked NG. For instance, lower indoor air exchange rates are generally more prevalent during the winter months, particularly for homes that rely on radiant heat, which could prolong exposure to any indoor NG emissions. (43,44) In regard to ambient air, photochemical activity (i.e., OH radical availability) is generally lower in the wintertime compared to summer, effectively increasing atmospheric lifetimes of certain NMVOCs such as BTEX. (45) Additionally, lower NG odorant content in the winter season was unexpected. One explanation could be that a higher odorant content is used during the warm season to counteract increased diffusivity of thiol compounds into pipe materials with increased temperatures; (46) however, it is ultimately unclear whether the variability in odorant content was intentional or a result of physiochemical processes.

Sample collection was designed to capture NG composition by the three largest local distribution companies in MA (LDCs, i.e., distribution utilities) to create a representative stratified sample (Figure 1). NG BTEX abundances varied by LDC and were elevated in Columbia's territory (utility A in Figure 1), as indicated by bootstrapped CIs (Table 2). All BTEX constituents were each significantly elevated in samples collected from Columbia's former territory compared to Eversource (utility B in Figure 1) and National Grid (utility C in Figure 1) (note—Eversource acquired all territories previously owned by Columbia Gas on October 13, 2020). Notably, Columbia's former service territory in MA entailed two geographically distinct territories—one in the Merrimack Valley (Lawrence, Andover, North Andover, MA—Figure 1), and the other encompassing a large swath south of Boston. Upon the finding of BTEX differences by territory, results were further differentiated by location and showed that elevated BTEX in the Merrimack Valley largely explained the elevated BTEX in Columbia's territory. For example, mean NG benzene concentrations were 439 ppbv in the Merrimack Valley compared to 102 ppbv in Sharon, MA—an area serviced within Columbia's former south Boston territory—and the differences were significant ($t = 4.08$; $p = 0.0006$).

Moreover, NG NMVOCs were significantly higher in the Merrimack Valley region compared to all other contiguous municipalities that were sampled. These results suggest that the geographic location better explains trace gas variability or is a better proxy for gas source and that service territory may have only a marginal or second-order effect.

Table 2. BTEX by Local Distribution Company Territory^a

	mean (95% CI) for BTEX constituents by LDC, ppbv			
	benzene	toluene	ethylbenzene	xylene
Columbia (utility A)	236 (171, 302)	249 (158, 349)	24.4 (14.8, 35.4)	146 (87.6, 213)
Eversource (utility B)	122 (78, 171)	98.5 (57.9, 147)	8.57 (5.08, 12.7)	50.3 (30.3, 74.0)
National Grid (utility C)	143 (109, 180)	123 (92.6, 157)	8.94 (6.03, 12.0)	54.3 (41.02, 69.5)
Difference (Columbia—Eversource)	114 (111, 116)	150 (147, 154)	15.8 (15.5, 16.2)	95.7 (93.8, 98.1)
Difference (Columbia—National Grid)	93.3 (90.8, 95.8)	126 (123, 129)	15.5 (15.1, 15.8)	91.7 (90.0, 94.1)
Difference (Eversource—National Grid)	-20.9 (-22.8, -19.0)	-24.1 (-25.9, -22.4)	-0.365 (-0.514, -0.215)	-4.00 (-4.76, -3.12)

^a

All statistics are derived from bootstrapped L3 data. Significant differences between LDCs are denoted by **italicized bold font**.

We next attempted to differentiate NG end use from its major transmission pipeline source; however, the complexity of the pipeline systems and lack of definitive pipeline flow data did not lend to clear source apportionment for individual samples. Both the Algonquin and Tennessee pipelines traverse the Greater Boston area from the west, which would coincide with estimates that, on average, 84% of the region's NG is generally sourced from the Marcellus formation. (47) A third transmission pipeline, the Maritimes & Northeast pipeline, traverses the Canadian Maritimes provinces and through Maine prior to reaching its terminus in the Merrimack Valley, which could indicate a distinct NG source into the area that could help explain the elevated NMVOCs observed in the Merrimack Valley. It is also worth noting that all sample collection in the Merrimack Valley occurred after the overpressurization event that took place on September 13, 2018. It is therefore unclear to what extent, if any, the subsequent replacement of ~77 km of underground pipelines had on results herein.

Provided that both substantial temporal and spatial variability were observed, we formally tested the differential variability of space (σ_{sp}^2) versus time (σ_t^2). σ_t^2 was calculated by pooling variances from each unique location having two or more samples across multiple dates. Similarly, σ_{sp}^2 was calculated by pooling variances from each sampling day having two or more samples across multiple locations. All BTEX constituents varied approximately twice as much across time than across space. In contrast, CH₄ varied approximately 3 times as much spatially versus temporally. However, while the CH₄ content of NG showed a higher spatial variance compared to temporal variance, this is not unexpected and is more likely an artifact of differential sample capture sufficiency rather than actual CH₄ variation across samples. See the [Supporting Information](#) Section 2.3 and [Supporting Information](#) Table S1 for details of this analysis.

While it was beyond the present scope to formally test root causes of seasonal and locational NMVOC variability in NG, possible sources of variation are likely some combination of (1) hydrocarbon sources via transmission pipeline, (2) natural reservoir variability, (3) NG processing steps and time in transit, and (4) utilization of supplemental hydrocarbon sources, such as upstream underground NG storage or LNG facilities. The observed ~1 month lag between temperature lows and peak BTEX could also signal an NG source change, whereby this latency period could represent a depletion of initial NG reserves and a switch to other NG sources (e.g., LNG-peak shaving). Increased C₄+ concentrations were observed by Liss et al., (9) where it was noted that LNG- and propane-peak shaving practices—deployed during peak demands during the heating season—had the greatest effect on the composition of NG delivered to customers. It is unclear to what degree peak shaving was utilized during sample collection herein; however, very low NG-propane content was observed, reducing the likelihood of any propane-peak shaving ($\mu = 579$ ppbv; see [Table S2](#)).

