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Article

Methane and NO_x Emissions from Natural Gas Stoves, Cooktops, and Ovens in Residential Homes

Eric D. Lebel,* Colin J. Finnegan, Zutao Ouyang, and Robert B. Jackson

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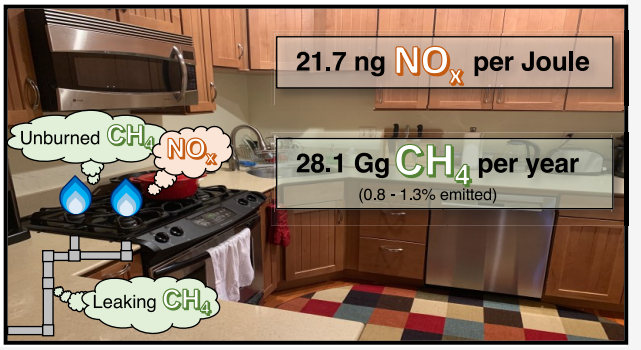

ABSTRACT: Natural gas stoves in >40 million U.S. residences release methane (CH_4)—a potent greenhouse gas—through post-meter leaks and incomplete combustion. We quantified methane released in 53 homes during all phases of stove use: steady-state-off (appliance not in use), steady-state-on (during combustion), and transitory periods of ignition and extinguishment. We estimated that natural gas stoves emit 0.8−1.3% of the gas they use as unburned methane and that total U.S. stove emissions are 28.1 [95% confidence interval: 18.5, 41.2] Gg CH_4 $year^{-1}$. More than three-quarters of methane emissions we measured originated during steady-state-off. Using a 20-year timeframe for methane, annual methane emissions from all gas stoves in U.S. homes have a climate impact comparable to the annual carbon dioxide emissions of 500 000 cars. In addition to methane emissions, co-emitted health-damaging air pollutants such as nitrogen oxides (NO_x) are released into home air and can trigger respiratory diseases. In 32 homes, we measured NO_x (NO and NO_2) emissions and found them to be linearly related to the amount of natural gas burned ($r^2 = 0.76$; $p \ll 0.01$). Emissions averaged 21.7 [20.5, 22.9] ng NO_x J^{-1}, comprised of 7.8 [7.1, 8.4] ng NO_2 J^{-1} and 14.0 [12.8, 15.1] ng NO J^{-1}. Our data suggest that families who don't use their range hoods or who have poor ventilation can surpass the 1-h national standard of NO_2 (100 ppb) within a few minutes of stove usage, particularly in smaller kitchens.

KEYWORDS: *cooking, climate, combustion, pollutants, post-meter emissions, appliances, houses*

INTRODUCTION

Since 1750, methane (CH_4) has contributed about one-quarter of the world's radiative forcing,[1] and its concentration continues to rise.[2] Methane is a shorter-lived gas than carbon dioxide (CO_2) but is nevertheless 34−86 times more potent than carbon dioxide on 100 and 20 year timescales, respectively.[3]

Leaks of natural gas (>90% methane) across the ∼3 million miles of pipeline in the United States' supply chain[4] have been studied extensively.[5] However, comparatively little work has been done on emissions inside homes and buildings, so-called "post-meter" emissions. The United States Environmental Protection Agency (USEPA) Inventory of U.S. Greenhouse Gas Emissions and Sinks only documents leaks from residential gas meters and incomplete combustion from residential gas appliances, but more information is needed on methane leaks from transitory on−off phases and quiescent steady-state-off emissions, particularly from gas stoves.[6] Previous work found that appliances in Boston and Indianapolis—excluding pilot light emissions—were estimated to emit 0.038% of the natural gas they consumed.[7] A separate study in California found that whole-home post-meter residential emissions totaled 0.5% of residential natural gas consumption in the state.[8]

Natural gas is a popular fuel choice for home cooking. Nationally, over one-third of households (>40 million homes) cook with gas.[9] In some states, the proportion is substantially higher; over 60% of households in California cook with gas, for instance.[10] People interact more directly with their stove than with other gas appliances, increasing potential exposure to any

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natural gas constituents and compounds formed during combustion, including formaldehyde (CH_2O) carbon monoxide (CO), and nitrogen oxides (NO_x). Here, we define NO_x as the sum of nitric oxide (NO) and nitrogen dioxide (NO_2). Among all gas appliances, the stove is unique in that the byproducts of combustion are emitted directly into home air with no requirement for venting the exhaust outdoors. In fact, some kitchens have "ductless" hoods that recirculate fumes through activated charcoal filters, which are generally less effective at cleaning the air.[11] Vented hoods have a range of effectiveness and function best when overhanging the stove.[12,13] Because exhaust hoods are separate from the stove and must be operated manually, vented hoods in practice are used only 25−40% of the time.[14−16]

To date, the majority of research concerning gas stoves has focused on their effect on indoor air quality.[17,18] In particular, research has measured long-term averaged indoor concentrations of NO_x and CO,[19,20] with limited direct measurements of emission rates. NO_2 is a respiratory irritant and can lead to asthma, coughing, wheezing, and difficulty breathing, occasionally resulting in hospitalization.[21,22] Chemically, NO forms NO_2 indoors upon interaction with ozone (O_3), and NO_2 concentrations will decrease through interactions with interior surfaces.[23] Concentrations of NO_x and other indoor pollutants depend greatly on the turnover rate of air in homes; over time, outside air will enter the house through ventilation and infiltration, reducing any gradient between indoor and outdoor concentrations.

There are currently no U.S.-based indoor exposure guidelines or standards for NO_2; however, Canada has a maximum residential exposure limit of 90 parts per billion (ppb) over a 1 h exposure and 11 ppb over the long term (>24 h).[24] In 2010 the EPA set a 1 h outdoor exposure limit of 100 ppb for NO_2 which was retained in 2018; the annual standard of 53 ppb was established in 1971 and has not been updated.[25] Recently, in 2021, the WHO set the outdoor standard for NO_2 to be approximately 13 ppb over 24 h.[26] NO_2 concentrations of 100 ppb are regularly exceeded if stoves are used without adequate indoor ventilation, either due to a low capture efficiency of the exhaust hood or the hood not being activated by the user.[17]

We measured methane emissions from stoves during all phases of operation (steady-state-off, turning on and off, and during combustion), with additional measurements quantifying NO_x emissions during combustion. We used a kitchen chamber approach to take measurements with a measured kitchen volume, sealed using a plastic partition. For cooktops, we focused our measurements on emissions from individual burners at different intensities to gain a fuller picture of total emissions. We quantified steady-state-off emissions from stoves because they were not included in most previous cooktop emissions studies and because, previously, we found steady-state-off emissions to be a substantial, sometimes dominant, component of total methane emissions from storage water heaters.[27]

MATERIALS AND METHODS

We define a "cooktop" as a flat surface with individual burners, typically four or five per stove, and an occasional "griddle" element. The "oven" is an enclosed heated space, usually containing two burners: a "bake" burner below the bottom steel plate and a "broil" burner typically exposed and affixed to the oven's roof. A "stove" (also referred to as a "range") is a freestanding unit that contains both a cooktop and an oven, typically both using gas, although some combine a gas cooktop with an electric oven.

