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An Estimate of Natural Gas Methane Emissions from California Homes

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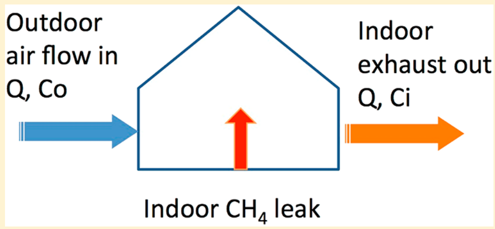
Supporting Information

ABSTRACT: We estimate postmeter methane (CH_4) emissions from California's residential natural gas (NG) system using measurements and analysis from a sample of homes and appliances. Quiescent whole-house emissions (i.e., pipe leaks and pilot lights) were measured using a mass balance method in 75 California homes, while CH_4 to CO_2 emission ratios were measured for steady operation of individual combustion appliances and, separately, for transient operation of three tankless water heaters. Measured quiescent whole-house emissions are typically <1 g CH_4/day, though they exhibit long-tailed gamma distributions containing values >10 g CH_4/day. Most operating appliances yield undetectable CH_4 to CO_2 enhancements in steady operation (<0.01% of gas consumed), though storage water heaters and stovetops exhibit long-tailed gamma distributions containing high values (∼1−3% of gas consumed), and transients are observed for the tankless heaters. Extrapolating results to the state-level using Bayesian Markov chain Monte Carlo sampling combined with California housing statistics and gas use information suggests quiescent house leakage of 23.4 (13.7−45.6, at 95% confidence) Gg CH_4, with pilot lights contributing ∼30%. Emissions from steady operation of appliances and their pilots are 13.3 (6.6−37.1) Gg CH_4/yr, an order of magnitude larger than current inventory estimates, with transients likely increasing appliance emissions further. Together, emissions from residential NG are 35.7 (21.7−64.0) Gg CH_4/yr, equivalent to ∼15% of California's NG CH_4 emissions, suggesting leak repair, improvement of combustion appliances, and adoption of nonfossil energy heating sources can help California meet its 2050 climate goals.



1. INTRODUCTION

1.1. California Total and Natural Gas Methane Emissions. Methane (CH_4) is a potent but short-lived greenhouse gas (GHG) that is emitted from a variety of natural and anthropogenic sources.[1] Lowering CH_4 emissions is an important part of California's climate goals to reduce GHG emissions by 40% to 80% by 2030 and 2050, respectively.[2] While anthropogenic CH_4 has agricultural, waste management, and oil and gas sources, emissions from the natural gas (NG) sector appear particularly important in urban areas where gas is consumed. Three atmospheric studies using other trace gases for source apportionment have found that natural gas sources may constitute 20−100% of regional CH_4 emissions from urban areas.[3−5] In this respect, NG emissions pose a potentially important challenge for successfully implementing "carbon-neutral" communities. For example, an ∼3% leak of unburned CH_4 produces the same short-term (20 yr) warming as the remaining ∼97% of carbon emitted as carbon dioxide from fuel combustion, assuming the IPCC[6] 20-yr global warming potential for methane (84 g CO_2eq/g CH_4).

While the origins of urban NG CH_4 emissions are uncertain, some studies have begun to disentangle this problem. For example, Lamb et al. measured emissions from NG distribution metering and regulating stations in 13 urban systems,[7] while Von Fischer et al. showed that leakage from distribution pipes varied with the age and the type of pipe materials.[8] In California, Hopkins et al. measured CH_4 plumes from a variety of sources in the Los Angeles area and used stable CH_4 isotope measurements to attribute emissions to biological versus thermogenic fossil CH_4 sources,[9] and Fischer et al. reported observable NG CH_4 emissions for a small sample of houses and appliances in the San Francisco Bay Area, suggesting the need for more comprehensive measurements.[10]

To provide quantitative estimates of postmeter NG CH_4 emitted from plumbing and appliance use, we report measurements of NG CH_4 emissions from a sample of 75 single-family California homes and a subset of their combustion appliances. We describe the broad characteristics of California homes and the range of house construction types that were selected for sampling. Two measurement methods were used to quantify 1) whole-house quiescent CH_4 emissions from the combination of pipe leaks and pilot lights when appliances are not operating and 2) CH_4 emissions from individual operating combustion appliances. We then describe the Bayesian statistical sampling procedure used to extrapolate from the study measurements to represent the larger California

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residential building stock. We describe the observed whole-house quiescent CH_4 emissions, CH_4 to CO_2 enhancements for steady operation of combustion appliances in the 75 houses sampled and transient operation of three separate tankless water heaters. We then discuss extrapolation of the measurements to estimate total residential NG CH_4 emissions in the California housing stock and compare the residential emissions with total NG CH_4 and total CH_4 emissions in California. We conclude with recommendations for further research and some avenues for emissions mitigation.

