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Via E-Filing

Ms. Lisa Felice
Executive Secretary
Michigan Public Service Commission
7109 West Saginaw Highway
Lansing, MI 48917

RE: MPSC Case No. U-21534

Dear Ms. Felice:

Please find enclosed the Accompanying Exhibits DAO-211 to DAO-215 (Part 2 of 3) for the Direct Testimony of Yunus Kinkhabwala on Behalf of Soulardarity and We Want Green, Too, along with proof of service for electronic filing in the above-referenced matter. Please do not hesitate to contact me with any questions or comments.

Sincerely,

A handwritten signature in black ink, appearing to read "Mark N. Templeton".

Mark N. Templeton, *pro hac vice*
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Chicago, IL 60637
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xc: Parties to Case No. U-21534



Furnaces and Boilers

Energy Saver

[Energy Saver](#) » [Home Comfort](#) » [Home Heating Systems](#) » Furnaces and Boilers

Most U.S. homes are heated with either furnaces or boilers. Furnaces heat air and distribute the heated air through the house using ducts. Boilers heat water, and provide either hot water or steam for heating. Steam is distributed via pipes to steam radiators, and hot water can be distributed via baseboard radiators or radiant floor systems, or can heat air via a coil. Steam boilers operate at a higher temperature than hot water boilers, and are inherently less efficient; however, high-efficiency versions of all types of furnaces and boilers are currently available. To learn more about furnaces, boilers and other types of home heating systems, explore our [Energy Saver 101 infographic on home heating](#).





📷 Upgrading to a high efficiency furnace or boiler is an effective way to save money on home heating.

Understanding the Efficiency Rating of Furnaces and Boilers

A central furnace or boiler's efficiency is measured by annual fuel utilization efficiency (AFUE). The Federal Trade Commission requires new furnaces or boilers to display their AFUE so consumers can compare heating efficiencies of various models. AFUE is a measure of how efficient the appliance is in converting the energy from fuel to heat over the course of a typical year.

Specifically, AFUE is the ratio of the furnace's or boiler's annual heat output compared to its total annual fossil fuel energy consumed. An AFUE of 90% means that 90% of the energy in the fuel becomes heat for the home and the other 10% escapes up the chimney and elsewhere. AFUE doesn't include the heat losses of the duct system or piping, which can be as much as 35% of the energy for output of the furnace when ducts are located in the attic, garage, or other partially conditioned or unconditioned space.

An all-electric furnace or boiler has no flue loss through a chimney. The AFUE rating for an all-electric furnace or boiler is between 95% and 100%. The lower values are for units installed outdoors because they have greater jacket heat loss. However, despite their high efficiency, the higher cost of electricity in most parts

of the country makes all-electric furnaces or boilers an uneconomic choice. If you are interested in electric heating, consider installing a heat pump system.

Read about the [standards for residential furnaces](#).

You can identify and compare a system's efficiency not only by its AFUE but also by its equipment features.

Older, low-efficiency heating systems:

- Natural draft that creates a flow of combustion gases
- Continuous pilot light
- 56% to 70% AFUE.

Mid-efficiency heating systems:

- Exhaust fan controls the flow of combustion air and combustion gases more precisely
- Electronic ignition (no pilot light)
- Compact size and lighter weight to reduce cycling losses
- Small-diameter flue pipe
- 80% to 83% AFUE.

High-efficiency heating systems:

- Condensing flue gases in a second heat exchanger for extra efficiency
- Sealed combustion
- 90% to 98.5% AFUE.

Retrofitting Your Furnace or Boiler

Furnaces and boilers can be retrofitted to increase their efficiency. These upgrades improve the safety and efficiency of otherwise sound, older systems. The costs of retrofits should be carefully weighed against the cost of a new boiler or

furnace, especially if replacement is likely within a few years or if you wish to switch to a different system for other reasons, such as adding air conditioning. If you choose to replace your heating system, you'll have the opportunity to install equipment that incorporates the most energy-efficient heating technologies available.

Retrofits are fuel-specific, so see the following sections for retrofit information:

- **Gas-Fired Furnaces and Boilers** (includes units fired with natural gas and propane)
- **Oil-Fired Furnaces and Boilers.**

Other retrofitting options that can improve a system's energy efficiency include installing **programmable thermostats**, **upgrading ductwork** in forced-air systems, and adding zone control for hot-water systems, an option discussed in **Heat Distribution Systems**.

Replacing Your Furnace or Boiler

Although older fossil fuel furnace and boiler systems have efficiencies in the range of 56% to 70%, modern conventional heating systems can achieve efficiencies as high as 98.5%, converting nearly all the fuel to useful heat for your home. Energy efficiency upgrades and a new high-efficiency heating system can often cut your fuel bills and your furnace's pollution output in half. Upgrading your furnace or boiler from 56% to 90% efficiency in an average cold-climate house will save 1.5 tons of carbon dioxide emissions each year if you heat with natural gas, or 2.5 tons if you heat with oil.

If your furnace or boiler is old, worn out, inefficient, or significantly oversized, the simplest solution is to replace it with a modern high-efficiency model. Old coal burners that were switched over to oil or gas are prime candidates for replacement, as well as natural gas furnaces with pilot lights rather than electronic ignitions. Newer systems may be more efficient but are still likely to be

oversized, and can often be modified to reduce their operating capacity.

Before buying a new furnace or boiler or modifying your existing unit, it is suggested that you first improve the energy efficiency of your home by adding insulation and/or new energy-efficient windows, then have a heating contractor size your furnace. Energy-efficiency improvements will save money on a new furnace or boiler, because you can purchase a smaller unit. A properly sized furnace or boiler will operate most efficiently, and you'll want to choose a dependable unit and compare the warranties of each furnace or boiler you're considering.

When shopping for high-efficiency furnaces and boilers, look for the ENERGY STAR® label. If you live in a cold climate, it usually makes sense to invest in the highest-efficiency system. In milder climates with lower annual heating costs, the extra investment required to go from 80% to 90% to 95% efficiency may be hard to justify. However, keep in mind that higher efficiency units will have lower emissions than units in the 80% range.

Specify a sealed combustion furnace or boiler, which will bring outside air directly into the burner and exhaust flue gases (combustion products) directly to the outside, without the need for a draft hood or damper. Furnaces and boilers that are not sealed-combustion units draw heated air into the unit for combustion and then send that air up the chimney, wasting the energy that was used to heat the air. Sealed-combustion units avoid that problem and also pose no risk of introducing dangerous combustion gases into your house. In furnaces that are not sealed-combustion units, back drafting of combustion gases can be a big problem.

High-efficiency sealed-combustion units generally produce an acidic exhaust gas that is not suitable for old, unlined chimneys, so the exhaust gas should either be vented through a new duct or the chimney should be lined to accommodate the acidic gas (see the section on maintaining proper ventilation below).