Site Selection. We measured methane emissions from stoves in 53 homes in 7 California counties between January 2020 and May 2021 (Table S1). Our sample set included private homes, properties for sale or rent by real estate agents, and, because of the constraints of COVID-19, Airbnb rentals. We measured methane emissions from gas cooktops (18 unique brands) at all 53 homes and tested gas ovens and broilers in 40 and 31 homes, respectively. For stoves with information available, we found the ages of stoves ranged between 3 and 30 years. Cooktop burners ranged in heat output from 4500 to 25 000 British thermal units (BTU) h^{-1}, and most oven burners ranged between 16 000 and 19 000 BTU h^{-1}, as per product specifications.

We additionally measured NO, NO_2, and NO_x emissions from 32 cooktops and 24 ovens of the stoves tested for methane emissions (Table S1). The NO, NO_2, and NO_x emission rates are important because—coupled with circulation, ventilation, infiltration, and room size—they determine the concentration of NO_x pollutants that build up in kitchens.

Sampling Overview. We measured each stove burner using a static flux chamber ("room chamber") method (Figure S1), which we tested in the lab and in homes before initiating the study. The stove was contained in an airtight portion of the room of known volume (average of 20 m^3, range of 8−83 m^3, Figure S2) by hanging plastic sheets (4−6 mil thickness) to partition the kitchen from surrounding space using a ZipWall Dust Barrier System (zipwall.com) with 8 or 10 ft tension poles. Clear plastic sheets were sealed along the ceiling, walls, and floor using a combination of ZipWall foam bars, painter's tape, and/or 3 kg sandbags. We used one or two Lasko 3300 Wind Machine Fans on low or medium to circulate the air within the space. We took care in selecting fan speed, position, and direction to minimize any disruptions to burners (Figure S1). An additional fan or two circulated the air outside the chamber to ensure that the "background air" was homogenous. We collected concentration measurements of methane (CH_4), ethane (C_2H_6), and CO_2 at a rate of <1 Hz on a Picarro cavity ring-down spectrometer (model G-2210i). In addition, we used one of two analyzers to measure concentrations of NO_x, including direct measurements of NO and NO_2: a Teledyne model 200A chemiluminescence analyzer and a Thermo Fischer Scientific 42iQ analyzer. Kitchen temperature was recorded using an Onset HOBO Logger (UX100-23A).

General Sampling Scheme. After partitioning the kitchen with plastic sheets, we set up two inlet hoses to the analyzers, one near the center of the enclosed kitchen area about 4−6 ft off the ground and a second in a space outside the enclosed area at a similar height for background measurements. We took background measurements of methane and NO_x for at least 2 min before collecting stove measurements.

To test the size and seal quality of the room chamber, we briefly entered the enclosed kitchen through a zipper door and released a 300−500 mL aliquot of ethane with a glass syringe ("ethane inject") to estimate the room volume by dilution. We found this method to be more straightforward for estimating kitchen volume than taking measurements of chamber dimensions, which proved challenging with cabinetry and nonstandard configurations of many modern kitchens, although both methods agreed well (Figure S2). Ethane was chosen for the dilution measurements because the Picarro measured it and there were no additional sources of ethane in

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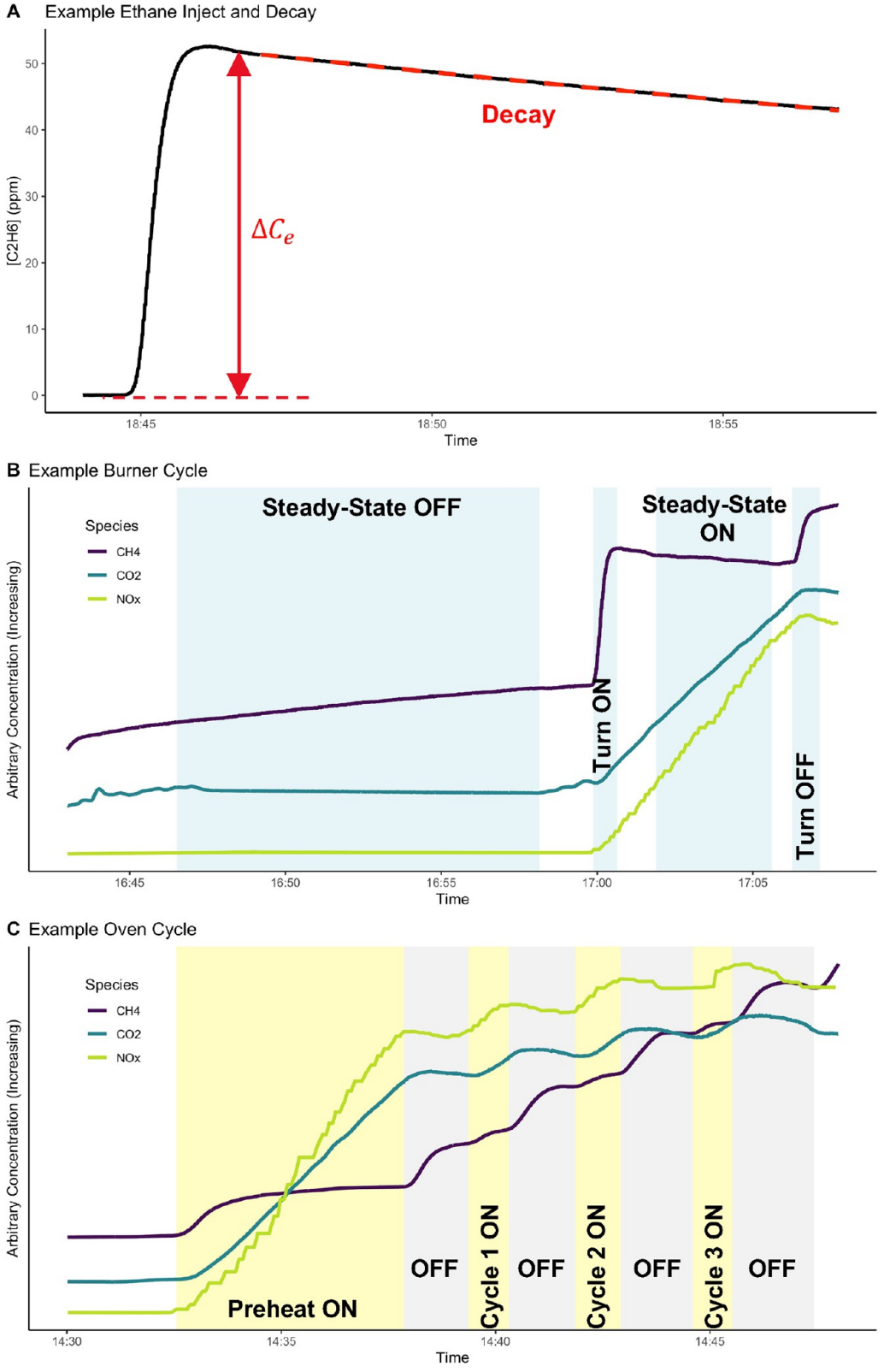