Overall, it is difficult to infer the generalizability of data collected in the Greater Boston area to areas outside of this. However, considering that most of the Northeastern U.S. sources NG in a similar manner, data collected herein are likely representative of the major cities along the northeast. We also note that benzene and hexane were highly correlated within our data set with an $R^2 = 0.80$ ($p < 0.0001$). Considering that publicly available midstream NG postings typically report NG hexane abundance, it is noteworthy these data appear to provide a reasonable approximation of the NG benzene content. From this study, the abundance ratio of benzene/hexane was 0.28 (95% CI: 0.26–0.30, RMA regression) (see the [Supporting Information](#) Section 2.4), which could be used as a scaling factor to infer benzene concentrations from hexane concentrations, assuming these relationships are robust outside of the data collected herein.

Using BTEX concentrations from this and other NG composition studies, it is also possible to calculate source-dependent ratios of individual constituents relative to benzene. While the magnitude of individual BTEX constituents can vary significantly across individual samples, the relative abundances of toluene, ethylbenzene, and xylenes to benzene collectively across all samples were relatively consistent ([Table 3](#)). In general, NG is benzene-dominant (or on par with toluene) relative to other anthropogenic sources of BTEX, such as gasoline combustion, which tends to be toluene-dominant, exhibiting a T/B ratio > 2 . (48) While it is

unclear whether whole NG NMVOCs measured in Boston are representative of other cities or upstream portions of its supply chain, NG BTEX abundance ratios in [Table 3](#) indicate that the geologic source likely exhibits a greater NG VOC variability than the variability between midstream and downstream portions of the supply chain. For example, [\(49\)](#) the distinct differences found in T/B molar ratios between SW PA and NE PA drilling regions possibly indicate distinct hydrocarbon makeup in the Marcellus region, PA alone. Similarly, Marrero et al. [\(26\)](#) found clear differences in NMVOC content measured downwind of various portions of the NG supply chain in the Barnett region of Texas. Nonetheless, given the difficulty in distinguishing BTEX sources in urban areas, [\(50\)](#) these data provide a distribution-grade NG source signature for comparison to other sources of VOCs and can be used in developing emission inventory estimates for fugitive NG from distribution- and beyond-the-meter source types. [Figure S9](#) provides BTEX RMA regressions used in calculating the relative abundances and their uncertainties from end user NG samples.

Table 3. Relative Abundances of Previously Published NG-Sourced BTEX Compared to the Present Study

	T/B (95% CI)	E/B (95% CI)	X/B (95% CI)
downstream—this study ^a	1.1 (1.0,1.2)	0.11 (0.10, 0.12)	0.59 (0.53, 0.65)
downstream—distribution-grade NG (<i>n</i> = 2) ^b	1.1 (NA)	0.10 (NA)	0.60 (NA)
midstream—NG (TX) ^c	1.29 (NA)	0.136 (NA)	1.28 (NA)
upstream—NG wells (WY) ^d	0.94 (0.73, 1.2)	0.13 (0.060, 0.26)	0.51 (0.34, 0.73)
upstream—NG (SW PA) ^e	1.45 (NA)	NA	NA
upstream—NG (NE PA) ^e	0.77 (NA)	NA	NA

^a

Ratio and 95% CI calculated from RMA regression of BTEX data at end user points.

^b

Burger et al., [\(3\)](#) ratios estimated through published chromatographs of distribution-level NG samples; therefore, no uncertainty estimates are available.

^c

Gulf South Pipeline Company LLC, [\(51\)](#) ratios of overall median values across seven separate interconnected pipelines near Houston, TX, reporting 5-minute data from Dec 1, 2020, through July 1, 2021.

^d

DiGiulio and Jackson, [\(52\)](#) ratio and 95% CI calculated from RMA regression of BTEX data scraped from a pdf table of Bradenhead well-head data.

^e

Goetz et al. (49) molar ratios for southwest (SW) and northeast (NE) PA via mobile monitoring in proximity of shale gas activity throughout the Marcellus, PA.

Inferred NG-Sourced BTEX Emissions for Greater Boston

Annual NG-sourced BTEX emissions were estimated for the Greater Boston area by combining our whole NG sample results with regional NG CH₄ emissions fluxes estimated by McKain et al., (17) Plant et al., (20) and Sargent et al. (22) (Table 4). Downstream NG system loss estimates indicate that NG-sourced BTEX emissions contribute to a modest NMVOC enhancement that is currently not accounted for in emissions inventories. Using NG CH₄ leak estimates from McKain et al., (17) Plant et al., (20) and Sargent et al., (22) an estimated 338–608 kg/yr (745–1340 lbs/yr) of total BTEX is annually emitted alongside CH₄ leakage throughout Greater Boston. For context, 216 kg/yr (476 lbs/yr) of benzene emitted annually (estimated via NG methane emissions from (17) equates to nearly 10% of on-road diesel light-duty vehicles for all of Massachusetts). (4) Of the 35 benzene source sectors tracked by the USEPA’s 2017 NEI, benzene emissions from NG leakage alone would rank 25th in MA. Depending on the NG CH₄ leak estimate used, our work suggests that benzene emissions from NG leaks are 11–31% of the median benzene sources tracked by the USEPA’s 2017 NEI. More specifically, annual NG benzene emissions associated with NG leakage (120–327 kg/yr) are likely greater than benzene emissions in MA from NG-combustion associated with industrial (129 kg/yr), residential (121 kg/yr), and commercial/institutional sectors (119 kg/yr). While VOC emissions from leaking NG are not currently accounted for in inventories and do not appear to be a major contributing source of VOCs, it is important to note that an uncertain fraction of these emissions occurs indoors and, therefore, may be subject to a relatively high human exposure-based intake fraction.

Table 4. Annual BTEX Emissions for the Entire 18,000 km² Area in McKain et al. (17) and the Approximately Similar Boston Bounds Used by Plant et al. (20) and Sargent et al (22)

reference	Boston region NG CH ₄ leaks (Tg/yr)	BTEX (kg/yr)* based on the Boston region NG leak estimates			
	CH ₄ (95%CI)	benzene (95% CI)	toluene (95% CI)	ethylbenzene (95% CI)	xylenes (95% CI)
McKain et al. (17) 2015a	0.33 (0.27, 0.4)	254 (174, 356)	272 (179, 398)	25.3 (15.8, 38.1)	153 (98.9, 223)
Plant et al. (20) 2019a	0.16 (0.12, 0.2)	120 (75.3, 175)	132 (81.6, 197)	12.1 (7.07, 18.9)	73.6 (43.5, 112)
Sargent et al. (22) 2021b	0.17 (0.14, 0.21)	132 (90.4, 186)	141 (92.9, 204)	13.4 (8.33, 20.2)	80.0 (51.0, 119)

18,000 km².

