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Unburned Methane Emissions from Residential Natural Gas Appliances

Zachary Merrin* and Paul W. Francisco

Applied Research Institute, Indoor Climate Research & Training, University of Illinois, Champaign, Illinois 61820, United States

Supporting Information

ABSTRACT: Methane, the primary component of natural gas (NG), is a potent greenhouse gas. NG is a common fuel for residential appliances because of low cost, high energy density, and relatively clean combustion. NG exhaust contains some unburned methane due to inevitable incomplete combustion. A field campaign measuring methane concentrations in exhaust from residential NG appliances was conducted in Boston and Indianapolis to determine their contribution to overall emissions. NG space heating, water heating, and cooking appliances were measured in 100 homes. Appliance exhaust typically exhibits a brief methane concentration spike during ignition and extinguishment and relatively low concentrations during steady-state operation. Exceptions to this pattern include ovens, suboptimal stove burners, and tankless water heaters, which either have a different operating pattern or nontrivial steady-state concentrations. Findings were combined with appliance usage and prevalence assumptions to estimate total emissions. Annually, ~30 [97.5% CI: 19−160] Gg of methane emissions can be attributed to U.S. residential NG appliances, corresponding to ~830 [530−4500] Gg carbon dioxide equivalent (CO_2e_{100}). This accounts for ~0.1% [0.08−0.7%] of U.S. anthropogenic methane emissions (which account for ~10% of total U.S. greenhouse gas emissions) and corresponds to an emission factor of 0.38 g/kg of NG consumed (0.038% [0.024%−0.21%]).

INTRODUCTION

Natural gas (NG) is a useful fuel, it is abundant, easily transported, has a high specific energy, and cleaner emissions than other common fossil fuels; as such it has diverse applications and is a practical and popular fuel for residential combustion appliances. Since NG is primarily methane, a potent greenhouse gas (GHG), it should be used in a way that minimizes uncombusted and fugitive emissions. Recent top-down research suggests widespread underestimations of anthropogenic methane emissions,[1,2] and multiple bottom-up efforts have been initiated to accurately quantify individual sources. Although significant research has been done to measure and model methane emission from NG processing and distribution infrastructure, limited research exists regarding the fate of methane delivered to residential end users.[3] Field research on residential NG space heating, water heating, and cooking appliances at 100 homes was conducted to determine their contribution to U.S. anthropogenic methane emissions.

METHODS

Sites and Appliances. Recruitment occurred through a variety of sources, including public advertisements and solicitations to home-performance program participants. Testing included 72 location-specific sites in Boston, MA and Indianapolis, IN (areas where substantial bottom-up research has revealed relatively high and low ambient methane levels, respectively[4]) and 28 additional sites (in IL and NY) with tankless water heaters (appliances suspected of having relatively high methane emissions). Appliances were on average 10.9 years old with the oldest being a 34+ year old boiler. Around two-thirds of furnaces and tankless water heaters were high efficiency condensing units, which achieve efficiencies above 90%; only one boiler and one conventional water heater used this technology. Sites and tested appliances are described in Table 1.

Testing. Testing of furnaces, boilers, and water heaters involved accessing the appliance exhaust (through an existing flue access point, a new hole drilled (and later sealed), or via the outdoor termination) and measuring gas concentrations during appliance ignition, operation, extinguishment, and cool down. Range burners were sampled through the device described in the equipment section. Other appliances (dryers, fireplaces, generators, pool heaters, etc.) were not tested for lack of sampling apparatus, protection of the equipment, or absence in the sample set.

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Table 1. Quantity, Distribution, and Characteristics of Appliances Tested

				stoves				water heaters			
	sites[a]	furnaces	boilers	stoves	range burners	ovens	broilers	conventional	tankless	outdoor grills	space heaters
total	101	62	19	57	213	47	30	49	30	2	1
Indianapolis	41	35	0	18	66	12	6	22	1	2	1
Boston	32	5	19	24	97	23	16	27	1	0	0
other	28	22	0	15	50	12	8	0	28	0	0
av age (yr)	NA	10.5	15.9		12.1			6.0	7.7	NA	NA
% 90+ eff.		65%	5%		NA			2%	67%		
multistage	NA	62%	0%		NA			0%	100%	NA	NA

[a]Indicates the total number of sites tested. Some data was rejected due to quality issues; the final data set includes data from 98 of the sites. The appliance numbers are based on those with usable data that are included in the analysis, not the total number tested.

Ovens were tested via the vent on top of the unit. The sequence was initially firing (ignition spike), preheat phase until methane concentrations stabilized, broiler activation (broiler spike, if equipped) and again waiting for concentrations to stabilize, and then deactivating the unit (extinguishment spike). Only seven ovens were observed beyond preheat into the burner cycling phase, when their burners would cycle on and off to maintain temperature; the oven steady-state analysis is based off of this subset.

Equipment. Testing utilized the Picarro G4301 cavity ring-down spectroscopy portable gas concentration analyzer. The instrument measured concentrations in parts per million (ppm) of methane (CH_4), carbon dioxide (CO_2), and water vapor (H_2O), as well as sample temperature and pressure. The instrument was calibrated with a zero gas and 80 ppm of CH_4 span gas multiple times during the field campaign.

The necessity for accurate low-level readings and rapid response to large concentration spikes was an instrumentation challenge, and measurements frequently exceeded the published operating range (0−800 ppm of CH_4, 0−3% CO_2). When concentrations exceeded instrument capabilities, gaps arose in the time series data. When possible the missing data was approximated by extrapolating nearby data (described in Supplement SI-1). To reduce these data gaps, Picarro modified the instrument firmware midproject by increasing the fitting parameters and decreasing the ring-down threshold, ultimately improving the response time and concentration range.