Figure 1. Example concentration plots for various measurement events using data from actual measurements in our study. (A) Ethane injection and decay: first, we injected 500 mL of ethane into the space created by the plastic wall to determine its volume based on dilution. ΔC_e is the change in ethane concentration, which resulted from our injection of ethane. Then, we allowed the elevated ethane to exponentially decay over time (dotted red line) to measure the air changes per hour (ACH) in the space created by the chamber. This value was used to correct CH_4, NO_x, and CO_2 values measured during other test phases. (B) Cooktop burner measurement cycle: "steady-state-off" before the burner is ignited, "turn on" for the pulse of gas emitted when the burner is ignited, "steady-state-on" for the emissions while the burner is on a steady state, and "turn off" for the pulse of emissions observed when the burner is extinguished; (C) oven measurement cycle: "preheat" for the increase in concentrations while the oven is reaching the set temperature and "cycling" for the on/off behavior of the oven while it maintains the set temperature.

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the kitchen while the stove was off beyond small emissions of ethane co-emitted in the natural gas from the stove or nearby pipes and fittings, which were at most 2 orders of magnitude lower than the amount of ethane we injected (Figure 1A; details and calculations in the Supporting Information (SI)). After leaving the kitchen, we measured the "ethane decay", which we used to calculate the diffusion and exchange of air outside the partitioned space through unavoidable, small gaps in the plastic and through leaky doors, windows, and vents (Figure 1A).

Simultaneously with ethane decay, "steady-state-off" measurements of methane from the stove were taken for 5−10 min (Figure 1B). Emissions from steady-state-off are not attributable to a specific source within the stove or piping but capture all emissions from the stove and piping within the chamber volume. Ethane injection/decay and steady-state-off measurements were repeated throughout sampling for a total of two to seven independent measurements of volume, air exchange, and steady-state-off methane emissions per stove.

To begin combustion measurements of methane and NO_x (NO and NO_2), we entered the kitchen and ignited one burner on the cooktop, which initiated the "on pulse". Because NO_x is a byproduct of combustion, we never observed pulses of NO_x while turning burners on or off. We left the burner on, and after the pulse was completely mixed in the room, typically 90 s after ignition, we exited the kitchen and collected a "steady-state-on" measurement of both methane and NO_x for at least 3 min while vacant. We then returned inside, waited 30 s, and turned off the burner (Figure 1B). After 90 s, another single burner was lit. We measured burners individually on some combination of low, medium, or high for both methane and NO_x emissions. We aired out the kitchen chamber volume whenever the concentration of methane inside approached the limit of the Picarro (~30 ppm CH_4). Details of the calculations, including correction for the measured air exchange rate, are presented in the SI.

We tested ovens similarly for methane and NO_x emissions using our kitchen chamber system. We used the oven's bake setting to set the temperature between 350 and 425 °F. We observed a distinct "preheat" phase of emissions as the oven warmed to temperature and a "cycling" phase as the oven maintained temperature, which we saw in the CH_4, CO_2, and NO_x data output (Figure 1C). We allowed the bake burner to preheat the oven and cycle at least three times, taking ~20−30 min. We tested broiler burners using the same method but only while preheating.

Scaling Methane Emissions. We scaled our appliance measurements to national estimates of total emissions using activity data from Chan et al.[28] and Zhao et al.,[18] assuming that stove usage does not vary substantively across the United States with climate the way that furnace usage does (Figure S7). Chan et al. and Zhao et al. report activity data from 80 residences, collectively, including the number and length of time that the occupants used individual burners on their stove and how often they used their ovens. We also obtained information on the numbers of appliances from the 2015 Residential Energy Consumption Survey.[9] The total number of appliances was assumed to be the total number of stoves and any secondary cooktops, 43.4 million appliances (standard deviation: 1.3 million) nationally.

Using Chan et al., Zhao et al., 2015 RECS, and our measurements, we created an "expanded dataset" with one observation for every possible combination of stove i (n = 53) and usage datapoint j (n = 80), creating 4240 unique observations of annual stove emissions. From here, we scaled our measurements using the number of appliances (43.3 million) reported in the RECS database for the United States.

For each pairing of stove i and usage datapoint j, we divided the time based on the selected usage environment into steady-state-off, steady-state-on, and oven usage periods, broken down into preheat and cycling phases. We calculated the total methane emissions that would be expected in a day given the selected stove's emissions for the total length of these periods and the number of expected pulses using eq 1

$$\begin{aligned}\text{emissions (g day}^{-1}) &= \text{SS. On}_i\ (\text{g h}^{-1}) \times \text{Hours. Cooktop}_j + \text{Oven}_i\ (\text{g h}^{-1}) \\ &\times \text{Hours. Oven}_j + \text{SS. Off}_i\ (\text{g h}^{-1}) \\ &\times (24 - \text{Hours. Cooktop}_j - \text{Hours. Oven}_j) \\ &+ \text{On/Off Pulse}_i\ (\text{g}) \times \text{Burner. Ignitions}_j \end{aligned} \quad (1)$$

with emissions in total grams CH_4 per day, SS.On$_i$ is the emission rate (g h^{-1}) during steady-state-on for stove i; SS.Off$_i$ is the emission rate (g h^{-1}) during steady-state-off for stove i; Oven$_i$ is the emission rate (g h^{-1}) for an oven set to 350−425 °F from kitchen i; Pulse$_i$ is the emission magnitude from each on/off pulse for stove i; Hours.Cooktop$_j$ is the total number of hours per day any burner from cooktop j was used, as reported by Chan et al. and Zhao et al.; Hours.Oven$_j$ is the total number of hours per day oven j was used; and Burner.Ignitions$_j$ is the total number of times a burner was ignited per day from stove j.