Maintaining Your Furnace or

Boiler

The following maintenance should be provided by a heating system professional.

All systems:

- Check the condition of your vent connection pipe and chimney. Parts of the venting system may have deteriorated over time. Chimney problems can be expensive to repair, and may help justify installing new heating equipment that won't use the existing chimney.
- Check the physical integrity of the heat exchanger. Boiler heat exchangers may leak water and are easy to spot. Furnace heat exchangers mix combustion gases with house air when they leak—an important safety reason to have them inspected. Combustion gases leaking into the house can cause carbon monoxide poisoning. It is suggested that every home have a working carbon monoxide alarm.
- Adjust the controls on the boiler or furnace to provide optimum water and air temperature settings for both efficiency and comfort.
- If you're considering replacing or retrofitting your existing heating system, have the technician perform a combustion-efficiency test.

Forced Air Systems:

- Check the combustion chamber for cracks
- Test for carbon monoxide (CO) and remedy if found
- Adjust blower control and supply-air temperature
- Clean and oil the blower
- Remove dirt, soot, or corrosion from the furnace or boiler
- Check fuel input and flame characteristics, and adjust if necessary
- Seal connections between the furnace and main ducts.

Hot Water Systems:

- Test pressure-relief valve
- Test high-limit control
- Inspect pressure tank, which should be filled with air, to verify that it's not

filled with water

- Clean the heat exchanger.

Steam Systems:

- Drain some water from the boiler to remove sediments and improve the heat exchange efficiency
- Test low-water cutoff safety control and high-limit safety control
- Drain the float chamber to remove sediments, which will prevent the low-water cutoff control from sediment clogs
- Analyze boiler water and add chemicals as needed to control deposits and corrosion
- Clean the heat exchanger
- See also [steam radiators](#).

Chimneys

Properly functioning chimney systems will carry combustion byproducts out of the home. Therefore, chimney problems put you at risk of having these byproducts, such as carbon monoxide, spill into your home.

Most older furnaces and boilers have natural draft chimneys. The combustion gases exit the home through the chimney using only their buoyancy combined with the chimney's height. Natural draft chimneys often have problems exhausting the combustion gases because of chimney blockage, wind or pressures inside the home that overcome the buoyancy of the gases.

Atmospheric, open-combustion furnaces and boilers, as well as fan-assisted furnaces and boilers, should be vented into masonry chimneys, metal double-wall chimneys, or another type of manufactured chimney. Masonry chimneys should have a fireclay, masonry liner or a retrofitted metal flue liner.

Many older chimneys have deteriorated liners or no liners at all and must be relined during furnace or boiler replacement. A chimney should be relined when any of the following changes are made to the combustion heating system:

- When you replace an older furnace or boiler with a newer one that has an AFUE of 80% or more. These mid-efficiency appliances have a greater risk of depositing acidic condensation in chimneys, and the chimneys must be prepared to handle this corrosive threat. The new chimney liner should be sized to accommodate both the new heating appliance and the combustion water heater (if present) by the installer.
- When you replace an older furnace or boiler with a new 90+ AFUE appliance or a heat pump. In this case, the heating appliance will no longer vent into the old chimney, and if the combustion water heater is present it will now vent through an oversized chimney. This oversized chimney can lead to condensation and inadequate draft. The new chimney liner should be sized for the water heater alone, or the water heater in some cases can be vented directly through the wall.

Other Ventilation Concerns

Some fan-assisted, non-condensing furnaces and boilers, installed between 1987 and 1993, may be vented horizontally through high-temperature plastic vent pipe (not PVC pipe, which is safely used in condensing furnaces). This type of venting has been recalled and should be replaced by stainless steel vent pipe. If horizontal venting was used, an additional draft-inducing fan may be needed near the vent outlet to create adequate draft. Floor furnaces may have special venting problems because their vent connector exits the furnace close to the floor and may travel 10 to 30 feet before reaching a chimney. Check to see if this type of venting or the floor furnace itself needs replacement. If you smell gases, you have a venting problem that could affect your health. Contact your local utility or heating contractor to have this venting problem repaired immediately.

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

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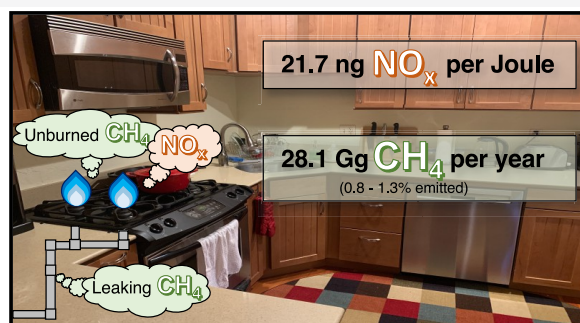
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Supporting Information



ABSTRACT: Natural gas stoves in >40 million U.S. residences release methane (CH₄)—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 CH₄ 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 NO₂) 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 NO₂ 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 NO₂ (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 (CH₄) 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 (CO₂) 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 chain⁴ 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