Cooktop burners were tested using a device ("CO Hot Pot" − Figure S3 in SI-2) that mimicked the presence of a cooking vessel and channeled the exhaust plume past a sampling port where representative readings could be taken.

Calculations. Annual methane emissions from an appliance type were calculated by combining average measured concentration, calculated exhaust flow, and appliance usage assumptions according to eq S1 and eq S2. The necessary volumetric flow rate of the appliance exhaust is difficult to measure, as appliance flue exhausts are often in inaccessible locations (e.g., roofs), and positive pressure flow measuring devices are inaccurate and frequently difficult to install in field environments. Therefore, the flow rate was calculated based off of the stoichiometry describing the combustion reaction, the excess air, and the appliance rated fuel consumption. These calculated flows were not validated against any field measurements, and their accuracy and uncertainty are unknown; high concentrations of CO_2 in the ambient combustion supply air or discrepancies between the rated and actual fuel consumption will affect this calculation.

Flue flow rate during operation is calculated from eq 1 using the appliance rated fuel consumption (in British Thermal Units [BTU]), and the excess air is calculated from the measured exhaust CO_2 concentration. Details on derivation and underlying assumptions for eq 1 are presented in Supplement SI-3. For multistage appliances (62% of furnaces and 100% of tankless water heaters), and those with variable input ratings, the highest listed consumption was used. The furnace testing procedure involved increasing the set point several degrees above the current temperature, and depending on thermostat and furnace logic, many may have been operating at their high level; undoubtedly, some were not, resulting in an incorrect consumption assumptions and overestimation of flue flows and emission.

Total Flue Volumetric Flow Rate

$$\text{Flue Flow}\left(\frac{\text{m}^3}{\text{h}}\right) = \text{nat. gas consumed}\left(\frac{\text{BTU}}{\text{h}}\right)*\left(\frac{2.84*10^{-5}}{\text{CO}_2(\text{dry, }\%)/100}\right) \quad (1)$$

Methane Mass. The mass of CH_4 emitted from the calculated volume depends on the flue gas density. Since CH_4 is a gas at these conditions, the density is determined by the temperature and pressure according to the ideal gas law. The time dependent density was calculated from the instrument reported pressure and temperature. The pressure and temperature measurements were taken within the instrument measurement cavity. While not within the flue itself, this measurement is immediately following extraction and is expected to closely reflect flue conditions. The CH_4 concentration data, volumetric emission rate, and the density were then combined to calculate the total mass of CH_4 emitted.

Methane Background. Average atmospheric methane levels are ∼2 ppm.[5] Indoor concentrations can be elevated from biological activities, NG leaks, and unvented combustion. When an appliance intakes combustion air, any additional fuel from ambient methane supplements the supplied NG and is indiscriminately burned.

Indoor methane concentrations encountered in this study had a median of 2.6 ppm (IQR = 2.2−3.5). The highest indoor reading was 35.9 ppm from a basement gas leak; even with this unusually high background concentration, ambient methane in this home only contributed 0.06% of the supplied fuel (50% excess air assumed). Although this negligibly affects the combustion calculations, it can significantly affect overall methane accounting. Since during steady state many appliances emit concentrations below ambient, in some cases, the process may be considered net methane-negative.

While this may be correct from an atmospheric methane accounting perspective, it is misleading from an appliance

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Table 2. Appliance Prevalence and Usage Assumptions

	source: 2015 RECS (except as noted below)[c]		appliance usage assumptions		
	no. of U.S. households using NG for specific end use [in millions][a] (RSE)	NG consumption [trillion BTU] (RSE)	days used per year	activations per day	av operation duration (min)
total	118.2 (0.0)	3965 (2.6)			
space heating[d]	57.7 (2.2)	2677.6 (2.9)	106.5	72	5
furnaces	46.5 (NA)				
boilers	6.6 (NA)				
water heating[e]	56.3 (2.2)	1019.1 (2.3)			
conventional	54.1 (NA)		365	12	5
tankless	2.8 (NA)		365	20	1
cooking[b]	39.0 (3.1)	113.0 (4.2)			
oven			150	1	30
broiler			5	1	15
stovetop			365	1	15
outdoor grilling	4.1 (9.6)	no data	NA	NA	NA
clothes drying	18.2	36.4 (5.2)	NA	NA	NA
pool heaters	NA	11.5 (18.1)	NA	NA	NA
hot tub heaters	NA	5.6 (16.2)	NA	NA	NA

[a]Total U.S. includes all primary occupied housing units in the 50 states and the District of Columbia. Vacant housing units, seasonal units, second homes, military houses, and group quarters are excluded. [b]Cooking includes fuels used by major cooking equipment (ovens, cooktops, and stoves). [c]SOURCE: 2015 Residential Energy Consumption Survey (except as mentioned below). [d]Main space heating equipment only. [e]Tankless water heaters estimated at 5% of total.

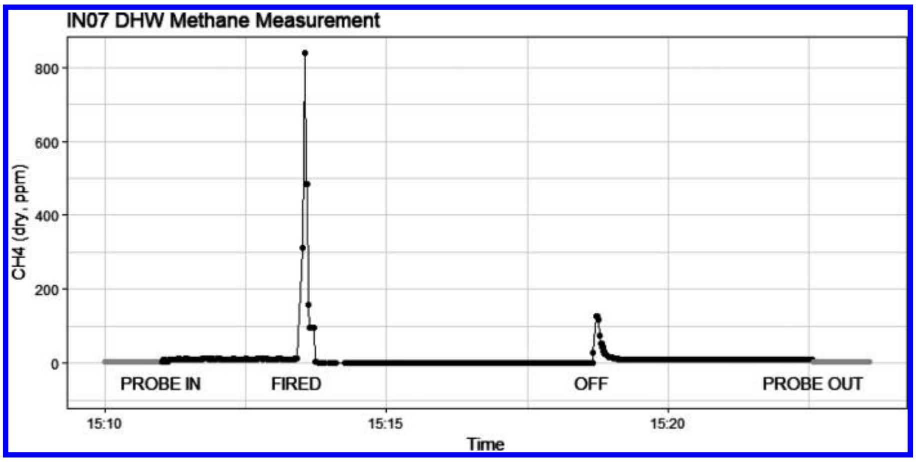

Figure 1. Example of typical CH_4 concentration vs time pattern.