We calculated the mean estimate for a stove (burners plus oven) as the mean of the expanded dataset and the 95% confidence interval from a bootstrap using 25 000 replicates and sample sizes of 53 to capture the full uncertainty associated with our stove emissions dataset.[29,30]

Total Volumetric Emissions. In total, previous inventories report that natural gas residential indoor cooking appliances use 113 billion cubic feet (bcf) of gas per year in the United States, the third most gas from residential appliances, after space heating (2677 bcf year^{-1}) and water heating (1019 bcf year^{-1}).[9] We estimated the total percentage of gas emitted using eq 2

$$\%_{\text{leaked}} = \frac{\text{Vol}_{\text{CH}_4}}{\text{Vol}_{\text{NG}}} \quad (2)$$

where Vol$_{CH_4}$ is the amount of methane emitted by the stoves and Vol$_{NG}$ is the volume of natural gas used by the stoves.

We estimated the annual volume of natural gas used by stoves with two different methods: (1) using the natural gas reported by RECS and (2) calculating the natural gas emissions using our recorded CO_2 data combined with the usage data. For both methods, we used our estimate of total emissions from stoves as the numerator in eq 2. In the first method, we used 113 billion cubic feet, provided by RECS. In the second method, we calculated the volume of natural gas using our direct CO_2 emissions and using the first two terms of eq 1 (steady-state-on and ovens) to calculate the assumed total emissions given the usage patterns and total number of national appliances.[9,18,28]

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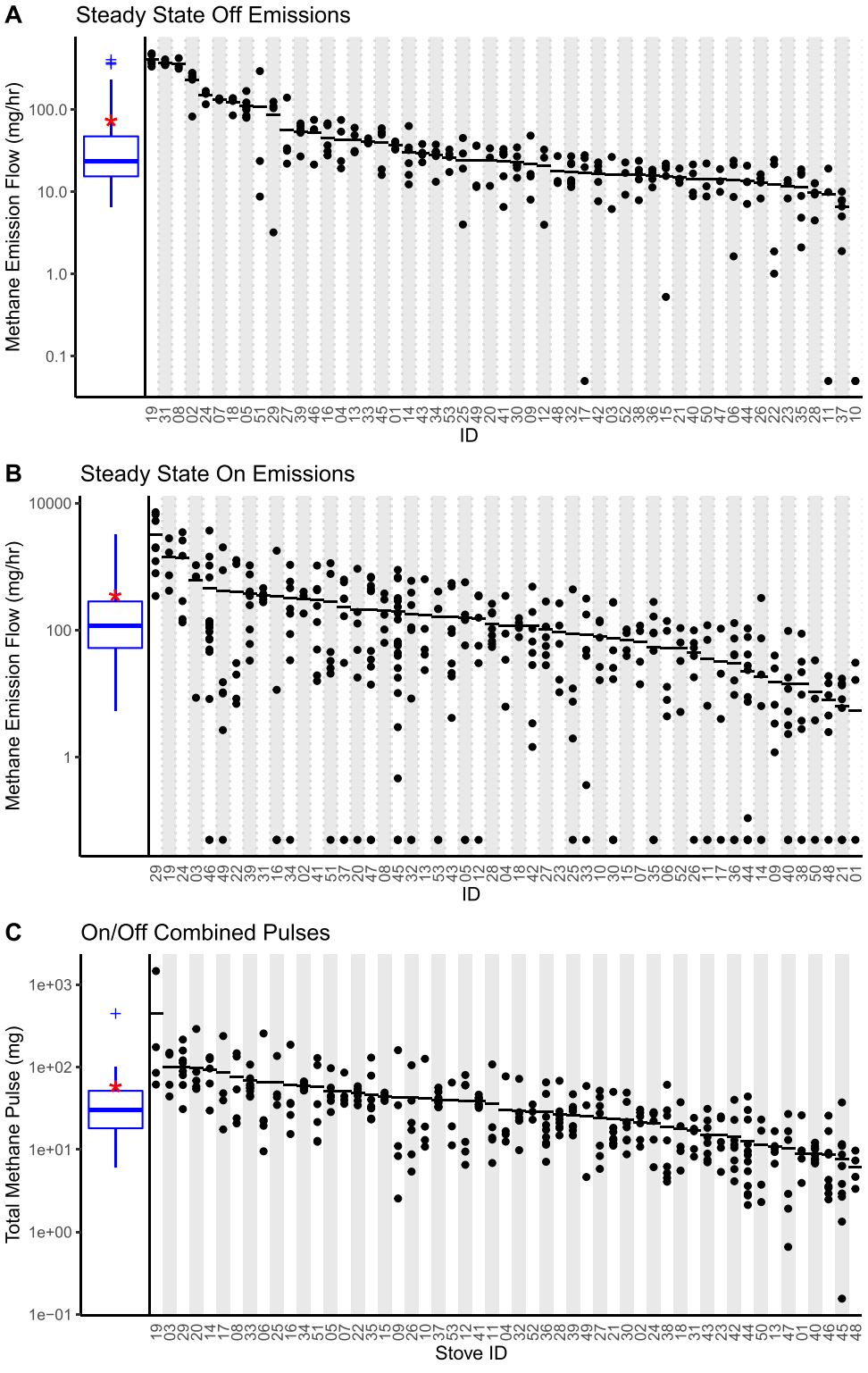

Figure 2. Stove emission components from each individual stove, represented on a log scale, from steady-state-off, steady-state-on, and the on/off pulses. The blue box-and-whisker plot on the left in each panel summarizes each component of stove emissions. The median, 25th, and 75th quartiles are shown, with blue + symbols denoting outliers. The red * represents the mean. On the right side of each plot, every "bin" above a stove ID represents either one measurement of steady-state-off as shown in (A) or a measurement of one burner's emissions, as shown in (B) and (C). The small black horizontal line represents the average for each stove. (A) All steady-state-off emissions are reported for each stove that we measured—most stoves were measured three to five times. The mean (58 mg CH_4 h^{-1}) was higher than the median (24 mg CH_4 h^{-1}). Negative values were set to 0.05 mg CH_4 h^{-1}. (B) Average steady-state-on emission was 259 mg CH_4 h^{-1}, which is slightly higher than the median (119 mg CH_4 h^{-1}). Negative values were set to 0.05 mg CH_4 h^{-1}. (C) Collective on/off pulses are represented by the individual black dots; one dot is the sum of the on-and-off pulse (if any) for an individual burner. The mean (46 mg CH_4) is again higher than the median (30 mg CH_4).