2. METHODS

2.1. Home Recruitment. We selected homes for this study to represent the California housing stock using information from the U.S. Census Bureau.[11] Because roughly 2/3 of California residences are single-family detached homes, our study focused on this housing type. In terms of fuel use, NG is the dominant source of energy for space and water heating and cooking in California single-family homes[12] (henceforth 2011AHS). Summary figures for 2011AHS are provided in Supplement S1. While not explicitly included in this study, we have made a simplifying approximation that CH_4 emissions from multifamily housing including apartments can be estimated based on results from single-family homes. We expect this to be reasonable because multifamily housing shares many important characteristics with single family housing (e.g., NG plumbing and smaller appliances), though we acknowledge some distinctions (e.g., the prevalence of wall heaters and centralized heating) deserve consideration in future work.

The homes in this study were recruited by an energy efficiency analysis and retrofitting contractor (Richard Heath & Associates Inc., henceforth RHA) using existing customers and professional contacts. In total, 75 homes were selected to span the ranges of building age, floor area, number of stories, and foundation type identified in the 2011AHS. Home eligibility criteria include owner-occupied, single-family detached homes that use NG for at least two of the following purposes: space heating, water heating, cooking, and clothes drying. Before conducting quantitative CH_4 leak measurements, study participants filled out an occupant survey, field technicians noted conditions of the gas appliances, and qualitative gas leaks were observed using either a hand-held electronic combustion gas leak detector (e.g., Sensit) or soap solution to detect bubbles. Here, we note that leak testing was performed to detect safety issues but were not comprehensive in that the technicians did not test pipes and fittings that were hard to reach (e.g., behind walls or recessed in shallow crawl spaces).

2.2. Methane Emission Measurements. The majority of the measurements described in this study were derived from whole-building quiescent and combustion appliance emission measurements in the 75 California homes by RHA as described below. Additional details of the measurement methods, including time dependence of indoor CH_4 during depressurization, attribution of CH_4 to natural gas sources, and transient tests of tankless water heaters, are included in Supplement S2.

2.2.1. Whole-Building Quiescent Emission Measurement. Methane emissions from interior leaks and quiescent appliances (with only pilot lights burning) were measured using a mass balance approach. As shown in Figure 1, a controlled flow of outdoor air is used to ventilate the house, while measuring both the indoor and outdoor air CH_4

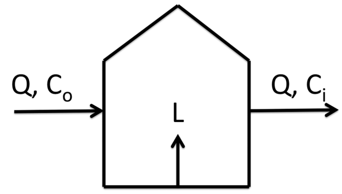

Figure 1. Schematic showing air flows into and out of a house during the building depressurization experiment and indoor CH_4 leak. The volumetric air flow, Q, of outdoor air with the mixing ratio, C_o, enters the home, mixes with indoor methane leaks, L, from gas pipes and pilot light emissions, and is exhausted at higher CH_4 concentration, C_i.

concentrations over time. Once indoor CH_4 concentration reaches steady state, the enhancement of indoor CH_4 relative to outdoor air $(C_i - C_o)$ combined with the known volumetric flow rate, Q, of air can be used to estimate indoor CH_4 emissions as

$$L = Q(C_i - C_o) \tag{1}$$

In this study, we used a commercial blower door system (The Energy Conservatory Inc., DG-1000) to ventilate (∼10 air changes per hour) and depressurize the house (∼−50 Pa at the blower door), opening all interior doors and applying small box fans in hallways to increase air mixing between locations with gas appliances to the blower door exhaust. CH_4 was measured with a portable total CH_4/CO_2 gas analyzer (Los Gatos Research, UGGA). The analyzer had a typical CH_4 measurement precision of ∼0.3 ppb for data collected at 1 sample per second, with both the CH_4 and CO_2 volumetric mixing ratios reported in total (moist) air. Indoor and outdoor measurements were alternated every 2 min using a solenoid valve controlled by the analyzer. The time response of the instrument and sample tubing was measured to have a 1/e response time of ∼10 s, more than sufficient to determine a valid mean value for indoor and outdoor CH_4 after excluding the first minute after each valve switch. Uncertainty in the leak rate, L, was estimated by standard propagation of measurement uncertainties in Q and $(C_i - C_o)$.

As a test of the instruments and mixing, we also conducted a controlled CH_4 release test for each house. We released 5 ± 0.6 g CH_4/day of CH_4 at a location roughly 5 m from the blower door and measured the step response of the indoor CH_4 enhancement $(C_i - C_o)$. The CH_4 was released for 10−15 min using 3.9 ± 0.1% CH_4 in air from a compressed gas cylinder through a regulator at a flow rate of 125 ± 15 sccm (standard cubic centimeters per minute), set using a calibrated rotometeric (ball gauge) flow meter (where we note 1 sccm CH_4 = 1.03 g CH_4/day). We note the uncertainty in the flow rate was estimated from typical drifts in the flow meter reading over time under experimental conditions. In practice, the estimated total CH_4 emissions due to the combination of the house and the additional source, $L_{house+cal}$, were estimated using eq 1, and the additional leak was then estimated from the difference as $L_{cal} = L_{house+cal} - L_{house}$. In the analysis section below, we examine the sensitivity of the distribution of whole-house results to cases where L_{cal} differs from the known value. Here, we note that while the depressurization will gather air containing CH_4 leaks in portions of the house with ventilating air flow, it is possible that leaks occurring in decoupled spaces with little or no induced air flow (e.g., a crawl space or pipes outside the house) will be underestimated with this technique.