^b

12,351 km².

Conclusions

Overall, we conducted a first-of-its-kind trace-gas characterization of unburned NG collected directly from indoor residential kitchen stovetops and outdoor building NG service lines throughout Greater Boston, Massachusetts. Trace gas analyses found that distribution-grade NG contains numerous NMVOCs, including HAPs, such as benzene, that were enhanced during the winter months. We also observed distinct trace gas variability by location but were unable to delineate root causes. Full implications extend beyond the need for additional research in other service territories to include several actions that would improve a general understanding of potential health risks posed by NMVOCs in distribution-grade NG. First, pipeline operators and utilities have a ready access to their own product and could regularly measure and report differentiated C6+ NMVOC content in NG postings; current reporting of NG constituents varies significantly between operators, even for the two major pipelines supplying Greater Boston. (53,54) Relatedly, NG odorization practices could be made publicly available in informational postings to better understand end use odorant content and its implications for low-level leaks behind-the-meter. Also, while more data is needed relating to behind-the-meter NG leakage, home inspectors and contractors serving in that capacity could easily perform NG-appliance leak detection surveys with measurement devices that detect in the low ppmv range, similar to radon tests done prior to the completion of a real estate transaction. Ultimately, we categorize this study as a hazard identification study within the larger health risk assessment context.

Future studies should better determine HAP source–concentration relationships from both pre- and post-combustion with considerations of microenvironments with either a high NG use (e.g., utility worksites, commercial kitchens) or built environments with poor indoor ventilation. Finally, alongside known NG leakage pathways, these data represent a source of VOCs in indoor and urban areas that is not currently accounted for in emissions inventories and provide a previously unaccounted for public health cobenefit associated with reductions of NG-methane emissions.

Home is Where the Pipeline Ends: Characterization of Volatile Organic Compounds Present in Natural Gas at the Point of the Residential End User

22 views

0 shares

0 downloads

Supplementary Information to Accompany:

Home is where the pipeline ends: Characterization of volatile organic compounds present in natural gas at the point of the residential end-user

Drew R. Michanowicz^{1,2,§,*}, Archana Dayalu^{3,§}, Curtis L. Nordgaard², Jonathan J. Buonocore¹, Molly W. Fairchild⁴, Robert Ackley⁵, Jessica E. Schiff⁶, Abbie Liu⁶, Nathan G. Phillips⁷, Audrey Schulman⁴, Zeyneb Magavi⁴, John D. Spengler⁶

¹Harvard T.H. Chan School of Public Health, C-CHANGE, Boston, Massachusetts, 02215 United States

²PSE Healthy Energy, Oakland, California, 94612 United States

³Atmospheric and Environmental Research (AER), Lexington, Massachusetts, 02421 United States

⁴Home Energy Efficiency Team (HEET), Cambridge, Massachusetts, 02139 United States

⁵Gas Safety Inc., Southborough, Massachusetts, 01772 United States

⁶Harvard T.H. Chan School of Public Health, Boston, Massachusetts, 02215 United States

⁷Boston University, Boston, Massachusetts, 02215 United States

*Corresponding author, email: Drew@PSEHealthyEnergy.org / Michanow@HSPH.Harvard.edu

§Contributed equally

Table of Contents

1. Methods	S2
1.1 Sample collection and processing	S2
1.2 Data processing	S4
2. Results	S10
2.1 Ethane to methane ratio	S10
2.2 Sensitivity tests	S11
2.3 Sample spatial and temporal variability	S11
2.4 Co-variance of benzene and hexane in natural gas	S12
2.5 Co-variance of BTEX components in natural gas	S13
2.6 Comparison of winter BTEX sample profiles: pre-COVID and COVID sample phases	S13
2.7 Comparison of BTEX across seasons	S15
2.8 Natural gas components and range of composition: This study and others	S16
2.9 NG Methane emissions and estimated corresponding BTEX fluxes: domain variations	S18
3. References	S21

Supplementary Information to Accompany

:

Home is where the pipeline ends: Characterization of volatile
organic compounds present in natural gas at the point of the
residential end

-

user

Drew R.

Michanowicz

1,2 ,

§,*

, Archana Dayalu

3

,§

, Curtis

L.

Nordgaard

2

, Jonathan J.

Buonocore

1

, Molly W. Fairchild

4

, Robert

Ackley

5

,

Jessica E. Schiff

6

, Abbie Liu

6

, Nathan

G.

Phillips

7

,

Audrey

Schulman

4

,

Zeyneb

Magavi

4

, John D. Spengler

6

1

Harvard T.H. Chan School of Public Health, C

-

CHANGE,

Boston, Massachusetts,

02215

United States

2

PSE Healthy Energy, Oakland, California,

94612

United States

3

Atmospheric and Environmental Research (AER), Lexington, Massachusetts,

02421

United

States

4

Home Energy Efficiency Team (HEET), Cambridge, Massachusetts,

02139

United

States

5

Gas Safety Inc., Southborough, Massachusetts,

01772

United States

6

Harvard T.H. Chan School of Public Health, Boston, Massachusetts,

02215

United States

7

Boston University, Boston, Massachusetts,

02215

United States

*Corresponding author, email:

Drew@PSEHealthyEnergy.org

/ Michanow@HSPH.Harvard.edu

§

Contributed equally

Table of Contents

1.

Methods

.....

.....

.....

.....

.....

S

2

1.1 Sample

c

ollection and

p

rocessing

.....

.....

.....

.....

.....

S

2

1.2

Data

p

rocessing

.....

.....

.....

.....

.....

.....

S

4

2.

Results

.....

.....

.....

.....

.....

S

10

2.1 Ethane to

m

ethane

r

atio

.....

.....

.....

.....

.....

S

10

2.2 Sensitivity

t

ests

.....

.....

.....

.....

.....

.....