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RESULTS AND DISCUSSION

Stove Steady-State-Off Emissions. During steady-state-off measurements, we found that stoves emitted 57.9 [95% CI from bootstrapping: 36.3, 84.0] mg CH_4 h^{-1} on average (Figure 2A). The data for steady-state-off measurements were long-tail skewed, with the top 5 stoves (9% of sampled units) emitting half (49%) of all steady-state-off emissions. Interestingly, all but four stoves had steady-state-off emissions greater than 10 mg CH_4 h^{-1}, suggesting that most stoves and associated nearby piping leak some methane continuously. We never measured any NO_x emissions while the burners were off because NO_x is formed from the oxidation of nitrogen in the high temperatures of combustion.

Cooktop Steady-State-On Emissions. Total steady-state-on methane emission rates (per unit time) from a single cooktop burner during combustion, averaged over all measured burner sizes and intensities, were 259 [95% CI: 151, 408] mg CH_4 h^{-1}, approximately 4.5× more than steady-state-off emission rates from the entire stove when off (Figure 2B). More methane is therefore emitted instantaneously during combustion than when the cooktop is idle, although the length of time during the day when burners are idle is substantially longer in homes, where emissions occur continuously from steady-state-off leaks. Steady-state-on emission measurements were also long-tailed: the top 5 stoves (9%) emitted 51% of all steady-state-on emissions. Methane emission rates while on were not correlated with burner intensity or CO_2 emission rates (Figures S4 and S5).

During steady-state-on, we also estimated that volumetric-based total NO_x emissions were directly proportional ($p \ll 0.01$; $r^2 = 0.76$) to the rate of natural gas combustion (Figure 3). Therefore, we measured the energy output from the burner using the flow rate of CO_2 to calculate the Joules (J) of energy usage. Average emissions from stoves during steady-state-on ranged between 13.9 and 28.3 ng NO_x J^{-1} (1.5−3.0 g NO_x $Therm^{-1}$), with an average value of 21.7 [20.5, 22.9] ng NO_x J^{-1}. The NO_x emission rate was comprised of 7.8 [7.1, 8.4] ng NO_2 J^{-1} and 14.0 [12.8, 15.1] ng NO J^{-1} (Figure 4B). Volumetrically, emissions were 15.8, 63.2, and 151.5 mL NO_x h^{-1} for burners on low, medium, and high, respectively (Figure 4A), further demonstrating that more NO_x is emitted from burners during higher intensity usage.

Cooktop On/Off Pulse Emissions. Methane emissions from one combined on/off pulse (extra gas emitted while igniting and extinguishing a burner) from a single burner averaged 45.9 [95% CI: 33.1, 64.8] mg CH_4 (Figure 2C). This on/off methane pulse equaled the amount of gas emitted during ∼10 min of average steady-state combustion.

Similar to the steady-state emissions, these on/off pulses exhibited a long-tail distribution driven predominantly by a few cooktops—even individual burners within cooktops—that emitted most of the methane. The highest emitters were the cooktops that ignited using a pilot light (total of eight burners); these burners had an average on/off pulse of 258 mg CH_4. One particularly large outlier—stove 19 (Figure 2C)—emitted on average 450 mg per pulse, nearly 4× higher than the second-highest emitter. This particular stove was one of the two stoves with a pilot light; on this specific unit, some burners took several seconds to ignite, while burners on other units with pilot lights took less time to ignite (thus releasing less methane), suggesting that delays in pilot light ignition may drive some emissions from on/off pulses.

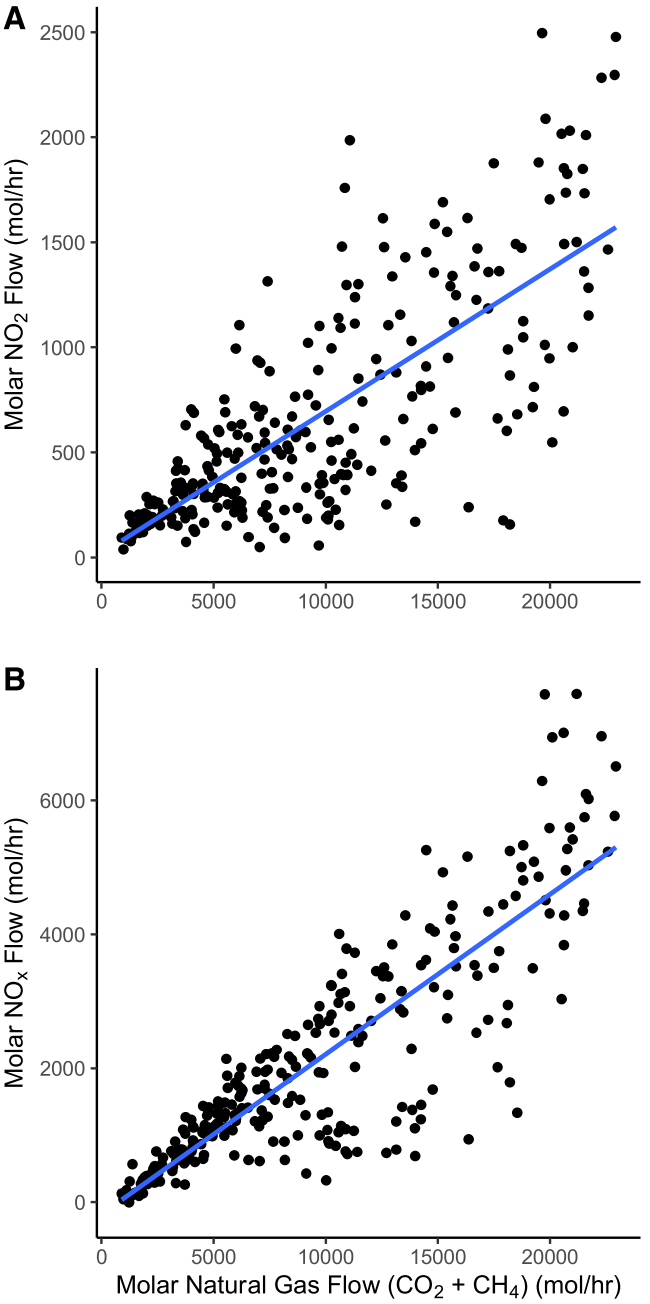

Figure 3. NO_2 and NO_x flows are linearly proportional to natural gas flow. Each black dot represents a measurement of steady-state-on, either for a cooktop burner, or for an oven broil or bake burner. (A) Emissions of NO_2 are plotted on the *y*-axis with the natural gas flow (i.e., emissions of CO_2 + CH_4) plotted on the *x*-axis. There is a clear correlation between the natural gas flow (estimated by summing the measured methane and carbon dioxide flow rates) and the NO_2 flow rate ($p \ll 0.01$; $r^2 = 0.59$). (B) Similarly, for NO_x (i.e., NO + NO_2), there is a correlation between natural gas flow and the NO_x flow rate ($p \ll 0.01$; $r^2 = 0.76$).