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In addition to the 75 homes studied, we re-examined 7 whole-building measurements of $^{13}CH_4$ isotope ratios measured in a previous study[10] that provide supporting evidence that the majority of those whole-building CH_4 enhancements are from natural gas sources (see Supplement S2 for details).

2.2.2. Combustion Appliance Emissions. Methane emissions were measured during steady operation for two combustion sources (either operating gas appliances or pilot lights) in each of the 75 homes. CH_4 emissions were estimated as the product of the fractional enhancement in CH_4 relative to enhancement of CO_2 in exhaust gas, ΔCH_4:ΔCO_2, and the measured volumetric gas consumption rate, Q_g, as

$$E = Q_g * \Delta CH_4 : \Delta CO_2 \tag{2}$$

where $\Delta CH_4:\Delta CO_2 = (CH_{4exh} - CH_{4bg})/(CO_{2exh} - CO_{2bg})$. Subscripts "exh" and "bg" refer to concentrations of CH_4 and CO_2 measured in exhaust and background air, respectively, and Q_g is estimated from repeated gas meter readings. Combustion measurements were made using the same portable gas analyzer used for whole-house measurements. Except for pilot lights which have much lower instantaneous gas flow than operating appliances and were not switched on and off, the gas use during operation was measured separately for each operating appliance. Each appliance was operated for 10−15 min, allowing a few minutes to reach equilibrium before the measurement. Exhaust gas was measured at a point to where CO_2 was elevated to between ∼400 and ∼20,000 ppm above background, and background air was sampled from within the space providing air to the appliance. Adjusting the sample location of exhaust air allowed the measurement to be accurate (within ∼5−10%) even for low ΔCH_4:ΔCO_2 enhancement ratios while reducing the chance that moisture in the exhaust stream could condense in the sample line. Additional details of the portable analyzer calibration and separate measurements of three tankless water heaters are reported in Supplement S2.

2.3. Statistical Estimation of California Emissions. The measurements of whole-house and operating combustion appliance emissions are extrapolated to state totals using a model that sums statewide homes and their NG usage by appliance types. Because emissions from pilot lights are captured in the whole-house measurements, we separately estimate and then subtract pilot light NG use from NG use by the appliance types before calculating emissions from operating appliances. As described below, both the whole-house emissions and the appliances are measured to have non-Gaussian distributions with a large number of near-zero values and a small number of high values that result in long-tails. To capture the effect of the non-Gaussian distributions, probability distributions (i.e., posterior distributions) are first estimated from the measurements using a Bayesian method (see Supplement S3 for details), and then samples from the inferred posterior distributions using a Markov chain Monte Carlo (MCMC) method are used to generate central estimates and confidence intervals for CH_4 emissions from whole-house and major appliances. Then, state-wide totals for whole-house emissions and combustion appliances, and total residential NG CH_4 emissions, are estimated by resampling the above distributions, with linear additive corrections for smaller appliance types with small estimated emissions.

2.3.1. Estimation of Statewide Whole-House Quiescent Emissions. We estimate statewide house leakage CH_4 emissions by multiplying the inferred whole-house quiescent leakage rate from our measurements by the number of housing units in California. We use the number of housing units from the Population and Housing Estimates for Cities, Counties, and the State data set prepared by California Department of Finance.[13] We use the total number of housing that is categorized as "Occupied". The total number of occupied housing units using natural gas is 12.2 million units for 2016, when a vacancy rate of 7.5% from the CDF data set is applied. This housing total estimate includes both single detached (65%) and multifamily (35%) units. As noted above, the estimate of quiescent whole-house emissions includes emissions from pilot lights, and so we estimate pilot light NG use and their likely contribution to whole-house CH_4 emissions separately as described below.

2.3.2. Estimation of Statewide Emissions from Combustion Appliances. We estimate CH_4 emissions from appliances by combining NG consumption with the ΔCH_4:ΔCO_2 ratio. Detailed NG consumption data are necessary to estimate emissions by appliance types. California total residential NG consumption for 2015 is 401 Gcft or ∼7850 Gg NG/yr.[14] To estimate NG consumption by the appliance type, we applied the relative consumption of NG from the 2009 California residential appliance saturation study[15] (henceforth 2009 RASS) to the 2015 state total NG consumption as well as estimating the fraction of NG consumed by pilot lights. For the pilot light NG consumption, we used RASS data to estimate the fraction of appliances using pilots and combined that with available estimates of NG usage in individual pilot lights for each appliance type. As described in the results, the appliance measurements captured a reasonably large number of water heating and stovetop cooking appliances but fewer space heaters or other appliances that consume small fractions of total residential NG use (e.g., clothes dryers, spas, and hot tubs). Hence, we jointly sample from probability distributions of the ΔCH_4:ΔCO_2 ratio for cooking and water heating using an MCMC method, which results in different central emission estimates (and uncertainty range) than the linear sum of individual results for cooking and water heating. Note that combining samples directly (e.g., joint sampling for the residential total) from the posterior distributions for the individual sectors handles possible correlations between the sector emissions. To obtain total combustion related emissions we also estimate approximate ranges for other NG appliances (space heating and spas/pools) and then sum those linearly with pilot light emissions and the combined MCMC result for water heating and cooking.