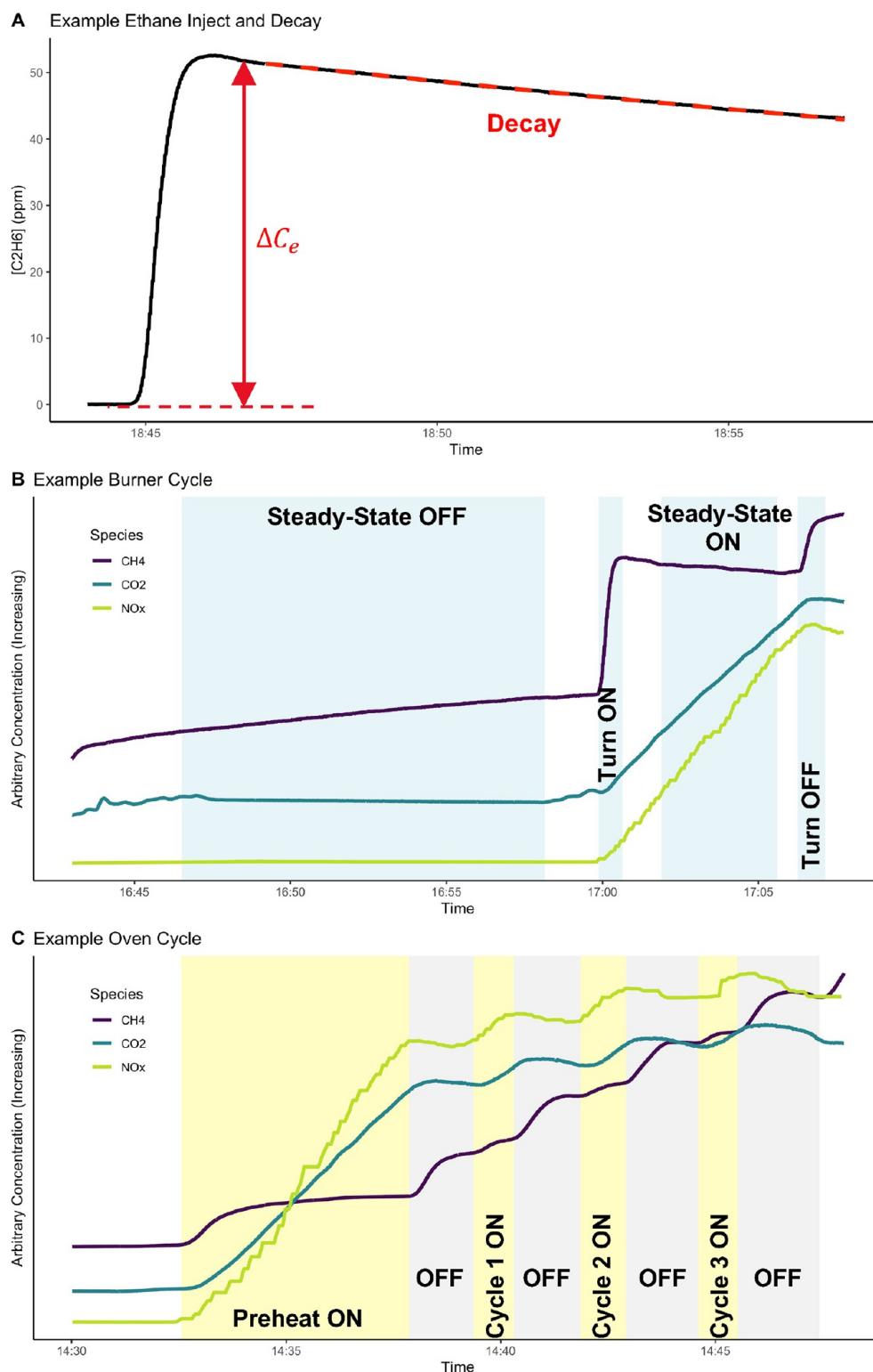


Figure 1. Example concentration plots for various measurement events using data from actual measurements in our study. (A) Ethane injection and decay: first, we injected 500 mL of ethane into the space created by the plastic wall to determine its volume based on dilution. ΔC_e is the change in ethane 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.

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

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

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 $\text{Burner.Ignitions}_j$ 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}

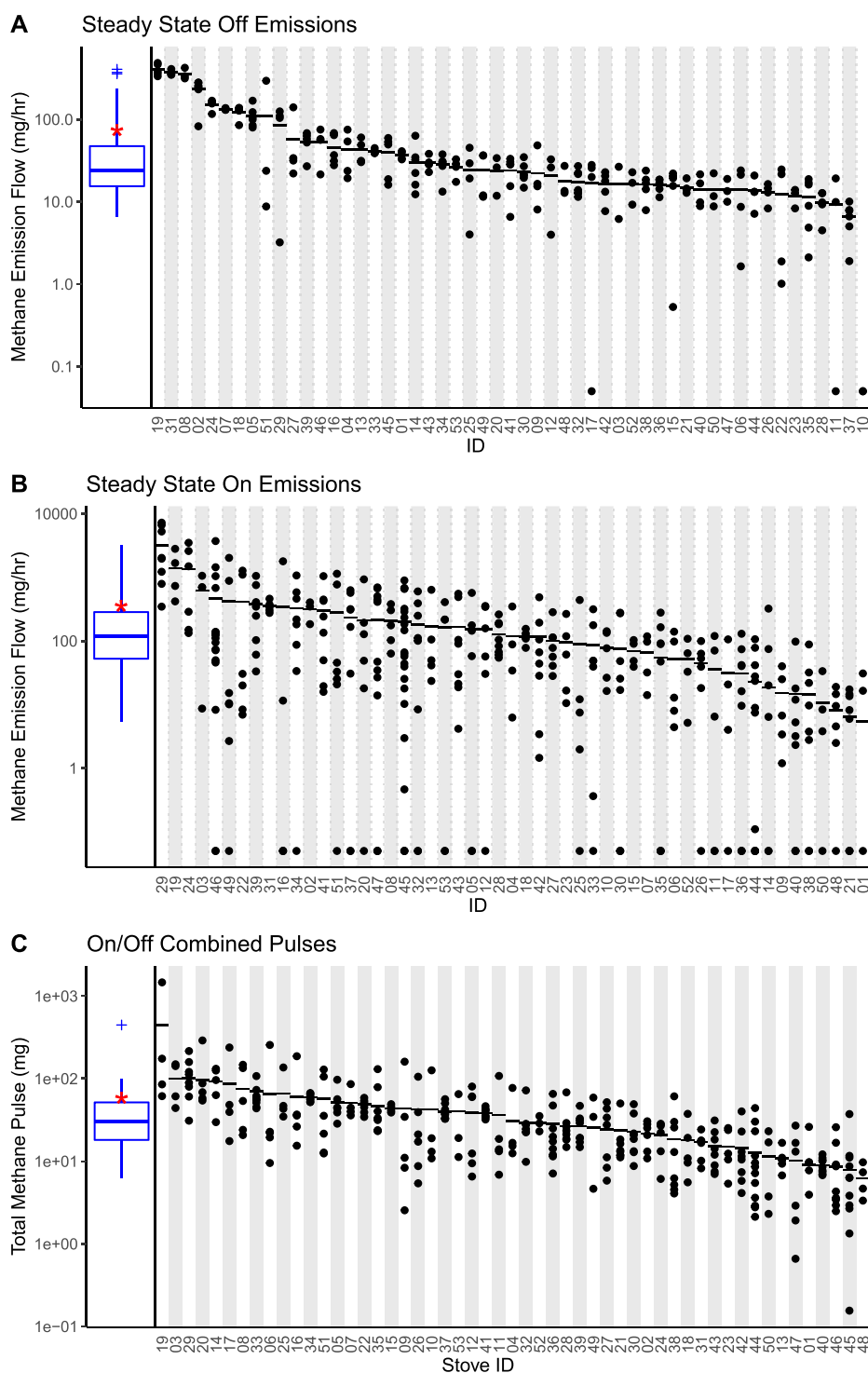


Figure 2. Stove emission components from each individual stove, represented on a log scale, from steady-state-off, steady-state-on, and the on/off pulses. The blue box-and-whisker plot on the left in each panel summarizes each component of stove emissions. The median, 25th, and 75th quartiles are shown, 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 ($58 \text{ mg CH}_4 \text{ h}^{-1}$) was higher than the median ($24 \text{ mg CH}_4 \text{ h}^{-1}$). Negative values were set to $0.05 \text{ mg CH}_4 \text{ h}^{-1}$. (B) Average steady-state-on emission was $259 \text{ mg CH}_4 \text{ h}^{-1}$, which is slightly higher than the median ($119 \text{ mg CH}_4 \text{ h}^{-1}$). Negative values were set to $0.05 \text{ mg CH}_4 \text{ h}^{-1}$. (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).