In contrast, 180 burners that were lit with a built-in operational electronic sparker emitted less methane, 38 mg CH_4. We also lit 22 burners with a match or lighter because their built-in electronic sparker did not work (but homeowners said they used them nonetheless). For these, we held a lit match next to the burner before turning the gas on. On average, these burners emitted 48 mg CH_4 per on/off pulse, similar to the burners with a functioning electric sparker. We

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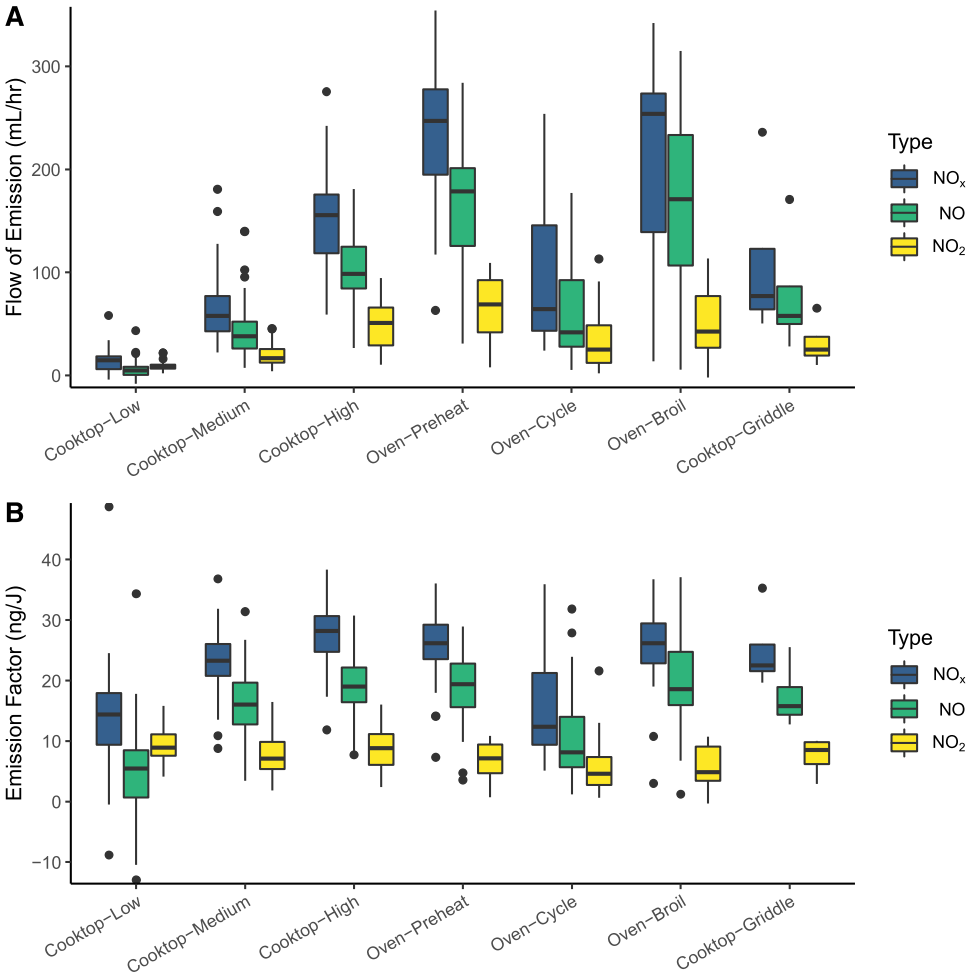

Figure 4. NO_x, NO, and NO_2 total emissions and normalized by energy output. NO_x emissions are shown in total emissions for various burner sizes and intensities and by constituents: NO and NO_2. We separate emissions into various phases of stove operation: cooktop emissions while burners are on low, medium, and high; oven emissions during preheat, cycling (maintaining temperature), and from the oven's broil burner; and emissions from those cooktops that had a griddle element. (A) Emissions are reported as a flow, in mL h^{-1}. These emissions were directly measured using our chamber-based approach, with concentrations measured on the real-time analyzer. (B) The emissions are normalized for the energy output of the burner, reported in Joules (J), and calculated from the total CO_2 emissions we simultaneously measured from each burner. The molecular weight of NO_x was assumed to be 46 g mol^{-1}, as is convention. For direct comparison, emission factors of NO also used 46 g mol^{-1} as the molecular weight.

analyzed these samples in our overall on/off pulse estimate because ignition with the match or lighter simulated "typical operation" for these burners according to the homeowners.

Oven Emissions. We measured the baking (main) burner for 40 ovens and found that during the preheat phase (from room temperature to the setpoint between 350 and 425 °F), ovens emitted on average 117 [95% CI: 74, 178] mg CH_4 over a period of about 13 min, with an average rate of 663 [408, 1030] mg CH_4 h^{-1}. Once the ovens reached preheat temperature, they "cycled", where the burner periodically ignited and extinguished to maintain the setpoint temperature. During this period, it emitted on average 759 [435, 1310] mg CH_4 h^{-1}. In comparison, the broiler burner only emitted 112 [50, 186] mg CH_4 h^{-1} while on. Unlike the bake burner, broil burners usually stayed lit and cycled on and off less often.

We also measured NO_x emissions from ovens and calculated them to be 25 ng NO_x J^{-1}. Volumetrically, this is equal to 232 mL NO_x h^{-1} (broken down into 163 mL NO h^{-1} and 69 mL NO_2 h^{-1}) during the preheat phase for the bake burner set to 350−425 °F, 92 mL NO_x h^{-1} (58 mL NO h^{-1} and 34 mL NO_2 h^{-1}) for the bake burner when it was in cycling mode, and 215 mL NO_x h^{-1} (162 mL NO h^{-1} and 53 mL NO_2 h^{-1}) for the broil burners. We found that ovens could produce enough NO_2 to exceed the 1-h ambient standard (100 ppb) within a few minutes (Figure S11). We also found that NO_2 and NO_x emissions were correlated with the size of the burner for both cooktops and ovens (Figure 3).