S

11

2.3 Sample

s

patial and

t

emporal

v

ariability

.....

.....

.....

.....

S

11

2.4 Co

-

variance of benzene and hexane in

n

atural

g

as

.....

.....

.....

...

S

12

2.5 Co

-

variance of BTEX components i

n

n

atural

g

as

.....

.....

.....

.....

S

13

2.6 Comparison of

w

inter BTEX sample profiles: pre

-

COVID and COVID sample phases

.....

S

13

2.7 Comparison of BTEX across seasons

.....

.....

.....

.....

S

15

2.8 Natural

g

as

c

omponents and

r

ange of

c

omposition: This study and others

.....

.....

S

16

2.9 NG Methane emissions and estimated corresponding BTEX fluxes: domain variations

.....

S

18

3.

References

.....

.....

.....

.....

S

21

1. Methods

1.1 Sample collection and processing

Whole-NG samples were collected from indoor NG-fired stovetop appliances and outdoor gas service lines across three local natural gas distribution territories. Compounds of interest included over 70 unique VOCs collected via grab samples in evacuated 1.4L Entech Silonite-lined canisters prepared by commercial environmental testing labs - Phoenix Environmental Laboratories (Phoenix Environmental Laboratories, Manchester, CT) and tested according to United States Environmental Protection Agency (USEPA) method TO-15. VOCs were subsequently separated by gas chromatography and measured by mass-selective detector techniques. Tentatively identified compounds (TICs) were also reported per sample. TICs were reported based on an evaluation of non-target compounds that had a peak area count > 10% of the nearest internal standard and met all mass spectral criteria. To further verify sufficient sample capture of NG, an aliquot from a subset of samples was tested for CH₄ and C₂H₆ using EPA method 3C performed by New England Testing Laboratory, West Warwick, RI (managed internally by Phoenix Environmental Laboratories). Analytical methods for ASTM D1945, ASTM D1946, and EPA 3C are identical, though differ by QC criteria and compounds specified for each method.

Stovetop sample collection entailed a direct in-line connection between stove natural gas outlet and sample canister via flexible Teflon-lined tubing (Figure S1). For all stoves sampled, the tubing diameter fully encompassed the gas outlet orifice (Figure S1, panel 4). The in-line connection effectively bypassed the stovetop ignition source (where present) and ensured that the sample was not diluted by ambient air and that minimal unburned NG was released. Once a secure connection was made, the gas flow knob was turned to medium-high. In most instances, the tubing was held in place by the second researcher. Once NG flow was detected from the open end via smell, the tubing was attached to an Entech Micro QT flow valve using a screw nut and cone washer (Figure S1, panel 7). The Micro QT valve was then engaged to the canister for sample collection that typically required 30 seconds for sample collection. Sample collection was completed once the canister stopped producing a hissing sound. Stovetop models with known automatic shut-off devices or aeration devices were excluded at the outset, though not all could be identified as evidenced post-hoc by very low canister CH₄ concentrations.

Building NG service line sampling was identical to stovetop sampling, except samples were collected from service lines that were typically connected to outdoor NG grills or firepits. Typically, building NG service line sample collection entailed disconnecting an existing grill hose at the service line riser and connecting a new hose (3/8 inch or 1/2 inch Dozyant) via an auto-coupler quick connect. An in-line sample collection flow devoid of ambient intrusion was created by inserting the Teflon-lined tubing at least two feet into the new grill hose. Once inserted and NG flow was detected, the line was purged for 10 seconds, and the sample collection proceeded similar to stovetop sample collection. Canisters were sampled within five days of arrival and returned within three days of sample collection. All lab analyses were completed within three weeks of sampling and chain of custody forms were maintained for all samples.

1

.

Methods

1.

1 Sample

c

ollection and

p

rocessing

Whole

-

NG samples were collected from indoor NG

-

fired stovetop appliances and outdoor gas

service

lines across three local natural gas distribution territories. Compounds of interest included

over 70 unique VOCs collected via grab samples in evacuated 1.4L Entech Silonite

-

lined

canisters

prepared by commercial environmental testing labs

-

Phoenix Environmental Laboratories

(Phoenix Environmental Laboratories, Manchester, CT) and tested according to United States

Environmental Protection Agency (USEPA) method TO

-

15. VOCs were subsequently separated

by gas chromatography and measured by mass

-

selective detector techniques. Tentatively identified

compounds (TICs) were also reported per sample. TICs were reported based on an evaluation of

non

-

target compounds that had a peak area count > 10% of the nearest internal standard and met all mass spect

ral criteria. To further verify sufficient sample capture of NG, an aliquot from a subset of samples was tested for CH

4

and C

2

H

6

using EPA method 3C performed by New England

Testing Laboratory, West Warwick, RI (managed internally by Phoenix Environmental

Laboratories). Analytical methods for ASTM D1945, ASTM D1946, and EPA 3C are identical, though differ by QC criteria and compounds specified for each method.

Stovetop sample collection entailed a direct in

-

line connection between stove natural gas outlet
and sample canister via flexible Teflon

-

lined tubing

(Figure S1

)

. For all stoves sampled, the tubing
diameter fully encompassed the gas outlet orifice

(Figure S1

, panel 4)

. The in

-

line connection

effectively bypassed the stovetop ignition source (where present) and ensured that the sample was

not diluted by ambient air

and that minimal

unburned NG was released.

Once a secure connection

was made,

the gas flow knob was turned to medium

-

high.

In

most instances

, the tubing was held
in place by the
second researcher.

Once NG flow was detected from the open end
via smell

, the

tubing was attached to an Entech

Micro QT flow valve using a screw nut and cone washer

(Figure

S1, panel 7

)

. The Micro QT valve was then engaged to the canister for sample collection that
typically required

30 seconds

f

or sample collection

.

Sample collection was completed once the

cannister stopped producing a hissing sound.

Stovetop models with known automatic shut

-

off

devices or aeration devices were ex

cluded at the outset, though not all could be identified as

evidenced post

-

hoc by very low canister CH

4

concentrations.

Building NG service line sampling was identical to stovetop sampling, except samples were collected from service lines that were typical

ly connected to outdoor NG grills or firepits.