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 CH₄ 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 CH₄ 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 CH₄ 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 CO₂ 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 CO₂ 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 NO₂ 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 CH₄ (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 CH₄. 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.

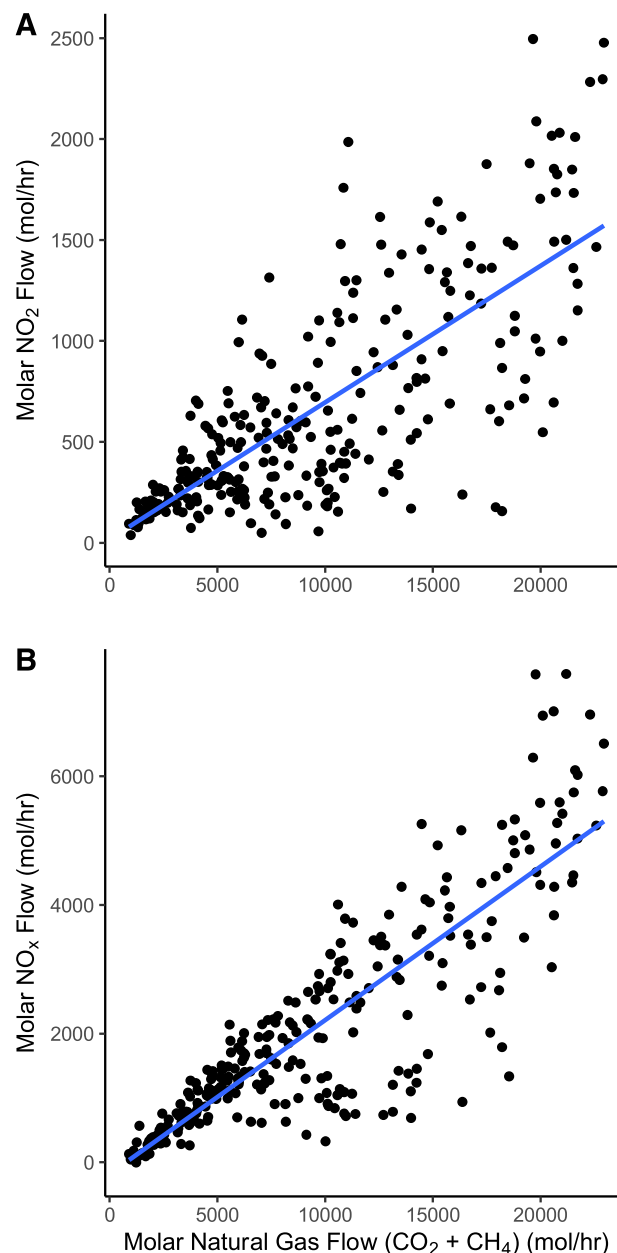


Figure 3. NO₂ and NO_x flows are linearly proportional to natural gas flow. Each black dot represents a measurement of steady-state-on, either for a cooktop burner, or for an oven broil or bake burner. (A) Emissions of NO₂ are plotted on the y-axis with the natural gas flow (i.e., emissions of CO₂ + CH₄) 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 NO₂ flow rate ($p \ll 0.01$; $r^2 = 0.59$). (B) Similarly, for NO_x (i.e., NO + NO₂), 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 CH₄. 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 CH₄ per on/off pulse, similar to the burners with a functioning electric sparker. We

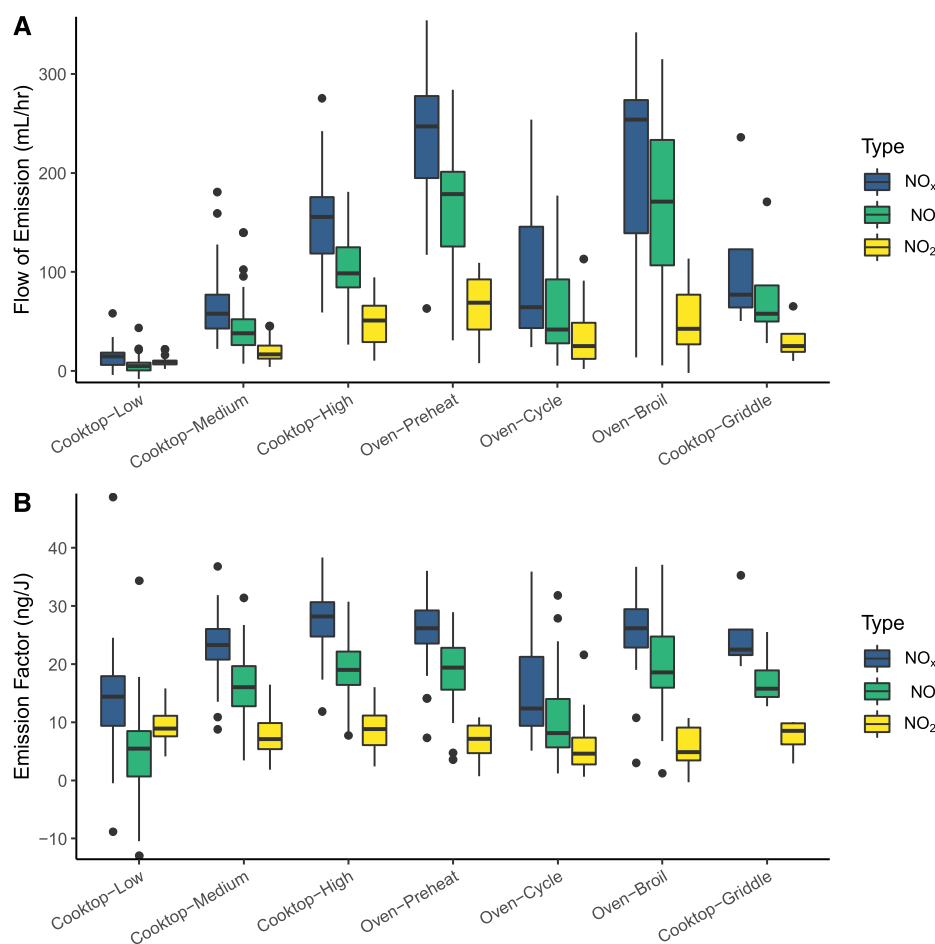


Figure 4. NO_x, NO, and NO₂ total emissions and normalized by energy output. NO_x emissions are shown in total emissions for various burner sizes and intensities and by constituents: NO and NO₂. We separate emissions into various phases of stove operation: cooktop emissions while burners are on low, 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 CO₂ 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 CH₄ over a period of about 13 min, with an average rate of 663 [408, 1030] mg CH₄ 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 CH₄ h⁻¹. In comparison, the broiler burner only emitted 112 [50, 186] mg CH₄ 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 NO₂ 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 NO₂ 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 NO₂ h⁻¹) for the broil burners. We found that ovens could produce enough NO₂ to exceed the 1-h ambient standard (100 ppb) within a few minutes (Figure S11). We also found that NO₂ 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 CH₄ 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 CH₄ 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