performance perspective. Regardless of ambient methane concentrations, incomplete combustion is inevitable, and comparable exhaust concentrations are expected. This issue is further complicated since elevated indoor concentrations are typically related to the appliance even if they are not a direct result of incomplete combustion. To deal with this complication, results are presented as absolute emission metrics with additional information provided about the quantity of ambient methane consumed.

Assumptions. Extrapolating real-world significance from the collected and calculated data requires several assumptions about U.S. appliance prevalence and usage patterns, which are summarized in Table 2 and explained in Supplement SI-4. Uncertainty in terms of relative standard error (RSE) for each metric in the Residential Energy Consumption Survey (RECS) is included for informative purposes; however, since they are small compared to the uncertainty from the distribution of measurements (see Uncertainty section), they are ignored in the conclusions.

RESULTS

Concentration vs Time Patterns. Most appliances exhibited similar CH_4 emission patterns with a rapid spike and decay on ignition, a low and steady concentration during operation (often below the ~2.6 ppm ambient concentrations encountered in many homes), and an additional spike and decay during extinguishment (Figure 1).

Some appliances diverged from the standard pattern. Ovens (Figure 2 – top) exhibit a unique operational pattern among residential appliances; the CH_4 emission profiles have an extended ignition decay (relative to other appliances) culminating at a relatively steady concentration. Once the thermostat is satisfied, the appliance regulates temperature by continuously cycling the burners on and off (Figure 2 – top-

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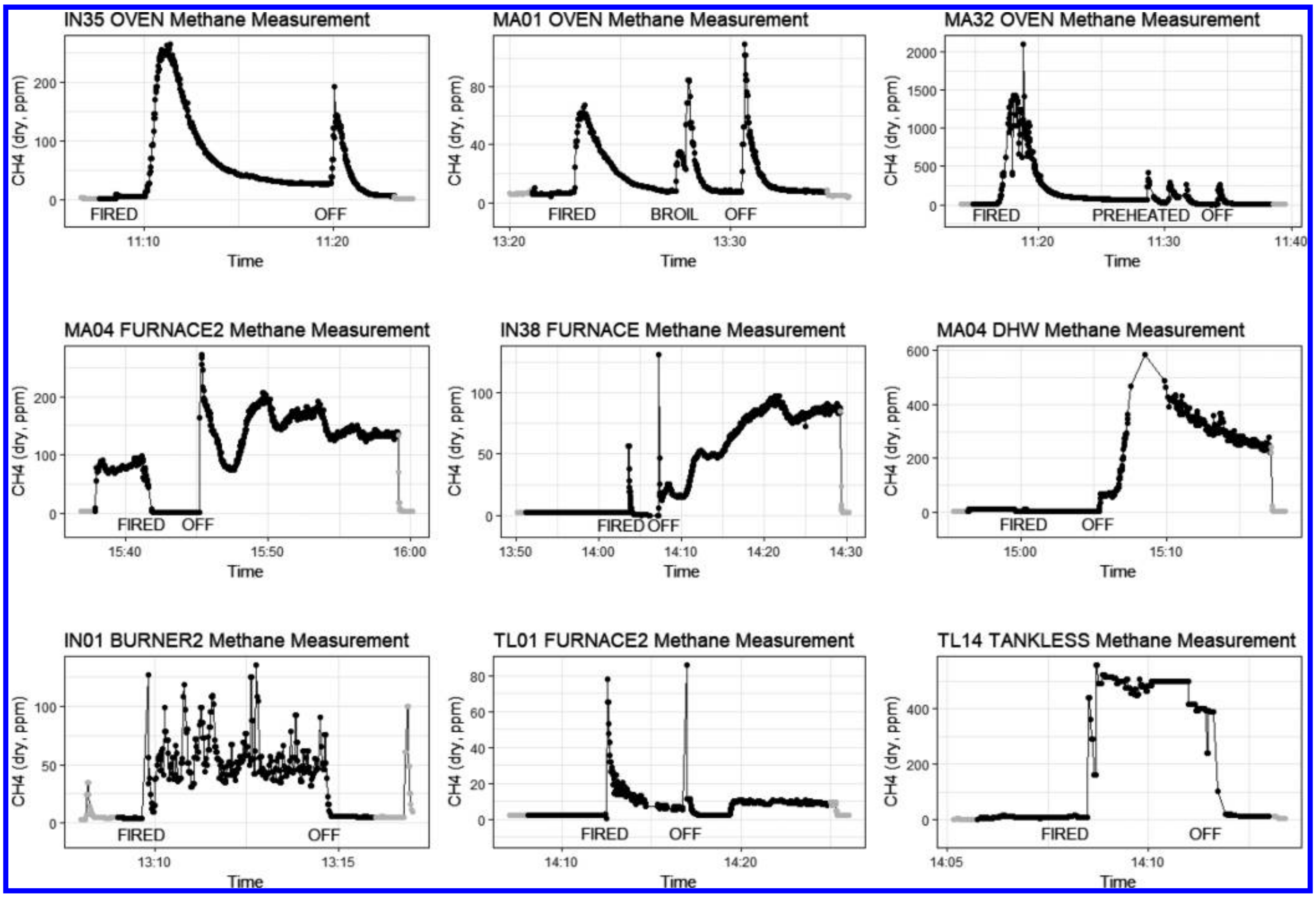

Figure 2. Top: Example of typical oven CH_4 concentration vs time patterns: without broiler (left), with broiler (center), including postpreheating burner cycling (right); Middle: Examples of high efficiency appliances with stable (left), increasing (center), and decreasing (right) standing CH_4 concentrations in flue following device extinguishment; Bottom: Example of appliances with nonzero steady-state CH_4 concentrations.