Typically, building NG service line sample collection entailed disconnecting an existing grill hose

at the service line riser and connecting a new hose ($\frac{3}{8}$ inch or $\frac{1}{2}$ inch Dozyant) via an auto

-

coupler

quick conn

ect. An in

-

line sample collection flow devoid of ambient intrusion was created by inserting the Teflon

-

lined tubing at least two feet into the new grill hose. Once inserted and NG flow was detected, the line was purged for 10 seconds,

and the sample collect

ion proceeded similar

to stovetop sample collection. Canisters were sampled within five days of arrival and returned


within three days of sample collection. All lab analyses were completed within three
three
weeks of
sampling and chain of custody forms were maintained for all samples.



Terms & Conditions

Most electronic Supporting Information files are available without a subscription to ACS Web Editions. Such files may be downloaded by article for research use (if there is a public use license linked to the relevant article, that license may permit other uses). Permission may be obtained from ACS for other uses through requests via the RightsLink permission system: <http://pubs.acs.org/page/copyright/permissions.html>.

- Corresponding Author

Drew R. Michanowicz - Harvard T.H. Chan School of Public Health, C-CHANGE, Boston, Massachusetts 02215 United States; PSE Healthy Energy, Oakland, California 94612, United States;  Orcid <https://orcid.org/0000-0002-2538-4819>; Email: Drew@PSEHealthyEnergy.org Michanow@HSPH.Harvard.edu

- Authors
 - Archana Dayalu - Atmospheric and Environmental Research (AER), Lexington, Massachusetts 02421, United States
 - Curtis L. Nordgaard - PSE Healthy Energy, Oakland, California 94612, United States
 - Jonathan J. Buonocore - Harvard T.H. Chan School of Public Health, C-CHANGE, Boston, Massachusetts 02215 United States
 - Molly W. Fairchild - Home Energy Efficiency Team (HEET), Cambridge, Massachusetts 02139, United States
 - Robert Ackley - Gas Safety Inc., Southborough, Massachusetts 01772, United States
 - Jessica E. Schiff - Harvard T.H. Chan School of Public Health, Boston, Massachusetts 02215, United States
 - Abbie Liu - Harvard T.H. Chan School of Public Health, Boston, Massachusetts 02215, United States
 - Nathan G. Phillips - Boston University, Boston, Massachusetts 02215, United States
 - Audrey Schulman - Home Energy Efficiency Team (HEET), Cambridge, Massachusetts 02139, United States
 - Zeyneb Magavi - Home Energy Efficiency Team (HEET), Cambridge, Massachusetts 02139, United States
 - John D. Spengler - Harvard T.H. Chan School of Public Health, Boston, Massachusetts 02215, United States
- Author Contributions

D.R.M. and A.D. contributed equally.
- Notes

The authors declare no competing financial interest.

This work was supported by grants from the Barr Foundation (grant #19-07957) and Putnam Foundation. A special thank you to all the study participants who welcomed us into their homes, particularly during uncertain times. Thank you to Jose Vallarino for help in designing the sampling protocols and hours of troubleshooting. Thank you to Megan Jones, Wendy Wang, and Sara Gillooly for the field sampling activities. Thank you to Marty Alvarez and

Gary Adamkiewicz for IRB guidance. The authors thank the editor and peer reviewers for their suggestions and contributions. Finally, thank you to Regina LaRocque, Barbara Taggart, Leanne Westfall, and Andee Krasner for their insights and early contributions.

This article references 54 other publications.


1. **1**

Energy Information Association. U.S. energy consumption by source and sector. https://www.eia.gov/totalenergy/data/monthly/pdf/flow/total_energy_2020.pdf (accessed 10/28/21). 2020.
[Google Scholar](#)

2. **2**

Lyons, W. C.; Plisga, G. J. *Standard Handbook of Petroleum and Natural Gas Engineering*; Elsevier, 2011.
[Google Scholar](#)


3. **3**

Burger, J. L.; Lovestead, T. M.; Bruno, T. J. Composition of the C6+ fraction of natural gas by multiple porous layer open tubular capillaries maintained at low temperatures. *Energy Fuels* 2016, 30, 2119– 2126, DOI: 10.1021/acs.energyfuels.6b00043
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)

4. **4**

USEPA. National Emissions Inventory 2017 Data. <https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei2020> (accessed 10/28/21).
[Google Scholar](#)

5. **5**

Francoeur, C. B.; McDonald, B. C.; Gilman, J. B.; Zarzana, K. J.; Dix, B.; Brown, S. S. Quantifying Methane and Ozone Precursor Emissions from Oil and Gas Production Regions across the Contiguous US. *Environ. Sci. Technol.* 2021, 55, 9129– 9139, DOI: 10.1021/acs.est.0c07352
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)

6. **6**

Pétron, G.; Karion, A.; Sweeney, C.; Miller, B. R.; Montzka, S. A.; Frost, G. J. A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin. *J. Geophys. Res. Atmos.* 2014, 119, 6836– 6852, DOI: 10.1002/2013jd021272
[Crossref], [CAS], [Google Scholar](#)

7. **7**

USEPA. Oil and natural gas sector: Emissions standards for new, reconstructed, and modified sources review. [EPA-HQ-OAR-2017-0757; FRL-10013-44-OAR].

https://www.epa.gov/sites/default/files/2020-08/documents/frn_oil_and_gas_review_2060-at90_final_20200812_admin_web.pdf (accessed 10/7/21), 2020.

[Google Scholar](#)

8. **8**

Field, R. A.; Goldstone, M. E.; Lester, J. N.; Perry, R. The variation of volatile organic compound concentrations in central London during the period July 1991 to September 1992. *Environ. Technol.* 1994, *15*, 931– 944, DOI: 10.1080/09593339409385501

[[Crossref](#)], [[CAS](#)], [Google Scholar](#)

9. **9**

Liss, W.; Thrasher, W.; Steinmetz, G.; Chowdiah, P.; Attari, A. *Variability of Natural Gas Composition in Select Major Metropolitan Areas of the United States*; American Gas Association Labs.: Cleveland, OH (United States), 1992, final report, august 1990 February 1992.