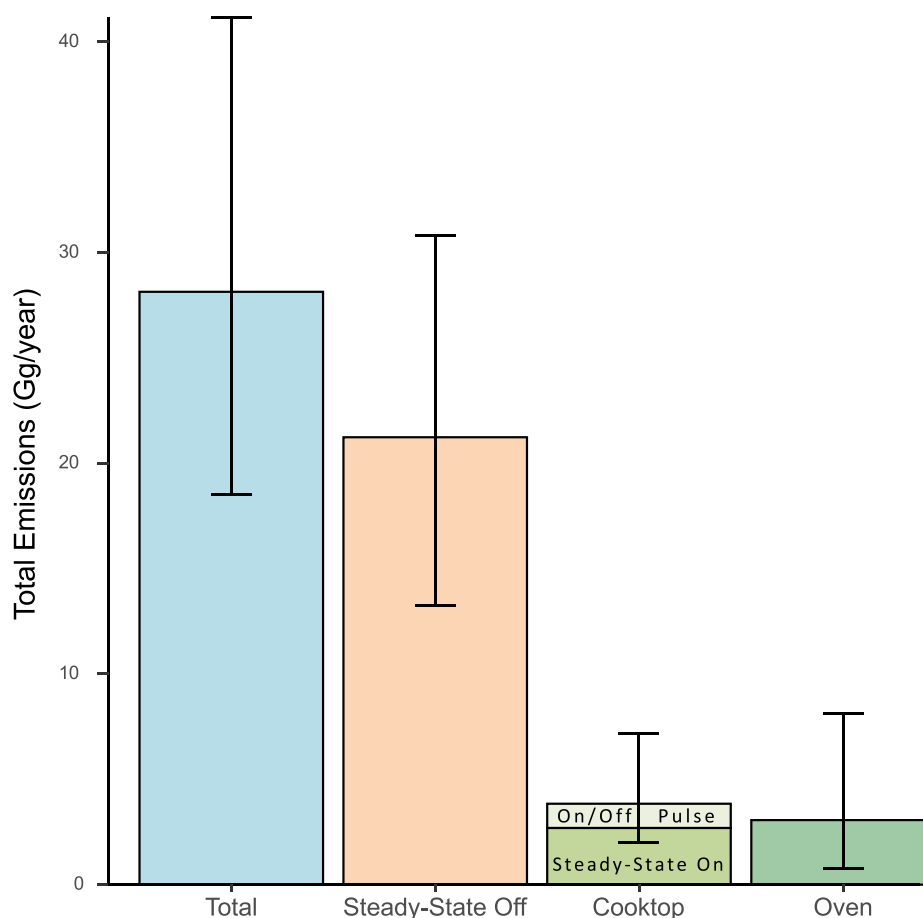


Figure 5. Total methane emissions by source. We report our scaled U.S. national estimates of methane emissions from stoves categorized into the various phases of emission sources: steady-state-off, cooktop emissions (broken down into steady-state-on and on/off pulses), and oven emissions. Bar height 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 CH₄ year⁻¹ during steady-state-off (i.e., emissions over most of the day); 2.7 [1.3, 5.3] Gg CH₄ year⁻¹ during steady-state-on from cooktops alone; and 1.1 [0.7, 1.9] Gg CH₄ 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 CH₄ 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 CO₂ 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 (MMT CO₂e) 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 CH₄ 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 CH₄ year⁻¹. From stoves alone, we

estimated total methane emissions to be about 28 Gg CH₄ 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 CH₄ 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 CO₂e emissions to combustion-based CO₂ 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 CO₂ emissions from gas stoves are 6.2 MMTCO₂ 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 MMTCO₂e from methane emissions, or 39% of the 6.2 MMTCO₂ 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 CH₄ year⁻¹.⁷ We estimated similar emissions nationally from the same sources, 3.8 Gg CH₄ year⁻¹ for burners (2.7 Gg CH₄ year⁻¹ for steady-state-on emissions and 1.1 Gg CH₄ 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 CH₄ 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 CH₄ year⁻¹.⁸ In our work, we scaled our emissions to the number of stoves in California using the 2019 RASS data³³ and found that total methane emissions from stoves in California were 4.8 [95% CI: 3.1, 7.0] Gg CH₄ year⁻¹, of which 1.2 Gg CH₄ 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 CH₄ year⁻¹ for California. We calculated steady-state-off emissions from stoves alone in California to be 3.6 Gg CH₄ 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 CH₄ 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 NO₂ 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

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

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Notes

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Pollutant concentrations and emission rates from natural gas cooking burners without and with range hood exhaust in nine California homes



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ABSTRACT

Combustion pollutant concentrations were measured during the scripted operation of natural gas cooking burners in nine homes. Boiling and simmering activities were conducted on the stovetop and in the oven with and without range hood exhaust ventilation or air mixing via a forced air system. Time-resolved concentrations of carbon dioxide (CO₂), nitric oxide (NO), nitrogen oxides (NO_x), nitrogen dioxide (NO₂), particles with diameters of 6 nm or larger (PN), carbon monoxide (CO), and fine particulate matter (PM_{2.5}) were measured in the kitchen and bedroom area of each home. Four of the nine homes had kitchen 1 h NO₂ exceed the national ambient air quality standard (100 ppb). In all homes, the highest 1 h integrated PN exceeded $2 \times 10^5 \text{ cm}^{-3}\text{-h}$, and the highest 4 h PN exceeded $3 \times 10^5 \text{ cm}^{-3}\text{-hr}$ in the kitchen. Range hood performance varied widely, but one with a large capture volume and a measured flow of 108 L/s reduced concentrations 80–95%. Increased awareness of the need to ventilate when cooking, along with building standards for minimum range hood flow rates and volume, could substantially reduce exposures to NO₂ and ultrafine particles in homes.

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1. Introduction

The combustion products of natural gas cooking burners (NGCBs) include pollutants that can degrade indoor air quality. While complete combustion directly produces water vapor and carbon dioxide (CO₂), the high flame temperatures also produce nitrogen oxides (NO_x) including nitrogen dioxide (NO₂), a respiratory irritant. Incomplete combustion can produce non-negligible emissions of other air pollutants including carbon monoxide (CO), formaldehyde (CH₂O), and nanometer-sized particles (PN) [25]. Residential NGCBs emit nanoparticles that grow to tens of nm but mostly remain within the <100 nm diameter threshold that defines ultrafine particles (UFP) [20,39].