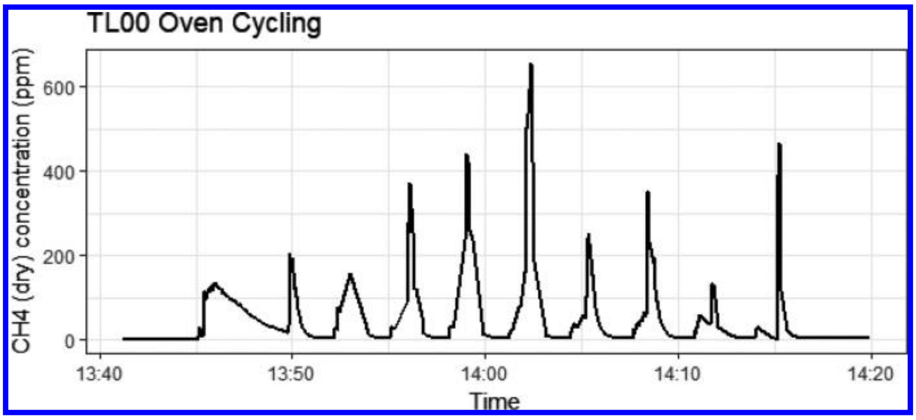

Figure 3. Example of oven data showing burner cycling to maintain temperature during extended test.

right). Not all oven measurements captured this cycling phase (Figure 2 − top-left). Ovens also exhibited an additional spike and decay when the broiler feature (if equipped) was activated (Figure 2 − top-center). When the oven is turned off, an extinguishment spike occurs, analogous to the other appliances.

During steady-state operation, the CH_4 concentrations continuously spike and decay in a cyclical sawtooth pattern (Figure 3). Previous research found that following preheating, ovens continuously cycle on and off at ~86-s intervals, with combustion occurring for approximately half of that time.[6] A minority of the tested tankless water heaters exhibited burner-cycling behavior similar to the ovens for all or part of their test (Figure S4).

Many sealed combustion appliances exhibited a standing CH_4 concentration in the flue preceding and/or following operation (Figure 2 − middle). In some appliances that exhibited this phenomenon, the concentration was stable or trending in one direction, and in some, it drifted with no discernible trend. This phenomenon suggests that the flue gases remain stagnant in the flue in these appliances, until the next call for operation occurs and the flue is flushed.

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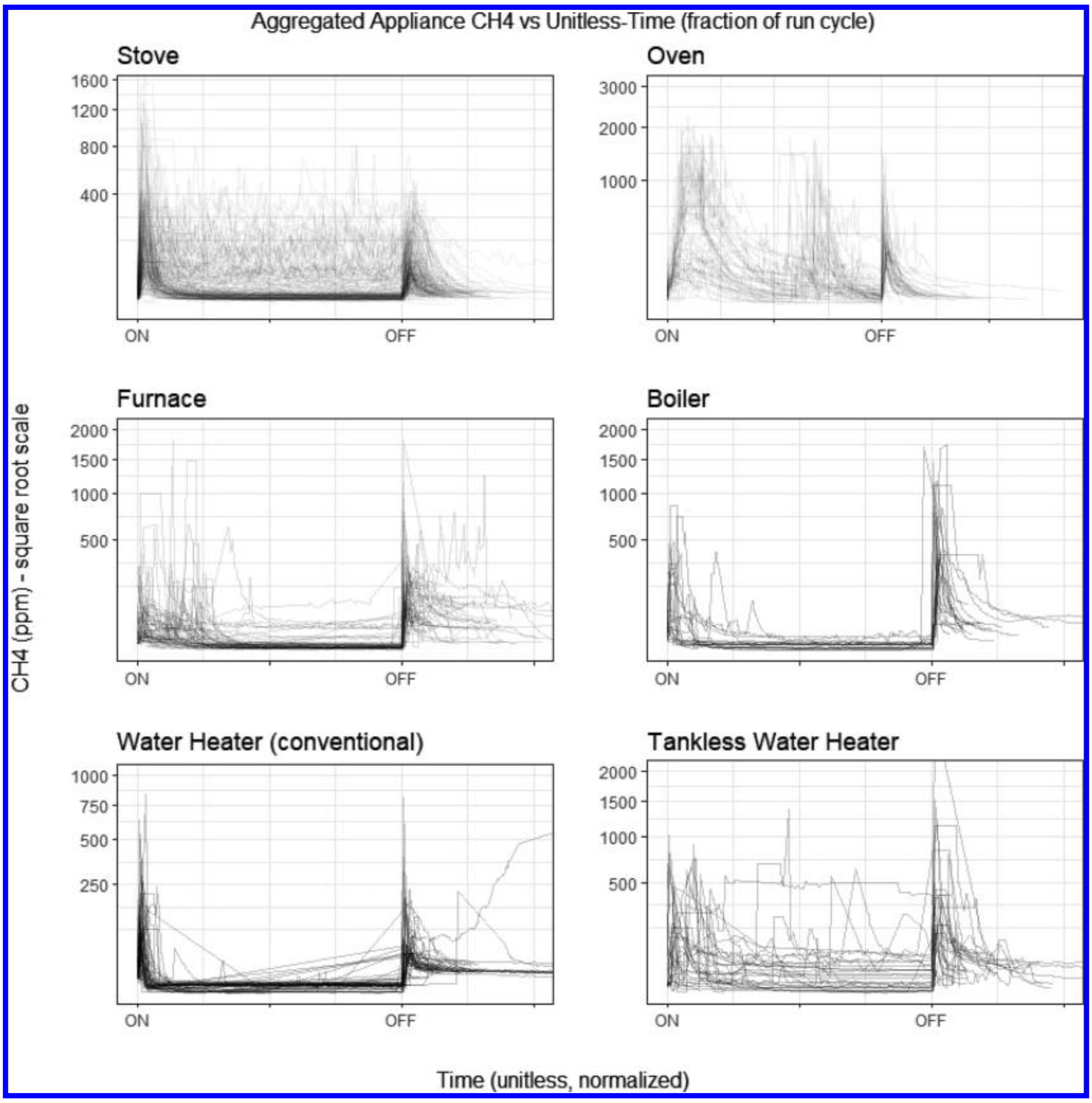