[Google Scholar](#)

10. **10**

Coggon, M. M.; Gkatzelis, G. I.; McDonald, B. C.; Gilman, J. B.; Schwantes, R. H.; Abuhassan, N. Volatile chemical product emissions enhance ozone and modulate urban chemistry. *Proc. Natl. Acad. Sci. U.S.A.* 2021, *118*, e2026653118 DOI:

10.1073/pnas.2026653118

[[Crossref](#)], [[PubMed](#)], [[CAS](#)], [Google Scholar](#)

11. **11**

McDonald, B. C.; De Gouw, J. A.; Gilman, J. B.; Jathar, S. H.; Akherati, A.; Cappa, C. D. Volatile chemical products emerging as largest petrochemical source of urban organic emissions. *Science* 2018, *359*, 760– 764, DOI: 10.1126/science.aaq0524

[[Crossref](#)], [[PubMed](#)], [[CAS](#)], [Google Scholar](#)

12. **12**

Fischer, M. L.; Chan, W. R.; Delp, W.; Jeong, S.; Rapp, V.; Zhu, Z. An estimate of natural gas methane emissions from California homes. *Environ. Sci. Technol.* 2018, *52*, 10205– 10213, DOI: 10.1021/acs.est.8b03217





[[ACS Full Text](#)  [ACS Full Text](#)], [[CAS](#)], [Google Scholar](#)

13. **13**

Hendrick, M. F.; Ackley, R.; Sanaie-Movahed, B.; Tang, X.; Phillips, N. G. Fugitive methane emissions from leak-prone natural gas distribution infrastructure in urban environments. *Environ. Pollut.* 2016, *213*, 710– 716, DOI:

10.1016/j.envpol.2016.01.094


[[Crossref](#)], [[PubMed](#)], [[CAS](#)], [Google Scholar](#)

14. **14**
Hopkins, F. M.; Kort, E. A.; Bush, S. E.; Ehleringer, J. R.; Lai, C.-T.; Blake, D. R. Spatial patterns and source attribution of urban methane in the Los Angeles Basin. *J. Geophys. Res. Atmos.* 2016, *121*, 2490– 2507, DOI: 10.1002/2015jd024429
[Crossref], [CAS], [Google Scholar](#)
15. **15**
Huang, Y.; Kort, E. A.; Gourdji, S.; Karion, A.; Mueller, K.; Ware, J. Seasonally resolved excess urban methane emissions from the Baltimore/Washington, DC metropolitan region. *Environ. Sci. Technol.* 2019, *53*, 11285– 11293, DOI: 10.1021/acs.est.9b02782
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)
16. **16**
Lebel, E. D.; Lu, H. S.; Speizer, S. A.; Finnegan, C. J.; Jackson, R. B. Quantifying methane emissions from natural gas water heaters. *Environ. Sci. Technol.* 2020, *54*, 5737– 5745, DOI: 10.1021/acs.est.9b07189
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)
17. **17**
McKain, K.; Down, A.; Raciti, S. M.; Budney, J.; Hutyra, L. R.; Floerchinger, C. Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proc. Natl. Acad. Sci. U.S.A.* 2015, *112*, 1941– 1946, DOI: 10.1073/pnas.1416261112
[Crossref], [PubMed], [CAS], [Google Scholar](#)
18. **18**
Merrin, Z.; Francisco, P. W. Unburned methane emissions from residential natural gas appliances. *Environ. Sci. Technol.* 2019, *53*, 5473– 5482, DOI: 10.1021/acs.est.8b05323
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)
19. **19**
Phillips, N. G.; Ackley, R.; Crosson, E. R.; Down, A.; Hutyra, L. R.; Brondfield, M. Mapping urban pipeline leaks: Methane leaks across Boston. *Environ. Pollut.* 2013, *173*, 1– 4, DOI: 10.1016/j.envpol.2012.11.003
[Crossref], [PubMed], [CAS], [Google Scholar](#)
20. **20**
Plant, G.; Kort, E. A.; Floerchinger, C.; Gvakharia, A.; Vimont, I.; Sweeney, C. Large fugitive methane emissions from urban centers along the US East Coast. *Geophys. Res. Lett.* 2019, *46*, 8500– 8507, DOI: 10.1029/2019gl082635
[Crossref], [PubMed], [CAS], [Google Scholar](#)
21. **21**
Saint-Vincent, P. M. B.; Pekney, N. J. Beyond-the-meter: Unaccounted sources of methane emissions in the natural gas distribution sector. *Environ. Sci. Technol.* 2019, *54*, 39– 49, DOI: 10.1021/acs.est.9b04657
[ACS Full Text  ACS Full Text], [Google Scholar](#)

22. **22**

Sargent, M. R.; Floerchinger, C.; McKain, K.; Budney, J.; Gottlieb, E. W.; Hutyra, L. R. Majority of US urban natural gas emissions unaccounted for in inventories. *Proc. Natl. Acad. Sci. U.S.A.* 2021, *118*, e2105804118 DOI: 10.1073/pnas.2105804118
[Crossref], [PubMed], [CAS], [Google Scholar](#)


23. **23**

Lebel, E. D.; Finnegan, C. J.; Ouyang, Z.; Jackson, R. B. *Methane and NO_x Emissions from Natural Gas Stoves, Cooktops, and Ovens in Residential Homes*; Environmental Science & Technology, 2022.
[ACS Full Text  ACS Full Text], [Google Scholar](#)


24. **24**

USEPA Office of Superfund Remediation and Technology Innovation. *National Functional Guidelines for Superfund Organic Methods Data Review* , 2015.
Washington D.C. OSWER 9355.0-132.
[Google Scholar](#)

25. **25**

Young, B. R.; Sleeth, D. K.; Handy, R. G.; Pahler, L. F. The Recovery of Volatile Organic Compounds and Volatile Sulfur Compounds in Fused-Silica Lined Canisters, Polyvinyl Fluoride/Tedlar Bags, and Foil-Lined Bags. *ACS Chem. Health Saf.* 2021, *28*, 426– 435, DOI: 10.1021/acs.chas.1c00027
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)


26. **26**


Marrero, J. E.; Townsend-Small, A.; Lyon, D. R.; Tsai, T. R.; Meinardi, S.; Blake, D. R. Estimating emissions of toxic hydrocarbons from natural gas production sites in the Barnett Shale region of Northern Texas. *Environ. Sci. Technol.* 2016, *50*, 10756– 10764, DOI: 10.1021/acs.est.6b02827
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)