The U.S. EPA sets national ambient air quality standards to protect both the general population and sensitive sub-populations [35,36]. The EPA limits for CO are 35 ppm averaged over 1 h and 9 ppm averaged over 8 h. The short-term exposure standard for NO₂ is 100 ppb over 1 h. There is a short term standard for fine particulate matter, PM_{2.5}, of 35 µg m⁻³ averaged over 24 h;

however, the particles emitted from NGCBs don't have sufficient mass to approach this threshold. Currently there are no widely recognized standards or guidelines for UFP exposure. An expert elicitation review of the available literature rated the likelihood of increased short-term UFP exposure causing health effects as medium to high [12]. Another review noted the substantial experimental evidence and plausible mechanisms for respiratory and cardiovascular effects of UFP intake, but deemed the evidence as “not sufficiently strong to conclude that short-term exposures to UFPs have effects that are dramatically different from those of larger particles” [11].

Emission factors of CO and NO₂ from NGCBs have been measured in laboratory and field studies [16,25,26,34]. Several studies have reported emission factors and/or indoor concentrations of ultrafine particles resulting from NGCB use without food preparation [2,20,25,39]. Many studies have reported elevated concentrations of CO and NO₂ in homes with natural gas cooking burners, compared to homes with electric cooking [8,18,23,24,29,30,41,42]. A recent study of 350 California homes reported that NO₂ and NO concentrations increased with increasing self-reported use of NGCBs across homes [18]. A modeling study of

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multifamily housing in Boston found that cooking with gas burners is a major source of NO₂ in homes [4].

While several measurement-based studies have reported time-resolved CO, (e.g. [18]), only a few have reported time-resolved or peak NO₂ concentrations resulting from NGCB use [5,7,17,19,22]. A recent simulation study found that the weekly highest 1 h mean NO₂ concentrations exceed 100 ppb in the majority of a representative sample of Southern California homes in which NGCBs were used without kitchen exhaust ventilation [13].

The primary strategy for mitigating exposure to pollutants from cooking burners is to use a venting range hood or other kitchen exhaust ventilation [33]. Recent assessments of range hoods in the U.S. indicate wide performance variations across devices and also across airflow settings and burner configurations for many devices tested [3,15,21,28]. Several of these studies used capture efficiency, CE, as the performance metric. CE indicates the fraction of pollutants generated at the cooking appliance that are removed or exhausted by the range hood before they can mix into the air of the home. These studies found that for many range hoods, CE is much higher for the back than for the front cooktop burners. The [13] modeling study of Southern California homes found that routine use of a venting kitchen range hood with a 52% CE (reflecting performance of a common hood for front burner cooking) should dramatically reduce the percentage of homes with 1 h mean NO₂ exceeding 100 ppb.

NGCB pollutants that are not exhausted directly by a range hood are removed from the air in the home by air exchange with outdoors and for NO₂ and PN by deposition to interior surfaces. Deposition rates for NO₂ in US homes can be estimated from data reported in several studies. A comparison of decay rates for NO₂ and the conserved tracer SF₆ following gas burner use in 5 homes yielded an estimated deposition rate of 0.8/h [31]. Comparing decay rates of NO₂ to those for NO, CO, and CO₂ – which are not removed by deposition – obtained in 21 homes using unvented gas fireplaces [6,9] yielded a mean deposition rate estimate of 0.7/h. Particle deposition rates vary with particle size and environmental conditions. Measurements in homes provide estimated deposition rates for 15–50 nm particles that range from 0.2/h to 3/h [1,10,14].

The primary objective of the research reported here was to quantify time-resolved concentrations of NO₂ resulting when NGCBs are used under realistic conditions, and specifically to investigate if the threshold of 100 ppb over 1 h is commonly exceeded. We also sought to measure concentrations of NO, NO_x, CO₂, CO, PM_{2.5}, and the number of particles with diameters ≥ 6 nm (PN, most of which are UFP) following controlled burner use. Another objective was to conduct a pilot study of the benefits of using venting range hoods to reduce in-home concentrations of pollutants emitted by NGCBs.

2. Methods

2.1. Overview

The study entailed operation of NGCBs and measurements of the resulting pollutant concentrations in nine homes in the San Francisco Bay area of California. Experiments were conducted, by permission, when residents were away from the home. Researchers controlled the operation of cooking appliances, ventilation, and forced-air heating systems. The NGCB operation sequences were designed to represent common cooking patterns. To avoid generating pollutants from food preparation, pots containing tap water were used as heat sinks. Air pollutants including NO_x, NO, number concentrations of particles ≥ 6 nm (PN), CO, CO₂, and estimated PM_{2.5} were measured in the kitchen and a hallway or bedroom that was far from the kitchen. CO₂ was also measured in a common

room between the other two locations but generally closer to the kitchen. NO₂ was inferred as the difference between NO_x and NO, even though that value likely includes non-negligible amounts of nitrous acid (HONO) [32].

The base set of experiments included operation of each type of cooking burner (cooktop, oven bottom burner, and broiler top burner, as available) with windows closed, no forced air unit (FAU) operation, and no mechanical exhaust. Additional experiments were conducted with the FAU operated in fan-only mode when this setting was available, and with a venting range hood when available.

2.2. Study homes

The nine homes varied in size and layout, as described in Table 1. They included seven detached houses, one flat (first floor of two-flat duplex), and a small apartment. There were three homes with open floor plans and no walls enclosing the kitchen. Four of the homes had kitchens that were distinct rooms, connected to other rooms in the home via standard interior doorways. Two homes had semi-open kitchens. One of these (labeled H6) had a small galley kitchen with both a floor-to-ceiling passage and a large pass-through connecting the kitchen to the adjacent dining room. The other (H9) had two wide, open passages between the kitchen and adjacent rooms.

Cooking appliances varied across homes. Table 2 summarizes the natural gas cooking appliances in each study home. The burner firing rates were obtained from the nameplate tag found on the appliance or by searching online product literature for the make and model. Five homes had a gas range with cooktop, oven bottom burner and broiler top burner; two homes had a gas range with only a cooktop and oven burner; and two homes had a gas countertop cooktop separate from an electric oven. A venting range hood was present in six homes. Six homes had FAUs that could be operated in fan-only mode.

Study home access was arranged with owners or renters who were paid \$200 for each day and \$200 for each overnight period that a home was unoccupied and made available for experiments, up to a total allowable payment per home of \$600. A single day of experiments required 11 h of access to the home without occupants.

Table 3 describes the kitchen exhaust fans in the study homes. Six of the homes had exhaust devices above the cooktop. Two of the venting hoods were “microwave over range” (MOR) appliances that combine the functions of a microwave and externally venting exhaust fan. Home H3 and H4 had no range hoods of any kind. H7 had a non-venting (recirculating) range hood that was operated during two experiments.