Figure 4. Aggregate appliance methane vs time (fraction of full on-cycle) profile.

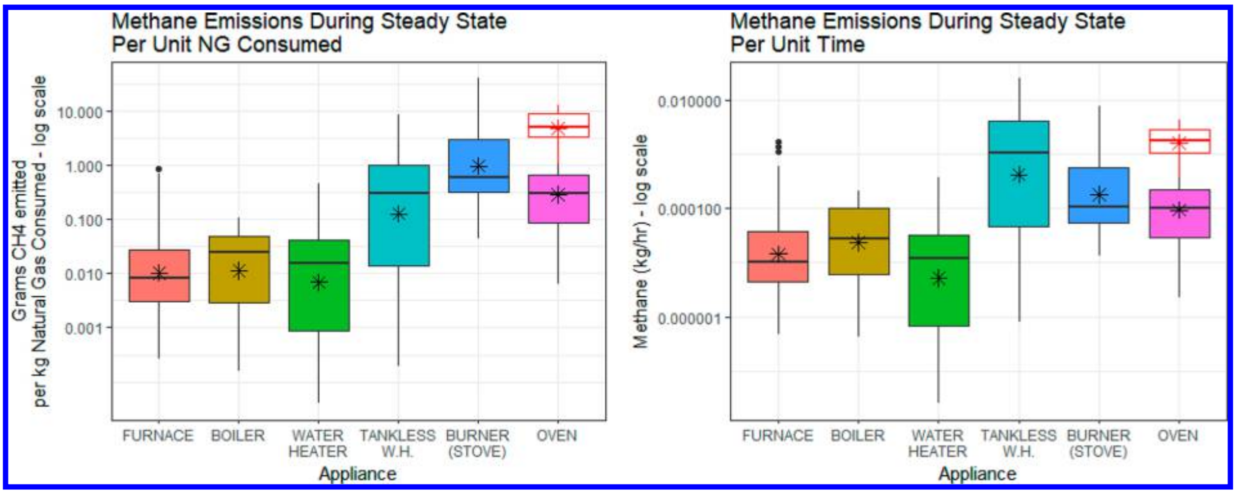

Figure 5. Emissions during steady-state operation (left) per unit gas consumed and (right) per unit time; oven results based on burner cycling indicated by unfilled red boxplot. Boxplots represent median and 25th−5th quartile, with the whiskers to the furthest point within 1.5× the interquartile range (IQR), star = mean.

Some appliances exhibited nonzero CH_4 concentrations during steady state (Figure 2 − bottom). This was most common in stove burners (Figure 2 − bottom-left) but was also present in numerous tankless water heaters (Figure 2 −

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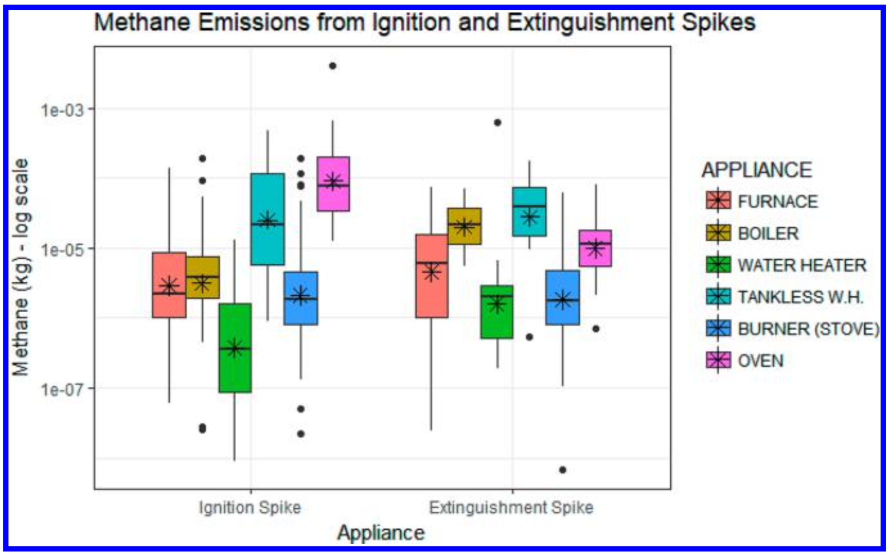

Figure 6. Emissions from ignition and extinguishment spikes. Boxplots represent median and 25th−75th quartile, with whiskers to the furthest point within 1.5× the IQR, star = mean.

bottom-right) and a few space heating appliances (Figure 2 − bottom-center). Figure 4 shows the aggregate emissions profiles for each type of appliance tested (normalized to the individual test duration).