2.3.3. Fitting Probability Distributions and Statistical Sampling of Statewide Emissions. To capture the non-Gaussian nature of the observations, we fit the measurements of quiescent house and operating appliance emissions to a long-tailed gamma distribution and compared quantiles of the observed and fit distribution in quantile−quantile (Q−Q) plots using an open-source statistical package.[16] To estimate the central, 5%, and 95% expected values, we apply a Bayesian method combined with an MCMC technique (see Supplement S3 for details). In this work, we set all zero values to an infinitesimal positive definite value. For comparison, we also estimate emissions using a bootstrap method with the simplifying assumption that the measurements are the best available samples for representing the unknown population without a normality assumption.[17] Because the Bayesian method with the MCMC technique sampling the gamma distributions yields larger uncertainty bounds than the

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bootstrapping method, we focus on results from the MCMC method as more conservative.

3. RESULTS AND DISCUSSION

3.1. Distribution of Buildings Selected for Measurement. The houses were recruited across a distribution of locations and building types identified as representative of California's housing stock, with 30 located in northern California and the Central Valley and 45 of them located in southern California and the Central Coast. A map of locations and tables summarizing construction characteristics is provided in Supplement S3 and briefly summarized here. Similar to 2011AHS, roughly 40% of homes were built between 1950 and 1990, with both older and newer homes on either end of the distribution. About half of the homes (55%) have a floor area of 1500−2500 ft^2 (∼140−230 m^2), and 71% are single-story. Similar to 2011AHS, crawlspace and slab are equally common among the sampled homes in northern California/Central Valley, while more houses were slab construction as common for homes in southern California and the Central Coast. In terms of appliances, the homes have 2−7 NG appliances with an average of 4.2. All of the 75 homes have NG water heaters, and all but one use NG for space heating. Storage tank water heaters are the most common ($N = 70$), with the remaining five homes using tankless water heaters. Most homes have central forced air NG furnaces ($N = 72$), while two homes use NG wall furnaces. The majority of the homes use NG cooktops ($N = 64$) and NG clothes dryers ($N = 53$), and about half have NG ovens ($N = 37$).

As part of the house inspection, field technicians detected minor NG leaks (none posing safety concerns) in pipe-fittings near 5 water heaters, 2 NG cooktops, 1 furnace, and 1 oven. As noted above, not all pipes and fittings were accessible, so these results likely represent a lower limit to the actual number of leaks.

3.2. Building Measurements. The methane emissions from the quiescent buildings and combustion appliance measurements from the 75 homes are reported below. In addition, a table combining the measurement results with the results of the field survey completed by measurement technicians is provided as a separate tabular Supplement file.

3.2.1. Distribution of Quiescent Whole-House Emissions. Emissions from quiescent buildings are shown as a histogram in Figure 2, ranging from near-zero (nondetection) to a maximum near 37 gCH_4/day, with median and mean values of 2.1 and 4.6 gCH_4/day, respectively. The distribution of the data is clearly non-Gaussian with a long-tail that will be characterized in the following analysis section. As described in the methods, we removed 10 whole-house measurements where the estimated calibration CH_4 release did not match the known rate to within 2 times the estimated measurement error. Here, we note the difference in the central value and 5 and 95% statistics was indistinguishable with those obtained using all data. As noted above, field technicians inspected pipe fittings near readily accessible house appliances, but we find that whole-house leakage does not vary significantly with the small number of detected pipe leaks. Thus, we suspect the leak testing may underestimate the actual number of pipe leaks in some homes. Whole-house leakage did not vary significantly ($p < 0.1$) with the number of NG appliances for all houses, but houses with emissions greater than 5 gCH_4/day showed a marginally significant ($p = 0.21$) increase with the number of appliances.

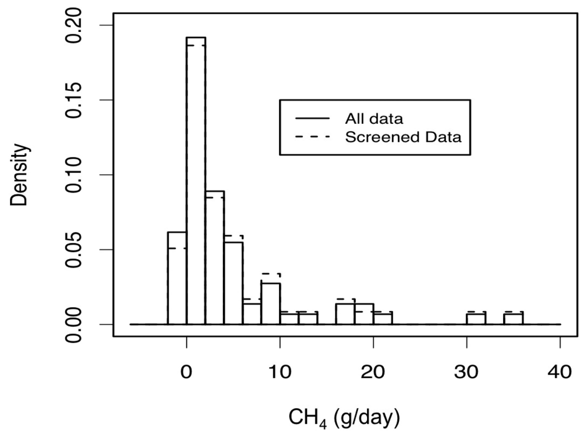

Figure 2. Distribution of measured whole-house quiescent CH_4 emissions (solid line) and the subset of houses screened where measured CH_4 gas addition matched the known value (5 g CH_4/day) to within a factor of 2 times the estimated measurement error (dashed line).