2.3. Burner operation and simulated cooking

A procedure was developed to simulate common usage scenarios for cooktop, oven, and broiler burners. The procedures, used in homes H2 to H9, are described in Table 4. The “Boil” and “Sauté” activities were done simultaneously in the “Cooktop” procedure. The procedures were not finalized until after experiments were completed in H1; H1 experiments featured variations of the procedures, as discussed in the Supplemental materials. The same clean pans and pots were used in all homes, but cooking appliances generally were operated as found. In a couple of cases the appliances were wiped with wet paper towels to remove large debris.

Upon arriving to each home, we reviewed the planned experimental procedures with the host (homeowner or renter) and obtained her/his signature for the agreed usage periods. With the host, we conducted a walk-through to identify potential hazards

Table 1
Study home and burner descriptions.

ID	Floor area (m ²)	Levels	BR/Ba	Year built	Kitchen design	Flooring ^a	Gas burners ^b	Venting range hood?	FAU fan only mode?
H1	134	1	2/2	1910	Closed	Hard	CT/O/B	Y	Y
H2	124	1	4/2	1963	Open	Hard	CT/O/B	Y	Y
H3	117	1.5 ^c	2/2	1904	Closed	Hard	CT/O	N	N
H4	26	1	1/1	<1990 ^d	Closed	Hard	CT/O	N	No FAU
H5	108	1	3/1	1925	Closed	Hard	CT/O/B	Y	No
H6	119	2	2/2.5	1991	Semi-open	Hard L1, carpet on stairs & L2	CT/O/B	Y	Y
H7	226	2	5/3	1990	Open	Hard L1, carpet on stairs & L2	CT	N	Y
H8	219	2	4/3	1990	Open	Hard L1, carpet on stairs & L2	CT	Y	Y
H9	139	2	3/2.5	1986 ^e	Semi-open	Hard K, other rooms carpeted	CT/O/B	Y	Y

^a L1 = level 1 or first story; L2 = level 2 or second story; K = kitchen.

^b CT = cooktop; O = oven; B = broiler (top of oven compartment).

^c Small room below kitchen connected via stairwell at back of kitchen (house on hill).

^d Building was renovated and expanded in 1990. Original year of construction was not determined.

^e Home was retrofitted for energy efficiency with extensive air sealing, insulation and windows; it thus has characteristics of a new, energy efficient home.

Table 2
Natural gas cooking appliances in study homes.

ID	Cooktop type	Cooktop burner type	Burner firing rate (kbtu/h)					
			Left front (LF)	Right front (RF)	Left rear (LR)	Right rear (RR)	Oven	Broiler
H1	Range	Sealed	9.5	16.2	14.2	5	18	15.5
H2	Range	Sealed	9.5	17	14	5	18	15.5
H3	Range	Open	9	9	9	9	18	None
H4	Range	Open	9	9	9	9	18	None
H5	Range	Sealed	12	12	9.2	9.2	18	15
H6	Range	Sealed	9.5	9.5	9.5	9.5	16	13.5
H7	Counter	Sealed	12–14 ^a	14–16 ^a	5 ^a	5 ^a	Elec.	Elec.
H8	Counter	Sealed	9.5	14.2	11	5	Elec.	Elec.
H9	Range	Sealed	9.5	12	9.5	5	16	12

^a Estimated. Cooktop specifications were not obtained for this home.

Table 3
Range hoods in study homes.

ID	Type	Make/model	Airflow from product literature (L/s)		Sound level from product literature (Sones)	
			Low speed	High speed	Low speed	High speed
H1	Vented range hood	Vent-A-Hood B200 MSC	Not listed	531	Not listed	6.5
H2	Microwave with exhaust fan	Electrolux FGMV174KFB	85	142	Not listed	Not listed
H5	Vented range hood	Zephyr Cyclone AK6500	142	142	1.5	5
H6	Microwave with exhaust fan	GE Space-maker JVM140	Not listed	230	Not listed	5.5
H7	Non-venting range hood	Broan QS3	Not listed	109	0.3–0.5 ^b	4.5–5.5 ^b
H8 ^a	Vented range hood	Kenmore 233.516891	50	76	Not listed	6
H9 ^a	Vented range hood	Broan 42000E	50	76	Not listed	6

^a The hoods in H8 and H9 are the same product, sold under different nameplates.

^b Varies with vent geometry.

Table 4
Burner operating procedures used for most experiments in homes H2 to H9.

Burner	Simulated Activity	Description
Cooktop	Boil (Simultaneous with Sauttee)	Add 4L cold tap water to 5L stainless steel pot with 23 cm diameter base. Place uncovered on largest cooktop burner. At start, set burner to high. When water reaches rolling boil, cover and turn down burner to just below boiling. Maintain this condition to complete 30 min total burn.
	Sauttee (Simultaneous with Boil)	Place stainless steel fry pan with 30.5 cm diameter base on second largest burner. At start, set burner to high. At 2 min from start, slowly add 1L cold tap water. At 6 min from start, turn burner to medium setting. Adjust burner down as needed to avoid boiling. Stop burner at 30 min total burn.
Oven	Roast	Remove top oven rack and place bottom rack to allow 5L stainless steel pot to fit into oven. At start, set oven temperature to 218 °C (425 °F). When oven reaches set point (or after 20 min if set point not yet reached), place uncovered pot containing 1L water on bottom oven rack. Maintain this condition for 30 min; then turn oven off and remove pot. (Note: total burn time varies with time to reach set point temperature.)
Broiler	Broil	Remove top oven rack and place bottom rack to hold 5L stainless steel pot 13 cm below broiler burner at top of oven. At start, set oven to “broil” with oven door ajar. After 20 min, place 5L pot containing 1L water into oven. With door ajar, maintain this condition for 15 min; then turn burner off, remove pot, and close door.

and confirm acceptability of the planned placement of monitoring equipment (for electrical capacity). The host then left the home to return at the agreed time.

Study homes were used for varying duration as summarized in [Supplemental Table S1](#). When experiments were conducted during consecutive days with the host residing during the intervening night, all instruments were shut down at the end of the first day then restarted when the host left the home the following morning. When the home was accessible for a night-day sequence, monitoring instruments operated continuously overnight.

The protocol prior to starting cooking experiments included set-up and airflow checks of air quality instruments, recording information about the home and appliances, and measurements to characterize range hood performance (if it vented to outdoors). Experiments were conducted with all windows closed and interior doors open with two exceptions. It was discovered mid-way through experiments at H4 that there was a bathroom window open about 15 cm; the window was left in this position for all experiments in H4. In H8 there was a large opening in the master bathroom ceiling, related to a home construction project; this was addressed by keeping the door to the master bath closed throughout experiments in H8.