Per Event Emissions. The following graphs (and Table S1) describe the magnitude of CH_4 emissions from each appliance type, including operational emission rates (Figure 5), and absolute quantities from ignition and extinguishment spikes (Figure 6). The total amount released from an operation cycle is the sum of the ignition and extinguishment spikes plus the steady-state emission rate times the operation duration (eq S3).

Calculating total emissions for ovens required unique methods, which considered intraoperational spikes and decays. The testing procedure prioritized observing broiler behavior over steady-state behavior, and most ovens were not observed following preheating. Consequently, oven steady-state results are calculated two ways: based on the stable concentrations measured at the end of the ignition spike which incorrectly assumes no burner cycling (magenta "OVEN" boxplot in Figure 5) and using assumptions based on the cycling behavior of the seven ovens for which operational cycling data exists (unfilled red boxplot in "OVEN" category in Figure 5). These results suggest that ovens generate the most unburned CH_4 per unit time because of their repeated burner cycling.

It is possible that some tankless water heaters cycle their burners when there is insufficient or inconsistent demand, but since most models have variable fuel input rates, this is not expected to be standard practice. Water heater analysis did not assume burner cycling.

Conventional water heaters typically had standing concentrations in their flues (with mean concentrations following extinguishment of 12.3 ppm and a standard deviation of 8.6 ppm), likely a result of the standing pilot light. There is nonzero flow through the flue (although extremely low compared to during operation) to support this combustion. The residual concentrations measured in water heater flues could not be extrapolated to total mass emissions because of the inability to calculate flue flow during that time (see Supplement SI-4 for more information); emissions related to pilot lights are not included in water heater emission rates. In a related study of residential emissions in California, ∼30% of appliance CH_4 emissions were attributed to pilot lights.[3]

With the exception of ovens, and their unique cycling behavior (depicted by the unfilled red boxplot in Figure 5), cooktop burners have the highest median steady-state emissions per unit fuel at ∼0.6 g/kg fuel, while tankless water heaters have the highest median steady-state emissions per unit time at ∼1 g/h (Figure 5). Both have steady-state emissions about 1−2 orders of magnitude greater than space heating and conventional water heating appliances, but stove burners have lower consumption (∼5−20 kBTU/h, Figure S5) compared to tankless water heaters (which generally have the highest consumption of any appliance, typically ∼150−200 kBTU/h, Figure S5). Furnaces, which tend to have the most usage of any appliance type, have the lowest median emission rate at about 0.008 g/kg fuel. Oven emission rates are about an additional order of magnitude above stovetop burners and tankless water heaters when cycling is considered.

Emissions from the ignition and extinguishment spikes (Figure 6) cannot be directly compared to those for steady-state operation, since they are expressed in a per-event absolute mass as opposed to per unit time rates. Additionally the amount released is highly dependent on duration of the spike. Ovens, with long drawn out ignition spikes, release more unburned CH_4 during the ignition process (median of about 0.079 g/event) than other appliances with shorter ignition spikes. This does not include emissions from oven burner cycling which is part of their normal steady-state operation.

Tankless water heaters have the second highest level of ignition spike emissions, with a median of about 0.02 g/event. They also had the highest extinguishment spike at a median of 0.04 g/event. For each appliance type, emissions from individual units ranged over 1 order of magnitude, with up to about 3 orders of magnitude for some types.

Uncertainty. Uncertainty is introduced from several sources during the data collection and analysis, including instrument limitations, sample size, exhaust flow rate assumptions/calculations, limited appliance observation, and extrapolation assumptions. Although each of these considerations factors into the overall uncertainty, it is difficult to quantify them or their aggregate effect with any confidence.

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Table 3. Per Appliance Annual Emissions

	emissions in kg/year (97.5% confidence interval)					
	total[a]	ignition spike	steady-state operation {% of total}[b]	extinguishment spike	sum	CH_4 consumed (kg/year) {% of total}
furnace	0.22 (0.14−0.51)	0.1 (0.048−0.2)	0.069 (0.022−0.1) {24.2%}	0.12 (0.067−0.68)	0.290	0.072 {33%}
boiler	0.32 (0.15−0.75)	0.17 (0.037−3.6)	0.041 (0.018−0.31) {9.8%}	0.21 (0.11−0.45)	0.420	0.17 {53%}
water heater	0.077 (0.02−0.084)	0.0083 (0.0041−0.034)	0.01 (0.01−0.24) {6.5%}	0.14[d] (0.0089−0.12)	0.160	0.037 {48%}
tankless W.H.	1.2 (0.98−41)	0.67 (0.28−3.6)	0.5 (0.31−120) {31.1%}	0.43 (0.12−5.8)	1.600	0.11 {9%}
stove	0.056 (0.04−0.071)	0.0025 (0.0016−0.0028)	0.054 (0.035−0.062) {93.2%}	0.0014 (0.0011−0.0017)	0.058	0.009 {16%}
oven	0.13 (0.11−0.14)	0.034 (0.018−0.045)	0.092 (NA[c]) {71.8%}	0.0021 (0.0016−0.0032)	0.130	0.003 {2%}

[a]Total is based on the average total for individual appliances and does not equal the sum of the row; calculation and conclusion are based on this value. [b]Percentage of total emissions attributed to steady-state operation (based on the aggregate average of the combustion components presented in this table). [c]Confidence interval is not available for oven steady state as the value is based on too few measurements. [d]Water heater extinguishment spike mean exceeds confidence interval due to unique 100 kBTU/h sealed combustion appliance which exhibited an extended extinguishment spike and decay.

Therefore results are presented as is, with the caveat that they are not exact; only the uncertainty from the measured results distribution is presented in a quantitative manner. As a result, the conclusions are considered at best an order of magnitude approximation.