3.2.2. Distribution of Combustion Appliance Emissions. Emissions from steady operation of two combustion appliances were measured in most of the 75 homes. Summary statistics for valid emission measurements by appliance type are shown in Table 1. Less than 1/2 of the measurements (1 of 6 furnaces, 16 of 56 domestic water heaters, and 23 of 51 stovetops) had ΔCH_4:ΔCO_2 enhancements greater than zero as indicated by Ntot and Nzero, respectively. Here, the cases identified as zeros had either no measurable CH_4 enhancement or showed CH_4 depleted in the exhaust gas relative to air supplying the appliance, indicating that the flames consumed part of the CH_4 present in the supply air. All tankless water heaters exhibited ΔCH_4:ΔCO_2 enhancements greater than zero, but with low values ranging from 0.05 to 0.1% (see Supplement S2 for additional results of detailed tankless water heater measurements).

For the cases with positive ΔCH_4:ΔCO_2 enhancement during steady operation, values generally ranged between 0.015% and 0.5%, with a few higher values ranging from 1 to 3% for tank heaters, stovetops, and wall heaters. Furnaces were an exception, with only one nonzero value of 0.03% observed out of six furnaces measured, consistent with a small number of measurements made as part of a previous CEC study.[10] Based on the low values in the small number of furnaces measured, we assume space-heating emissions from forced air furnaces contribute only a small amount of CH_4 in the state-wide analysis described below. For the stovetops and domestic water heaters, we note that there was no significant relationship between the measured ΔCH_4:ΔCO_2 enhancement ratios and appliance age.

Pilot light flames all exhibited measurable ΔCH_4:ΔCO_2 enhancement ratios. Because the number of total pilot light measurements was small, the distributions of water heater and furnace pilot lights cannot be distinguished. Grouping them together yields mean and median ΔCH_4:ΔCO_2 enhancement ratios of 0.059% and 0.065% and a standard deviation of 0.03%, respectively. Based on these results, we include pilot lights as a separate category of combustion appliances and evaluate their importance for California's total residential CH_4 emissions below.

3.3. Statistical Estimation of California Emissions. *3.3.1. Emissions from Quiescent House Leakage Including*

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Table 1. Summary Statistics for Combustion Appliance $\Delta CH_4:\Delta CO_2$ Enhancement Ratios (%)

	min.	1st Qu	median	mean	3rd Qu	max.	Ntot	Nzero
tank WH	0.000	0.000	0.000	0.136	0.100	1.000	62	40
tank WH pilot	0.150	0.400	0.500	0.530	0.800	0.800	5	0
dryer	0.000	0.000	0.035	0.068	0.103	0.200	6	2
furnace	0.000	0.000	0.000	0.005	0.000	0.030	6	5
furnace pilot	0.230	0.515	0.800	0.677	0.900	1.000	4	0
stovetop	0.000	0.000	0.000	0.242	0.100	3.000	54	28
tankless WH	0.050	0.065	0.080	0.077	0.090	0.100	5	0
wall heater	0.000	0.250	0.500	0.500	0.750	1.000	2	1

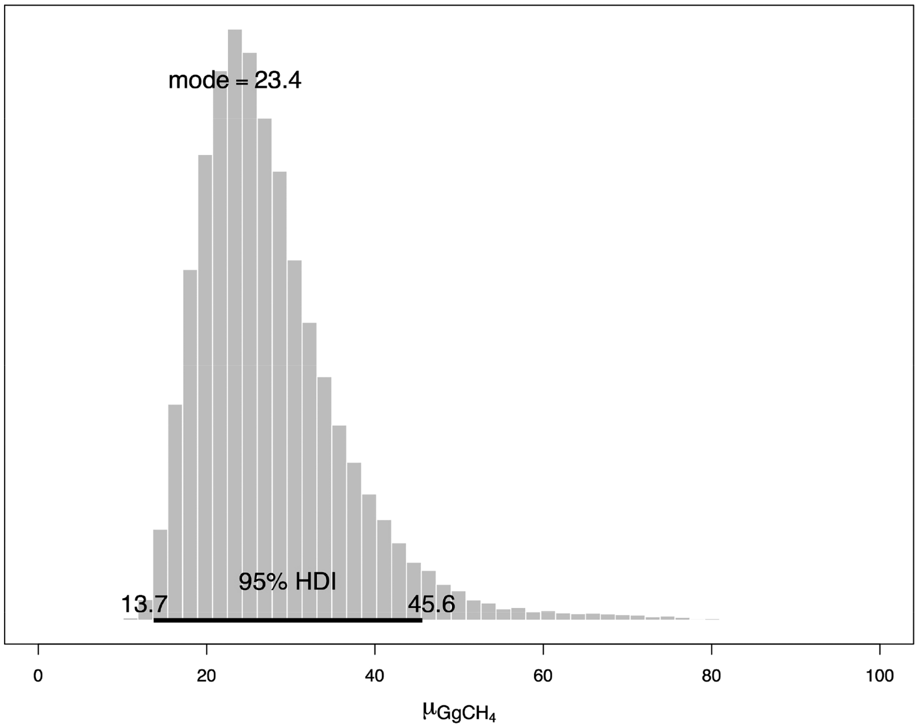

Figure 3. Posterior distribution of California whole-house quiescent leakage (Gg CH_4/yr) including emissions from pipe leaks and pilot lights. Throughout the paper, we report mode values (i.e., the maximum a posteriori probability estimate) for the mean as central estimates.