Cumulative Methane Emissions. For our methane emission measurements, we scaled our measurements to calculate the total amount of methane emitted from stoves overall, employing the usage patterns reported by Chan et al. and Zhao et al.[18,28] (see the Materials and Methods section). We estimated that an average stove (burners plus oven) emitted 649 [95% CI: 427, 949] g CH_4 $year^{-1}$ (Figure S5). When scaled to total U.S. emissions, and taking into account uncertainty in the estimate of the number of appliances reported in RECS, we estimated total national methane emissions from stoves were 28.1 [18.5, 41.2] Gg CH_4 $year^{-1}$. Using a 20 year timescale for the lifetime of methane, these methane emissions were comparable in climate impact to the carbon dioxide emissions of approximately 500 000 gas-powered cars.[31]

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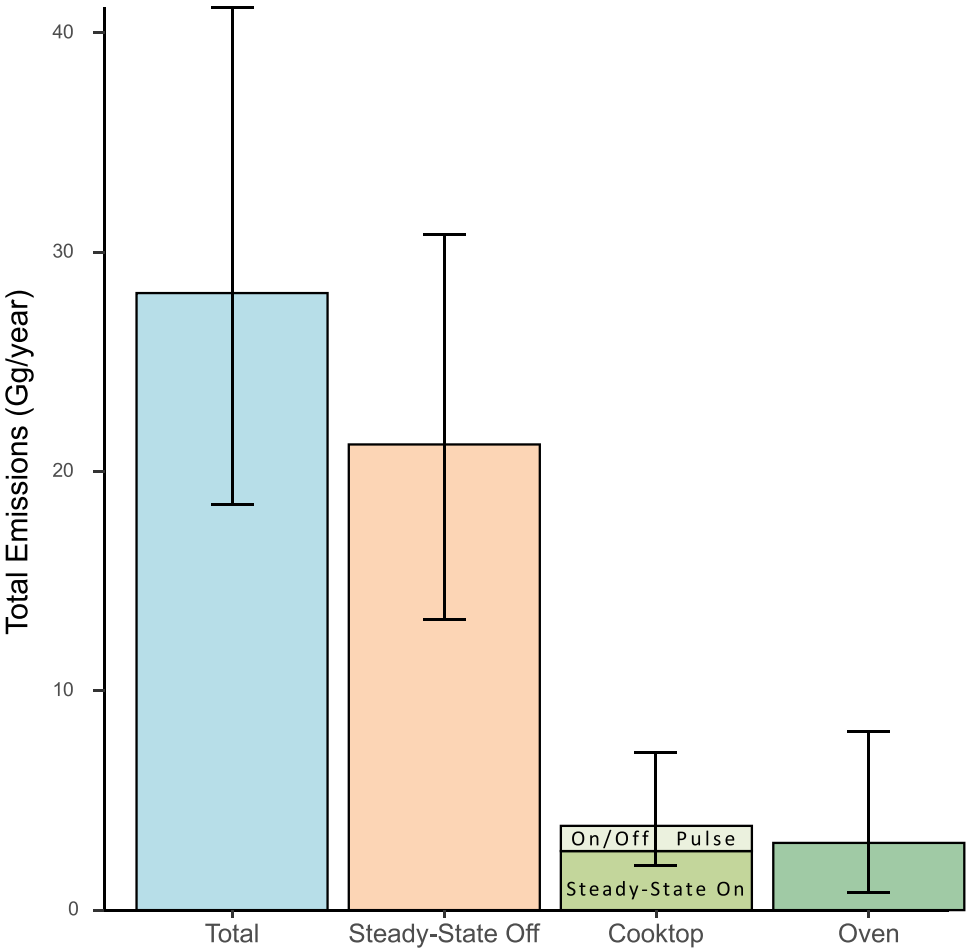

Figure 5. Total methane emissions by source. We report our scaled U.S. national estimates of methane emissions from stoves categorized into the various phases of emission sources: steady-state-off, cooktop emissions (broken down into steady-state-on and on/off pulses), and oven emissions. Bar height and error bars (95% confidence intervals) show the total methane emissions expected from each category. We found that 76% of the total methane emissions from stoves originated during steady-state-off.

Similar to our previous measurements in homes and to the results of other researchers,[7,8,27] our results were long-tail skewed. Considering our expanded dataset, a plot of the cumulative count of measurements vs fraction of emissions showed that the top 10% of all observations in the expanded dataset were responsible for emitting 47% of total emissions (Figure S6). These data approached the "5/50 rule" where the top 5% of emitters typically emit 50% of all emissions.[32]

We also calculated the total emissions based on whether the stove was on or off. Including both emission and usage data, we estimated that total emissions were 21.2 [13.3, 30.8] Gg CH_4 $year^{-1}$ during steady-state-off (i.e., emissions over most of the day); 2.7 [1.3, 5.3] Gg CH_4 $year^{-1}$ during steady-state-on from cooktops alone; and 1.1 [0.7, 1.9] Gg CH_4 $year^{-1}$ from the on/off pulses. For ovens alone, we only used emission data from the bake burner because we lacked comprehensive broil burner usage information but suspect it is small (~10%) based on user-reported datasets. Overall, we estimated that ovens emitted 3.1 [0.8, 8.1] Gg CH_4 $year^{-1}$ while on (Figure 5).

Volumetric Percentage Emissions. We used two methods to estimate the total percent methane emitted, the first using the report of gas used from the 2015 RECS survey and the second based on our CO_2 measurements combined with usage data (see the Materials and Methods section). From the first method, the volumetric fraction of our methane emissions using the RECS data was 1.3% of gas consumed, assuming that the volume of natural gas was measured at standard pressure and 60 °F. In the second method, we estimated that the total volumetric fraction emitted was lower—0.8%—because the appliances we measured used more gas than the national average from RECS. This approach assumes that our usage data and selection of low/medium/high burner intensities approximated usage in all regions. For example, stove usage does not seem to vary regionally in the United States in the same way that furnace usage varies attributable to local climate (Figure S7).[9]

Comparison to National Inventories. In 2019, the United States emitted an estimated 158 million tons of carbon dioxide equivalent ($MMTCO_2e$) of methane from natural gas systems. Because the EPA used a 100 year global warming potential of 25 for methane, this is equivalent to 6.3 Tg CH_4 $year^{-1}$.[6] The USEPA included specific emission sources such as residential and commercial meters and leaks from mains and services and also included a category for "stationary combustion" from various sectors (assumed to be incomplete combustion) but did not take into account leaks from appliances. While the USEPA did not report emissions from specific residential natural gas appliances, they reported methane emissions for residential stationary combustion from natural gas: 24 Gg CH_4 $year^{-1}$. From stoves alone, we

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estimated total methane emissions to be about 28 Gg CH_4 $year^{-1}$, more than the emissions currently reported by the USEPA from stationary combustion from all appliances, suggesting that the USEPA substantially underestimated emissions from residential natural gas combustion. Our previous work estimated that water heaters alone emitted 82 Gg CH_4 $year^{-1}$ from leaks and incomplete combustion, further suggesting that the USEPA is underestimating emissions from stationary combustion.[27]

We combined the total methane and carbon dioxide emissions from stoves and calculated that methane emissions add an extra one-third of CO_2e emissions to combustion-based CO_2 emissions from stove natural gas use. Using the estimate of natural gas consumption from RECS (113 billion cubic feet per year),[9] we estimated that CO_2 emissions from gas stoves are 6.2 $MMTCO_2$ $year^{-1}$ as a direct result of combusting the natural gas they use. Over a 20-year timescale, the global warming potential of methane is 86 times greater than carbon dioxide.[3] Using this scaling factor, we calculated the impact of methane over 20 years added an additional 2.4 $MMTCO_2e$ from methane emissions, or 39% of the 6.2 $MMTCO_2$ $year^{-1}$. Reducing these short-term climate impacts due to methane emissions is important for achieving climate goals in the coming decades.