Several of the emission metrics with a sufficiently large sample approached log-normal distribution (Figure S7-Figure S10). The modified Cox method[7] was employed to quantify the 97.5% confidence intervals of the results, which are presented in Table 3.

Overall U.S. Emissions. *Appliance Annual Emissions.* Using the assumptions presented in Table 2, annual emissions were calculated for each appliance type and are summarized in Table 3. In this table, the "total" column is the average overall CH_4 emissions for a specific appliance type based on individually tested appliances. However, because of asymmetric emission rate distributions within appliance types, the "total" value does not equal the sum of the individual component averages (ignition spike, steady state, extinguishment spike). The sum of the average individual components is shown in the "sum" column and is used to calculate the percent attributed to steady-state operation. The "CH_4 consumed" column describes the quantity of ambient methane the appliance consumes from the combustion air, and the percentage refers to the percent of total emissions that it represents; a value of 100% implies that the process is net methane neutral.

Tankless water heaters have the highest per-appliance annual methane emissions, at about 1.2 kg each, followed by space heating appliances, at about a quarter of that level, despite their greater runtimes. Despite cooking appliances having higher emission rates per unit fuel, their duty cycles are such that they contribute substantially less total methane per individual appliance compared to tankless water heaters and space heating equipment.

Total National Emissions. CH_4 emission rates by appliance type were comparable regardless of location. Although climate driven differences will affect usage and total emissions, since appliances are mass-produced with widespread distribution, it is unlikely that location will influence the unburned CH_4 emission rate. Average emission rates from all tested appliances were assumed representative of nationwide appliance performance.

Table 4 summarizes the total amount of unburned CH_4 emitted in the U.S. annually from each of the main residential appliances, for a total emission of ∼30 Gg (97.5% CI: 19−160 Gg). The net methane generated (considering methane consumed from the ambient air) is ∼21 Gg (97.5% CI: 11−154 Gg). These emission calculations exclude appliances not tested in this study (e.g., dryers), but since space/water heating and cooking account for ∼96% of residential NG consumption (Table 2), contributions from other appliances should be minimal.

Table 4. Total Methane Emissions

appliance	methane emissions [Gg/year] (95% confidence interval)	contribution to total U.S. residential NG appliance emissions (based on mean values)	net methane generation [Gg/year][a]
furnace	11.5 (7.3−26.6)	39%	7.7 (3.6−22.8)
boiler	2.4 (1.1−5.5)	8%	1.1 (−0.2−4.3)
water heater	4.1 (1.1−4.5)	14%	2.1 (−0.9−2.5)
tankless W.H.	3.2 (2.8−115.4)	11%	2.9 (2.5−115.1)
stove	3.3 (2.3−4.2)	11%	2.7 (1.8−3.6)
oven	5 (4.3−5.5)	17%	4.8 (4.2−5.3)
total	29.5 (18.9−161.7)	100%	21.3 (11−154)

[a]Accounts for CH_4 consumed from ambient air.

This calculated appliance use consumes ∼80 Tg of NG annually (slightly under the ∼4.4 million ft^3 or ∼91 Tg of NG delivered to residential end users in 2017,[8] which includes NG used by appliances not tested in this study, and fugitive emissions), resulting in an emission factor for methane of 0.38 g per kilogram of NG consumed (0.038%, 97.5% CI: 0.024%−0.21%). Fischer calculated an overall residential postmeter emission factor of 0.5% including pipe leaks and pilot lights and ∼0.11% for the appliance contribution (excluding pilot lights).[3] It is important to note that Fischer's methods differed in that only steady-state emissions were measured, as did their appliance assumptions, since the study was based in temperate California, where for example relatively low-emitting space

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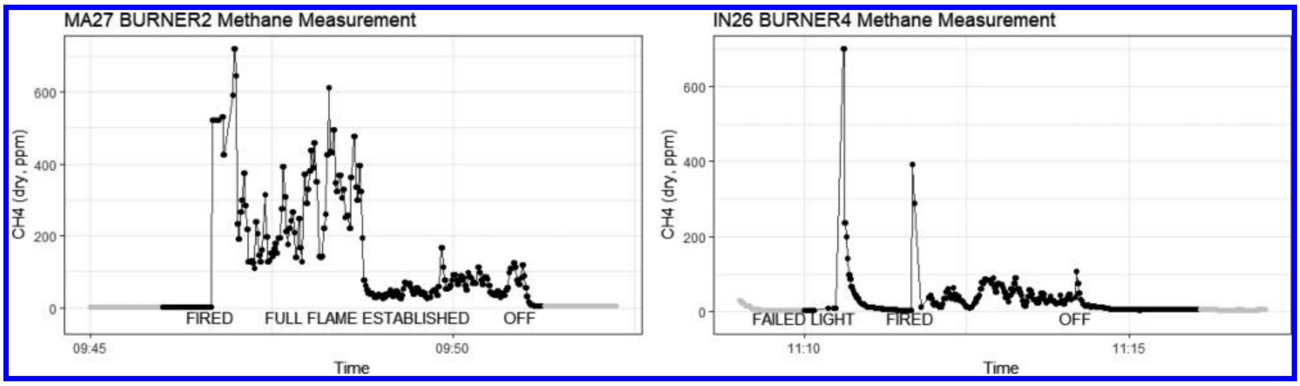

Figure 7. CH_4 concentration profiles from stove burners not performing optimally, showing delayed full flame establishment (left) and unsuccessful ignition and incomplete combustion (right).

heating is a less significant end use. When evaluating comparable results from Fischer's and this study (only steady-state emissions, just applied to California's population, excluding space heating), Fischer's emissions were ~15× and ~3× higher for water heating and stove top end uses. Because of the large uncertainty on the calculated tankless water heater emissions, Fischer's water heating results are within the confidence interval of this data.