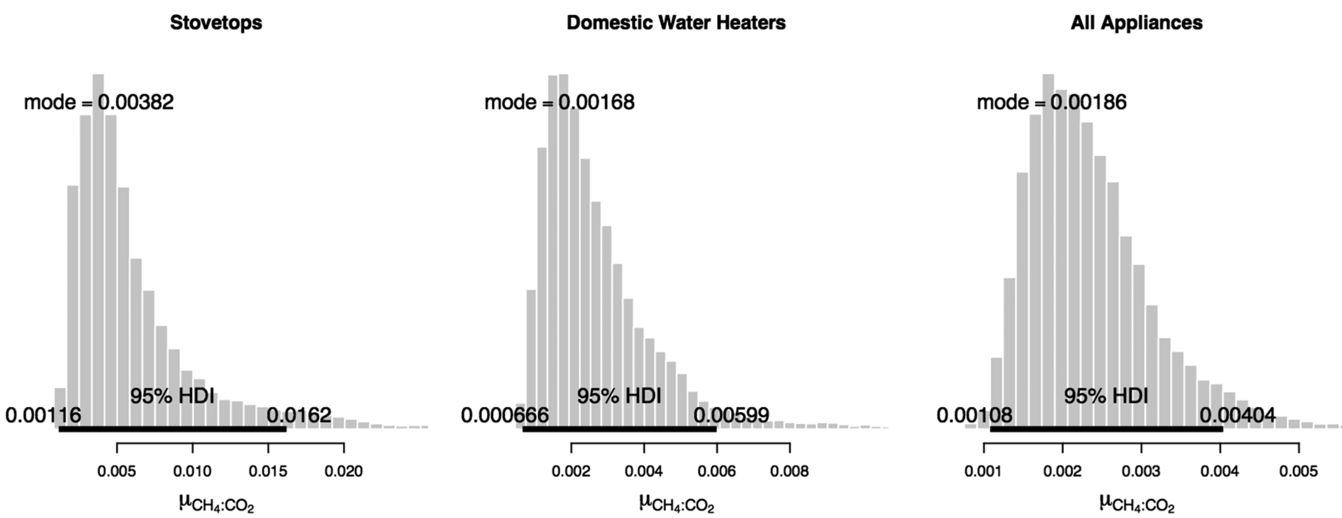

Figure 4. Posterior distributions $\Delta CH_4:\Delta CO_2$ enhancement ratios for operating stovetops, domestic water heaters, and all operating combustion appliances taken together (not including pilot lights).

Pilot Lights. We estimate CH_4 emissions from quiescent house leakage and pilot light emissions in California as the product of the distribution estimated above and the 12.2 million occupied California residences using NG. Figure 3 shows the posterior

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Table 2. Estimated Quiescent CH_4 Emissions from California Homes and Combustion Appliances

estimation type	description	lower CH_4:CO_2 ratio[a] (%)	lower CH_4 emitted (Gg CH_4/yr)	central CH_4: CO_2 ratio[a] (%)	central CH_4 emitted (Gg CH_4/yr)	upper CH_4:CO_2 ratio[a] (%)	upper CH_4 emitted (Gg CH_4/yr)	lower CH_4MCMC (Gg CH_4/yr)	central CH_4MCMC (Gg CH_4/yr)	upper CH_4MCMC (Gg CH_4/yr)
quiescent whole-house	whole-house leakage							13.7	23.4	45.6
appliance combustion	space heating	0.005	0.1	0.014	0.4	0.04	1.1			
	water heating	0.07	2.2	0.205	6.5	0.6	19.1	2.1	5.4	19.1
	cooking	0.11	0.5	0.420	1.7	1.6	6.6	0.5	1.6	6.6
	pool and spa	0.07	0.1	0.205	0.4	0.6	1.3			
	clothes dryer	0.005	0.0	0.032	0.1	0.2	0.5			
MCMC-appliance combustion[b]	water heating + cooking							**3.3**	**7.5**	**22.7**
total MCMC[b]	water heating + cooking + whole-house leakage							**21.3**	**34.6**	**60.6**
minor appliances[c]	space heating + pool/spa + dryer		**0.4**		**1.1**		**3.4**			

[a]Ratios for water and cooking values taken from fitted distributions; others are minimum value greater than zero or max of observed values, with pool and spa assumed the same as heaters for domestic water. [b]MCMC sampling of joint distributions yields estimates that differ from the linear sum over individual distributions. [c]Total emissions reported in the text are estimated by summing minor appliances linearly with MCMC results.

distribution (with summary statistics) for the mean CH_4 emissions from house leakage, estimated using the Bayesian method treating the unknown mean CH_4 emission as a random variable. As shown in Figure 4, the posterior estimate for mean whole-house emissions is 23.4 (13.7−45.6, hereafter 95% confidence) Gg CH_4/yr when only including measurements for houses where the prescribed calibration flow is obtained. This result is not very sensitive to removing data, where emissions estimated using all measurements yield whole-house emissions of 20.9 (12.5−37.5) Gg CH_4/yr, with the slightly smaller confidence interval likely due to including more data. For comparison with the Bayesian method, using the same data directly in a bootstrap method yields a narrower confidence interval of 15.3−31.7 Gg CH_4/yr, and we adopt the Bayesian result as a more conservative estimate.