Comparison to Previous Methane Studies. We compared our methane results to past studies that measured residential methane emissions. Merrin and Francisco estimated that national U.S. methane emissions from cooktop use—including both steady-state-on and on/off pulses for burners (but without steady-state-off emissions from stoves)—were 2.7 Gg CH_4 $year^{-1}$.[7] We estimated similar emissions nationally from the same sources, 3.8 Gg CH_4 $year^{-1}$ for burners (2.7 Gg CH_4 $year^{-1}$ for steady-state-on emissions and 1.1 Gg CH_4 $year^{-1}$ from on/off pulses). However, unlike Merrin and Francisco,[7] we also measured steady-state-off emissions from stoves nationally and estimated their emissions to be 21.2 Gg CH_4 $year^{-1}$, far larger than estimates for burners during combustion and on/off pulses.

Another study, Fischer et al., estimated that emissions from cooking appliances during steady-state-on from California alone totaled 1.6 Gg CH_4 $year^{-1}$.[8] In our work, we scaled our emissions to the number of stoves in California using the 2019 RASS data[33] and found that total methane emissions from stoves in California were 4.8 [95% CI: 3.1, 7.0] Gg CH_4 $year^{-1}$, of which 1.2 Gg CH_4 $year^{-1}$ comes from steady-state-on, on/off pulses, and ovens. Additionally, Fischer et al.[8] measured steady-state-off measurements in their "whole-home quiescent" leakage rates, which accounts for steady-state-off emissions from all appliances as well as interior pipeline leaks. They estimated this source to be 23.4 Gg CH_4 $year^{-1}$ for California. We calculated steady-state-off emissions from stoves alone in California to be 3.6 Gg CH_4 $year^{-1}$. Applying previous work by our lab to the numbers of appliances in California, we estimated that water heaters emit an additional 12.9 Gg CH_4 $year^{-1}$ during steady-state-off alone.[27] Together with the estimate of quiescent emissions from Fischer et al.,[8] our work provides strong evidence that steady-state-off emissions provide the largest source of natural gas leakage in homes.

Comparison to Previous NO_x Studies. Relatively little recent research has measured total NO_x flow rates from stoves, with the exception of Singer et al.,[17] who estimated NO_x emissions to be ~40 ng NO_x J^{-1} (ranged between 30 and 45 ng NO_x J^{-1}). Traynor et al. estimated that NO_x emissions from stoves had a geometric mean of 32 ng NO_x J^{-1},[34] and Singer et al. estimated NO_x emissions to be 30−36 ng NO_x J^{-1}.[35] Overall, these values are slightly higher than our estimate from cooktops: 21.7 [20.5, 22.9] ng J^{-1}. Previous modeling-based approaches suggest that residents are exposed to NO_2 levels above outdoor acute (1 h) exposure guidelines 62% of the time while using their stove but not using their range hoods,[36] and people use their range hoods only 28−36% of the time.[15]

Relationship between Emissions with Stove Age, Price, and Median Income. We also tested potential relationships among emissions and age, purchase price, and average income levels of where the stoves were located. For the stoves in our dataset with a known manufacturing date, we found no evidence of a relationship between either the age of the stove or purchase price of the stove with methane emissions nor with NO_x emissions normalized by burner energy (Figures S8 and S9; p values all >0.5). Finally, median income from census tracts where the stove was located had no relationship with methane emissions (Figure S10, p = 0.79).

STUDY LIMITATIONS AND RECOMMENDATIONS FOR FUTURE DIRECTIONS

To improve future home-appliance emissions research, we provide the following recommendations, building on suggestions from previous researchers:[7,8,37]

1. A rapid technique such as a portable methane detector could be used to locate the largest emitters qualitatively during steady-state-off conditions. In this study, we estimated that emissions from steady-state-off were responsible for 76% of all methane emissions from gas stoves.
2. Because of sampling limitations from COVID-19, we were limited in where and how we could sample homes and could not include a representative selection of low-income, multifamily homes; future work should emphasize measurements within this demographic.
3. More comprehensive data on appliance usage and activity should be collected. Such information would help researchers scale the number of on/off pulses per day and average consumption periods. Having better information about the number of burners that are on simultaneously and their emissions during real-world use would also help inform estimates for indoor NO_x pollution and indoor air quality modeling projects.
4. Additional measurements of direct emission rates of other pollutants from stoves should be collected, such as CO and formaldehyde, as such data are currently lacking. Additionally, we recommend measuring NO_x emission rates from other gas appliances and from electric appliances for comparison (i.e., as a control for comparison). Our data and additional data collection on emission factors could be used for indoor air quality modeling to calculate concentrations of NO_x that home occupants are exposed to under various cooking and ventilation scenarios.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c04707.

Details of calculations; photo of the setup; room volume estimates; volunteer information; methane emissions per

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stove; correlation plots; methane cumulative emissions from stoves; gas usage per household; NO_x concentrations from the use of an oven (PDF)

AUTHOR INFORMATION

Corresponding Author

Eric D. Lebel − *Department of Earth System Science, Stanford University, Stanford, California 94305, United States; PSE Healthy Energy, Oakland, California 94612, United States;* orcid.org/0000-0001-5255-6893; Email: elebel@stanford.edu, elebel@alumni.stanford.edu

Authors

Colin J. Finnegan − *Department of Earth System Science, Stanford University, Stanford, California 94305, United States*

Zutao Ouyang − *Department of Earth System Science, Stanford University, Stanford, California 94305, United States*

Robert B. Jackson − *Department of Earth System Science, Stanford University, Stanford, California 94305, United States; Woods Institute for the Environment, Stanford, California 94305, United States; Precourt Institute for Energy, Stanford, California 94305, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.1c04707

Notes

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