Although tankless water heaters account for only a small portion of all residential NG appliances (assumed as 5% of all water heaters), they are responsible for a disproportionate part (~11%) of the total CH_4 released and ~40% of all CH_4 attributed to water heating.

DISCUSSION

Significance. Methane emissions are typically discussed regarding their global warming potential in terms of carbon dioxide equivalent on both a 20-year (CO_2e_{20}) and a 100-year (CO_2e_{100}) time scale.[9] Both metrics are important to consider, especially for methane, which absorbs more infrared radiation than CO_2 but has a shorter atmospheric lifetime. Methane has a CO_2e_{20} of ~84 and a CO_2e_{100} of ~28,[10] meaning 1 g of methane is equivalent to 28 g of CO_2 when considering a 100-year time horizon. The U.S. EPA estimated 657.4 Tg CO_2e_{100} of methane was released in 2016, primarily from energy production/distribution/use, agriculture, and waste management.[11] The ~30 (97.5% CI: 19−160) Gg of methane calculated from this research equals ~830 Gg CO_2e_{100} (97.5% CI: 530−4500 Gg) and ~2.5 Tg CO_2e_{20} (97.5% CI: 1.6−14 Tg), accounting for ~0.13% (97.5% CI: 0.08−0.7%) of total methane emissions. To add additional context to these metrics, combustion of the quantity of NG assumed for this analysis generates ~205 Tg of actual CO_2. Previous research estimates that total U.S. CO_2 emission from residential space and water heating (from all fuel sources, including electric) is 338 Tg and 157 Tg, respectively.[12]

Given the assumptions required for this analysis and the large variation in individual appliance performance, the results including the confidence intervals should be interpreted as an approximation.

Specific End Uses. Space heating is the most significant residential energy end use, accounting for 42% of overall energy consumption,[13] and an even larger percentage of NG consumption. Considering space heating is also the most common end use of NG (Table 2), it is not surprising that it is responsible for approximately half of residential unburned methane emissions.

To minimize the standby losses associated with conventional storage water heaters, tankless water heaters lack (or significantly reduce) storage of preheated water. Consequently, they require powerful burners and rapid cycling to satisfy hot water demands. As a result, per individual unit tankless water heaters generate the most unburned methane of the tested appliances. Tankless water heaters generate the second highest amount of unburned methane from their ignition spike (after ovens, which have a uniquely long ignition spike and decay) and the highest from the extinguishment spike (see Supplement SI-5). This, combined with their frequent activation and deactivation (every time there is a call for hot water), results in their large overall emission.

The amount of energy savings associated with tankless systems varies with household characteristics and system configuration. Households with low hot water consumption will have higher savings than those with moderate to high consumption.[14] A configuration with a tankless water heater near every hot water outlet will result in the highest energy (and water) consumption reduction, at the cost of higher equipment and installation costs. If the popularity and prevalence of tankless water heaters continues to grow, overall methane emissions attributed to residential appliances will increase. It is important to note that methane represents a small portion of the GHG emissions from NG appliances, with the considerably larger volume of CO_2 accounting for the majority. For appliances such as tankless water heaters, the CO_2 reduction from any efficiency gains relative to their conventional counterparts should more than compensate for the larger CH_4 emissions from an overall GHG perspective.

Stove burner emissions can be significantly higher when improperly seated burner caps or other factors (such as cleanliness or disrepair) inhibit the stove's ability to establish a complete flame. Figure S6 shows a variety of stove burners that were not operating optimally, and Figure 7 shows measured CH_4 emissions measurements from two of those burners.

Figure S6 (left) shows an improperly seated stove burner cap resulting in an asymmetric flame with large flame cones on one side and unignited burner ports on the other. This situation results in incomplete combustion with high concentrations of unburned CH_4. When the burner cap was repositioned correctly, the flames established a much more normal pattern (not pictured), and the emissions were significantly reduced.

Figure S6 (center) shows a stove burner that had difficulty establishing a complete flame; once the flames were fully established, the CH_4 emissions dropped (Figure 7, left). When

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burners have difficulty establishing a complete flame, the process can be assisted with an external ignition source or by gently blowing on the established flames and agitating them to ignite adjacent flames. Figure S6 (right) shows a stove that had difficulty lighting with the built-in igniter; after the CH_4 plume from the unsuccessful ignition dissipated, the stove was successfully lit using a utility lighter (Figure 7, right). This burner was described by the field tech as "gusty", alluding to the sound the burner makes by releasing uncombusted gas. Following ignition, both of these burners emitted relatively high concentrations of CH_4 (Figure 7) confirming that they were not in optimal operational condition.

Ovens had the lengthiest ignition spike relative to other appliances; they also exhibited the highest operational emissions per unit time because of their continuous burner cycling behavior to modulate temperature. One aspect of realistic cooking that was not emulated by the testing procedure was oven door opening. Ovens are designed to operate with the door closed; opening the door disrupts the airflow and can lead to incomplete combustion, potentially releasing CO and CH_4.[15]

Opportunities. Although unburned methane is not a typical metric used to evaluate appliances, it does impact efficiency ratings. Appliance manufacturers have expended serious efforts to make use of all the possible energy from a given fuel input. Some of the unburned methane (for example during ignition and extinguishment spikes) could be a result of other combustion priorities such as safety or equipment reliability and longevity.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b05323.

> Information on data correction procedures, additional figures (including histograms of emission distributions), supplemental calculations, usage assumption justifications, and summary table of emissions metrics[16−22] (PDF)

AUTHOR INFORMATION

Corresponding Author

*E-mail: zmerrin@illinois.edu.

ORCID

Zachary Merrin: 0000-0001-8600-9193

Notes

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