27. **27**

Deighton, J. A.; Townsend-Small, A.; Sturmer, S. J.; Hoschouer, J.; Heldman, L. Measurements show that marginal wells are a disproportionate source of methane relative to production. *J. Air Waste Manage. Assoc.* 2020, *70*, 1030– 1042, DOI: 10.1080/10962247.2020.1808115
[Crossref], [CAS], [Google Scholar](#)

28. **28**

Gilman, J. B.; Lerner, B. M.; Kuster, W. C.; De Gouw, J. A. Source signature of volatile organic compounds from oil and natural gas operations in northeastern Colorado. *Environ. Sci. Technol.* 2013, *47*, 1297– 1305, DOI: 10.1021/es304119a
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)

29. **29**
Broderick, B. M.; Marnane, I. S. A comparison of the C2–C9 hydrocarbon compositions of vehicle fuels and urban air in Dublin, Ireland. *Atmos. Environ.* 2002, 36, 975– 986, DOI: 10.1016/s1352-2310(01)00472-1
[Crossref], [CAS], [Google Scholar](#)
30. **30**
Massachusetts DEP. Ambient air toxics guidelines, 2021.
<https://www.mass.gov/service-details/massdep-ambient-air-toxics-guidelines>. 2021 (accessed 11/30/21).
[Google Scholar](#)
31. **31**
Gkatzelis, G. I.; Coggon, M. M.; McDonald, B. C.; Peischl, J.; Gilman, J. B.; Aikin, K. C. Observations confirm that volatile chemical products are a major source of petrochemical emissions in US cities. *Environ. Sci. Technol.* 2021, 55, 4332– 4343, DOI: 10.1021/acs.est.0c05471
[ACS Full Text  ACS Full Text], [CAS], [Google Scholar](#)
32. **32**
D’anna, A.; Kent, J. H. A model of particulate and species formation applied to laminar, nonpremixed flames for three aliphatic-hydrocarbon fuels. *Combust. Flame* 2008, 152, 573– 587, DOI: 10.1016/j.combustflame.2007.08.003
[Crossref], [CAS], [Google Scholar](#)
33. **33**
Senkan, S.; Castaldi, M. Formation of polycyclic aromatic hydrocarbons (PAH) in methane combustion: comparative new results from premixed flames. *Combust. Flame* 1996, 107, 141– 150, DOI: 10.1016/0010-2180(96)00044-2
[Crossref], [CAS], [Google Scholar](#)
34. **34**
Vovelle, C.; Delfau, J.-L.; Pillier, L. Laminar hydrocarbon flame structure. *Combust. Explos. Shock Waves* 2009, 45, 365– 382, DOI: 10.1007/s10573-009-0047-z
[Crossref], [Google Scholar](#)
35. **35**
Speight, J. G. *Natural Gas: A Basic Handbook*; Gulf Professional Publishing, 2018.
36. **36**
Tenkrat, D.; Prokes, O.; Hlincik, T. *Natural Gas Odorization*; INTECH Open Access Publisher, 2010.
37. **37**
Esfandiari, K.; Banihashemi, M.; Mokhtari, A.; Soleimani, P. Experimental investigation of influencing parameters on natural gas odor fading in gas distribution networks. *J. Nat. Gas Sci. Eng.* 2021, 95, 104191, DOI: 10.1016/j.jngse.2021.104191
[Crossref], [CAS], [Google Scholar](#)

38. **38**
Wise, P.; Rowe, S.; Dalton, P. Odorization of Natural Gas: What are the Challenges?. *bioRxiv* 2021, DOI: 10.1101/2021.07.10.450231
39. **39**
Clanton, C. J.; Schmidt, D. R. Sulfur compounds in gases emitted from stored manure. *Trans. ASAE* 2000, *43*, 1229, DOI: 10.13031/2013.3016
[\[Crossref\]](#), [\[CAS\]](#), [Google Scholar](#)
40. **40**
Nagata, Y.; Takeuchi, N. Measurement of odor threshold by triangle odor bag method. *Odor Meas. Rev.* 2003, *118*, 118– 127
[Google Scholar](#)
41. **41**
Dong, J.; Pinto, J. M.; Guo, X.; Alonso, A.; Tranah, G.; Cauley, J. A. The prevalence of anosmia and associated factors among US black and white older adults. *J. Gerontol. A Biol. Sci. Med. Sci.* 2017, *72*, 1080– 1086, DOI: 10.1093/gerona/glx081
[\[Crossref\]](#), [\[PubMed\]](#), [\[CAS\]](#), [Google Scholar](#)
42. **42**
Khan, A. M.; Kallogjeri, D.; Piccirillo, J. F. Growing Public Health Concern of COVID-19 Chronic Olfactory Dysfunction. *JAMA Otolaryngol. Head Neck. Surg.* 2022, *148*, 81, DOI: 10.1001/jamaoto.2021.3379
[\[Crossref\]](#), [\[PubMed\]](#), [\[CAS\]](#), [Google Scholar](#)
43. **43**
Deng, S.; Lau, J. Seasonal variations of indoor air quality and thermal conditions and their correlations in 220 classrooms in the Midwestern United States. *Build. Environ.* 2019, *157*, 79– 88, DOI: 10.1016/j.buildenv.2019.04.038
44. **44**
Underhill, L. J.; Dols, W. S.; Lee, S. K.; Fabian, M. P.; Levy, J. I. Quantifying the impact of housing interventions on indoor air quality and energy consumption using coupled simulation models. *J. Expo. Sci. Environ. Epidemiol.* 2020, *30*, 436– 447, DOI: 10.1038/s41370-019-0197-3
[\[Crossref\]](#), [\[PubMed\]](#), [\[CAS\]](#), [Google Scholar](#)
45. **45**
Rasmussen, S. G.; Ogburn, E. L.; McCormack, M.; Casey, J. A.; Bandeen-Roche, K.; Mercer, D. G. Association between unconventional natural gas development in the Marcellus Shale and asthma exacerbations. *JAMA Intern. Med.* 2016, *176*, 1334– 1343, DOI: 10.1001/jamainternmed.2016.2436
[\[Crossref\]](#), [\[PubMed\]](#), [\[CAS\]](#), [Google Scholar](#)
46. **46**
Saadatmand, M.; Foroughi, H.; Dai, T.; Misra, T.; Bensabath, T.; Farnood, R. Odor fading in natural gas distribution systems. *Process Saf. Environ. Prot.* 2015, *94*, 131– 139, DOI: 10.1016/j.psep.2015.01.005
[\[Crossref\]](#), [\[CAS\]](#), [Google Scholar](#)

47. **47**

ICF. Supplemental greenhouse gas analysis of the Danskammer Energy Center,
Prepared for: Danskammer Energy, LLC.