We estimate the contribution of pilot lights to the whole-house measurements in California as the product of the average number of pilot lights in an average house, the amount of NG consumed by pilot lights, and the fraction of CH_4 emitted unburned from the CH_4:CO_2 enhancement ratio, with details provided in Supplement S6. Using the 2009 RASS data, we estimate that there are approximately 0.82−1.26 pilot lights per house with the majority associated with domestic water heaters. Corresponding NG use for residential appliance pilot lights is assumed to range from 200−400 Btu/h (∼90−180 gCH_4/day) per pilot, depending on the typical size of the burner. From Table 1, the mean ΔCH_4:ΔCO_2 ratio for pilot lights is ∼0.6 ± 0.3%. Combining these factors for each appliance category, we estimate total NG consumed by pilot light emissions is roughly 4.7 (3−10) Gg CH_4/yr, where the uncertainty is assumed due to uncertainty in NG consumed by pilots and the ΔCH_4:ΔCO_2 ratio. This suggests that roughly 25% of the estimated whole-house leakage may be due to pilot lights, though the fraction is quite uncertain. We note that under these assumptions, NG consumption from all pilot lights is ∼740 Gg CH_4 /yr and is subtracted from the NG consumption by appliance class before estimating NG from operating appliances below.

3.3.2. Emissions from Residential Combustion Appliances. Figure 4 shows the posterior distributions for the estimated mean ΔCH_4:ΔCO_2 ratios for operating stovetops and domestic water heaters with tanks (which comprise the majority of the measurements) as well as all appliance types together. Generally speaking, stovetops are found to have roughly double the ΔCH_4:ΔCO_2 ratio of domestic water heaters in steady operation.

Total CH_4 emissions estimated by appliance types are summarized in Table 2. The largest single category is emissions from domestic water heating which total 5.4 (2.1−19.1) Gg CH_4/yr (at 95% confidence). For comparison, emissions from cooking are estimated to be 1.6 (0.5−6.6) Gg CH_4/yr. We note that although the mean ΔCH_4:ΔCO_2 ratio is higher for the stovetops (mode = 0.0038) than for the water heater (mode = 0.0017), the NG usage for the cooking is only ∼14% of that of the water heating. Estimating emissions from joint MCMC sampling of water heating and cooking together yields emissions of 7.5 (3.3−22.7) Gg CH_4/yr. Here, we note that joint sampling for the sum of water heating and cooking does not yield the same result as that from the linear sum of individual sampling results due to non-Gaussian likelihood distributions and sampling uncertainty (inherent in working with samples).

The other appliance types are estimated to have comparatively much smaller emissions (furnaces, spas, etc.). Here, we use the lower 25% and upper 75% estimates for ΔCH_4:ΔCO_2 ratio together with gas consumption to estimate the central value as the geometric mean of the lower and upper estimates. For example, this results in estimated emissions of 0.4 (0.04−1.1) Gg CH_4/yr for space heating. Here, we also note that in areas where a significant fraction of space heating is done with inefficient heaters (e.g., wall furnaces), these emissions will likely be higher. Emissions from spa/hot tubs and clothes dryers are estimated to contribute small but uncertain amounts to the combustion related emissions. Lacking better information, we sum emissions for these classes linearly with a total estimate of 1.1 (0.4−3.4) Gg CH_4/yr for space heating, pools and spas, and clothes dryers together (see Table 2).

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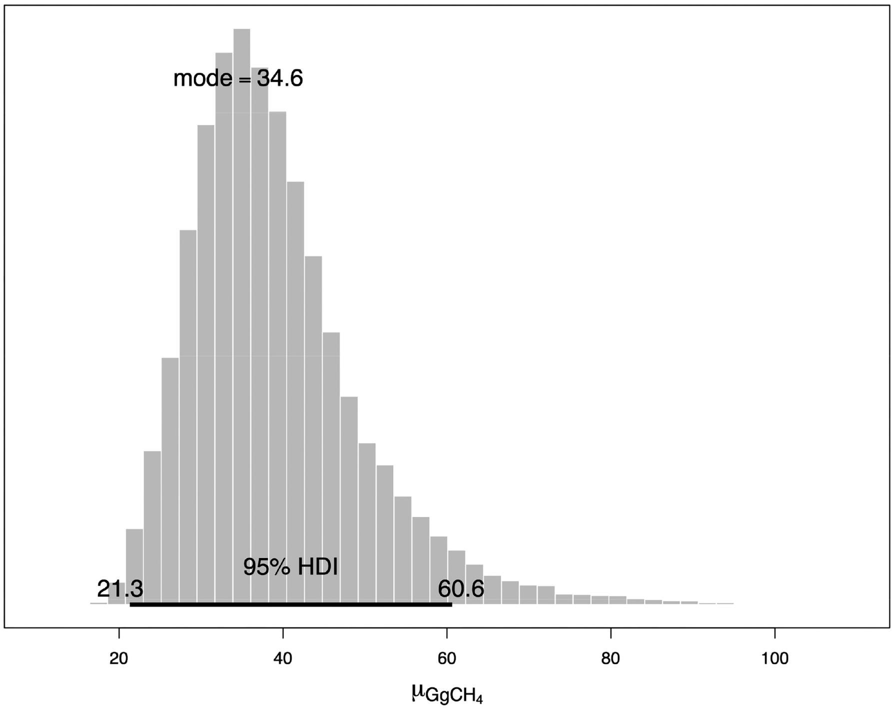