[http://documentsdpsnygov/public/Common/ViewDoc.aspx?](http://documentsdpsnygov/public/Common/ViewDoc.aspx?DocRefId=%7B6C430CE8-D270-4D09-A4A0-031523905E63%7D)

[DocRefId=%7B6C430CE8-D270-4D09-A4A0-031523905E63%7D](http://documentsdpsnygov/public/Common/ViewDoc.aspx?DocRefId=%7B6C430CE8-D270-4D09-A4A0-031523905E63%7D) (accessed 7/20/20).

[Google Scholar](#)

48. **48**

Barletta, B.; Meinardi, S.; Simpson, I. J.; Khwaja, H. A.; Blake, D. R.; Rowland, F. S. Mixing ratios of volatile organic compounds (VOCs) in the atmosphere of Karachi, Pakistan. *Atmos. Environ.* 2002, 36, 3429– 3443, DOI: 10.1016/s1352-2310(02)00302-3

[[Crossref](#)], [[CAS](#)], [Google Scholar](#)

49. **49**

Goetz, J. D.; Avery, A.; Werden, B.; Floerchinger, C.; Fortner, E. C.; Wormhoudt, J. Analysis of local-scale background concentrations of methane and other gas-phase species in the Marcellus Shale. *Elementa* 2017, 5, 1, DOI: 10.1525/elementa.182

50. **50**

Gelencsér, A.; Siszler, K.; Hlavay, J. Toluene– benzene concentration ratio as a tool for characterizing the distance from vehicular emission sources. *Environ. Sci. Technol.* 1997, 31, 2869– 2872, DOI: 10.1021/es970004c

[[ACS Full Text](#)  [ACS Full Text](#)], [[CAS](#)], [Google Scholar](#)

51. **51**

Gulf South Pipeline Company LLC. Receipt Point Gas Composition.

[https://infopostbwpipelinescom/Frameset.aspx?](https://infopostbwpipelinescom/Frameset.aspx?url=%2FPosting%2FGasCompositionPage.aspx&tspid=1)

[url=%2FPosting%2FGasCompositionPage.aspx&tspid=1](https://infopostbwpipelinescom/Frameset.aspx?url=%2FPosting%2FGasCompositionPage.aspx&tspid=1). 2021 (accessed 7/1/21).

[Google Scholar](#)

52. **52**

DiGiulio, D. C.; Jackson, R. B. Impact to underground sources of drinking water and domestic wells from production well stimulation and completion practices in the Pavillion, Wyoming, field. *Environ. Sci. Technol.* 2016, 50, 4524– 4536, DOI: 10.1021/acs.est.5b04970

[[ACS Full Text](#)  [ACS Full Text](#)], [[CAS](#)], [Google Scholar](#)

53. **53**

Algonquin, E. Gas Transmission Hourly Gas Quality. Available from:

<https://infopostenbridgecom/InfoPost/AGHome.aspx?Pipe=AG> (accessed 4/4/22).

[Google Scholar](#)

54. **54**

Morgan, K. Tennessee Gas Pipeline Hourly Gas Quality Data. Available from:

[https://pipeline2kindermorgancom/GasQuality/RepMainLinePoints.aspx?](https://pipeline2kindermorgancom/GasQuality/RepMainLinePoints.aspx?code=TGP&type=5)

[code=TGP&type=5](https://pipeline2kindermorgancom/GasQuality/RepMainLinePoints.aspx?code=TGP&type=5) (accessed 4/4/22).

[Google Scholar](#)

Cited By

Citation Statements

beta

- Supporting

1

- Mentioning

15

- Contrasting

0

Explore this article's citation statements on scite.ai

powered by **scite**

This article is cited by 3 publications.

1. Misbath Daouda, Annie Carforo, Darby Jack, Diana Hernández. Correspondence on “Home is Where the Pipeline Ends: Characterization of Volatile Organic Compounds Present in Natural Gas at the Point of the Residential End User”. *Environmental Science & Technology* **2023**, 57 (4) , 1848-1849.
<https://doi.org/10.1021/acs.est.2c09423>
2. Eric D. Lebel, Drew R. Michanowicz, Kelsey R. Bilsback, Lee Ann L. Hill, Jackson S. W. Goldman, Jeremy K. Domen, Jessie M. Jaeger, Angélica Ruiz, Seth B. C. Shonkoff. Composition, Emissions, and Air Quality Impacts of Hazardous Air Pollutants in Unburned Natural Gas from Residential Stoves in California. *Environmental Science & Technology* **2022**, 56 (22) , 15828-15838. <https://doi.org/10.1021/acs.est.2c02581>
3. Bart Eklund. Comment on “Home is Where the Pipeline Ends: Characterization of Volatile Organic Compounds Present in Natural Gas at the Point of the Residential End User”. *Environmental Science & Technology* **2022**, 56 (17) , 12783-12784.
<https://doi.org/10.1021/acs.est.2c05195>

Abstract

Figure 1

Figure 1. Unique sample locations (black markers) by municipality (red outline) across the three major Greater Boston NG utility providers. Note, some locations were sampled multiple times. *Utility A was acquired by Utility B on October 13, 2020.

Figure 2

Figure 2. BTEX content of NG in raw (markers) and monthly averages (blue). Monthly averaged regional air temperature (°F) from the National Oceanic and Atmospheric Administration (NOAA) is displayed in red. The COVID-related sampling pause from mid-March 2020 through mid-July 2020 is highlighted in the grey box.