Figure 5. Posterior distribution of total California residential CH_4 emissions (Gg/yr) combining whole-house quiescent leakage, water heating, and cooking.

4. DISCUSSION

Methane emissions from California residences are estimated for the combination of quiescent house leakage and operating combustion appliances combining MCMC emission samples from these two sectors (Figure 5). Including the additional emissions from minor appliances, total CH_4 emissions from residential sector NG consumption are 35.7 (21.7−64.0) Gg CH_4/yr (and 0.9 (0.5−1.6) Tg CO_2eq, using the global warming potential of 25 gCO_2eq/gCH_4 adopted by the CARB GHG inventory), equivalent to 0.5% (0.3−0.9%) of residential consumption. This is equivalent to roughly 15% of the estimated California inventory for NG related CH_4 emissions (6.4 Tg CO_2eq) and 2% of total inventory CH_4 emissions (39.6 Tg CO_2eq) in 2015 (CARB, 2017). In terms of cost to consumers, if 0.5% of California's residential NG gas consumption is emitted at an average price of ∼$12/Mcft in 2015, the economic value of lost gas is approximately $30 million/yr that could be applied to reducing sources of postmeter CH_4 emissions.

Comparing these results with atmospheric studies, work in the San Francisco Bay Area found between 0.3−0.5% (95% confidence interval) of NG CH_4 delivered to customers is emitted to the atmosphere,[5] which is nominally consistent with the residential estimate if before-meter distribution leakage is comparatively small and/or the emitted fraction of NG used in other sectors (e.g, commercial buildings, and industrial activities) is smaller than that for the residential sector. A different atmospheric study of Los Angeles, NG CH_4 emissions of 1.6 ± 0.5% of gas delivered,[4] suggested postmeter residential emissions are unlikely to dominate CH_4 emissions in that area. Last, results from an atmospheric study of Boston[3] found emissions of 2.7 ± 0.6%, which is nearly 5 times larger than our residential estimate, suggesting premeter leaks in the distribution system dominate or that results obtained in California underestimate emissions in Boston due to differences in some combination of climate, housing type, or equipment.

Summing linearly across all aspects of combustion appliances, CH_4 emissions from major operating appliances (7.5 (3.3−22.7) Gg CH_4 /yr), minor appliances (1.1 (0.3−4.4 Gg CH_4 /yr), and pilot lights (4.7 (3−10) Gg CH_4 /yr) yield 13.3 (6.6−37.1) Gg CH_4 /yr, which is roughly equivalent to 0.17 (0.08−0.47)% of total gas consumed. Converting combustion related CH_4 emissions to 100-yr CO_2 equivalent units we note the estimate of 0.33 (0.15−0.89) Tg CO_2eq is more than an order of magnitude larger than residential natural gas combustion emissions (0.01 Tg CO_2eq) reported in the 2015 state GHG inventory.[2] Here, nearly 30% of the total appliance emissions are estimated from pilot lights, suggesting a value in moving toward electronic ignitions. Last, we note that appliance emissions may be larger than the steady state measurements reported for 75 homes because of emission transients during burner startup and shutdown as found in the separate measurements of tankless water heaters. This suggests that future work should include measurement of transient emissions across a sample of appliance types and manufacturers should consider designing new products that minimize CH_4 emissions during startup and shutdown.

These findings suggest that CH_4 emissions from residential buildings can be reduced not only through a combination of inspection and repair of gas leaks, particularly regular checks for unlit pilot flames, but also leak testing readily accessible pipe-fittings (e.g., at point of sale or during energy retrofits) and improved ignition and combustion efficiency for gas appliances. In the longer term, while CH_4 emissions from houses are small compared to most other sources of anthropogenic CH_4, California's ambitious climate goals (e.g., 80% reduction by 2050) suggest value in promoting a transition to renewable nonfossil energy sources and high-

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efficiency technologies (e.g., heat pumps, induction heating) for residential water and space heating and cooking.[18,19]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b03217.

S1, summary of California housing stock; S2, detailed measurement methods; S3, Bayesian and bootstrap statistical sampling methods; S4, building characteristics of study homes; S5, probability distributions for whole-house quiescent emissions and appliance ΔCH_4:ΔCO_2 ratios; S6, estimation of pilot light gas use and CH_4 emissions (PDF)

Measurement results (XLSX)

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Notes

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