Each experiment included the following elements. (1) Air movement and mixing in the home was driven either by extant temperature and pressure variations or by operation of the forced air unit (FAU). (2) If present, the venting range hood was either operated or not. (3) The simulated cooking procedure was followed. (4) Following the end of the cooking procedure, and range hood use if it was part of the experiment, ventilation and mixing in the home were held in the same condition for 60–90 min or more, to enable analysis of mixing and decay. When the FAU was used, in most cases it was started approximately 10 min prior to the cooking activity, and operated until the mixing condition was established for the next experiment. For experiments involving range hood use, the hood was started approximately 1 min prior to the start of cooking, and remained in operation until 5 min after the cooking procedure ended. Range hoods were most commonly operated on the highest setting, though lower settings were used in several experiments. Excluding one experiment in H1 in which the range hood was started 15 min after the end of a cooking procedure, mixing and decay periods varied from a minimum of 54 min to a maximum of overnight.

2.4. Air quality measurements

Instruments continuously monitored the concentrations of carbon dioxide (CO₂), carbon monoxide (CO), nitric oxide (NO), nitrogen oxides (NO_x), total number concentration of particles greater than 6 nm (PN), number concentrations of particles in 6 size bins from 0.3 μm to 10 μm, and the estimated mass of PM_{2.5} via forward light scattering. Temperature and relative humidity were also monitored. The instruments used to measure each of these parameters are noted in [Supplemental Table S2](#) with performance specifications provided in [Table S3](#). Photos of the instrument carts deployed in a kitchen and a bedroom are provided as [Figs. S1 and S2](#). The values reported for NO₂ are based on the difference between measured NO_x and NO, and likely include HONO [\[32\]](#); for simplicity, we refer to the NO₂ value reported by the instrument as NO₂ throughout this report.

Air quality monitoring occurred in two primary locations and one secondary location in each home. The primary locations were the kitchen and a bedroom or hallway location far from the kitchen. The bedroom area location (henceforth described simply as bedroom) was used to determine an approximate lower bound of combustion pollutant concentrations and potential exposures. The kitchen and bedroom locations had nearly identical collections of

instruments, mounted on mobile carts. The kitchen cart was situated to ensure that the air inlet was no closer than 2 m from the closest cooktop burner in all homes other than the small apartment H4, in which this distance was slightly less than 2 m. The devices on the cart sampled air at heights of roughly 1.4–1.65 m. The third monitoring package was installed on a table in a common room (living room, dining room, great room) closer to the kitchen, or connected to the kitchen in an open floor plan. Air quality data were recorded at 1 min or more frequent intervals.

Particle instruments for the kitchen and bedroom carts were operated side-by-side in a lab test chamber to confirm consistency prior to deployment. CO₂ instruments were calibrated after the field study and calibration factors were applied to the raw data. CO instruments were used as received new from the manufacturer, without an independent calibration. The NO_x and NO₂ analyzers were single-point calibrated at the start of the field study, then checked for zero and span calibration in the laboratory prior to most home deployments. Despite this, it was observed that in most homes, the NO and NO₂ (NO_x–NO) concentrations were offset between instruments at times when the home was thought to be well mixed. The observed differences between the two NO_x analyzers were used to determine an offset adjustment. Because the NO concentrations reported by the bedroom instrument were consistently in line with data obtained from nearby regulatory monitoring sites, at times when the homes were not impacted by indoor NO_x emissions, the offset was typically applied to the NO_x analyzer on the kitchen cart.

2.5. Range hood performance characterization

Measurements were made to quantify airflow and sound characteristics of all the venting range hoods in the study. The capture efficiency (CE) of cooking burner combustion products was measured for five of the six venting hoods. CE could not be determined for the hood in H1 because the range hood vent was inaccessible to make the required exhaust CO₂ measurements.

Exhaust air flow from each range hood was measured using a balanced-pressure flow hood method described by Ref. [\[38\]](#). A calibrated and pressure-controlled variable-speed fan (Minneapolis Duct Blaster, Energy Conservatory¹) was connected to either the exhaust inlet (preferred approach) or outlet. The Duct Blaster was connected at each site using a custom transition constructed with cardboard and tape. Using a pressure sensor, the Duct Blaster fan was controlled to match the flow of the exhaust fan while maintaining neutral pressure to the room at the exhaust inlet. The pre-calibrated speed versus flow relationship of the Duct Blaster provided the flow through the exhaust fan.

Sound levels were measured using the AudioTools app (version 8.9.X) from Studio Six Digital² on an iPhone6. We used the “Real Time Analyzer” tool, a 1/3-octave band analyzer, which provides sound pressure (in decibels, dB) as a function of frequency. The sound pressure distribution was measured for background conditions (hood off) and for each available range hood speed when the house was in quiescent condition, i.e. with no air quality monitoring devices operating. We report the A-weighted total sound pressure (dBA) reported by the app as a summary statistic. Additionally, we applied the procedure that is used to determine the sound level reported by the Home Ventilation Institute (HVI) in their Certified Home Ventilating Products Directory; the calculation procedure is described in HVI Publication 915, available on the HVI web site.³

¹ www.energyconservatory.com.

² www.studiosixdigital.com.

³ www.hvi.org.

Capture efficiency (CE) refers to the fraction of pollutants emitted from the cooking burner (and cooking, when applicable) that are removed by the venting range hood before mixing into the air of the kitchen. CE can be estimated by calculating both the mass flow of CO₂ exiting through the range hood, and the mass generation rate based on fuel composition and the assumption of complete combustion [28]. In this study, we used a simpler approach that compares the flow of CO₂ through the hood under the normal operating condition to the flow of CO₂ when a foil curtain is used to extend the hood over the cooktop to ensure perfect or nearly perfect capture. This approach assumes no change in airflow between the conditions, meaning the CO₂ mass flow changes proportionally with the CO₂ concentration. CE is calculated using CO₂ concentrations measured under the normal operating condition (C_N) and with the hood extended to create nearly perfect capture conditions (C₁₀₀), and background concentrations with the cooking burners off (C₀), as shown in Equation (1).

$$CE = (C_N - C_0) / (C_{100} - C_0) \quad (1)$$

CO₂ concentrations in the exhaust from the range hood were measured using a PPSystems EGM-4 analyzer drawing from the ducting above the range hood. An example of the setup for this procedure, from H8, is shown in Supplemental Fig. S3.

2.6. Attributing concentration profiles to individual cooking events

The primary objective of this study was to quantify the impact of using NGCBs on pollutant concentrations. However, in order to complete as many experiments as possible in the limited time available in each house, the interval between experiments was not long enough to allow concentrations to return to background levels before the next experiment. Therefore the analysis included a *disentanglement* procedure, in which the data for each analyte were decomposed into estimated concentration profiles for each cooking event.

Disentangling the data for a particular cooking event involved four steps: (1) fitting a first-order decay model to the experimental data during each interval between NGCB operation; (2) using this model to predict how the previous cooking event would have evolved, had the current event not taken place; (3) using the same model to predict how the current cooking event would have evolved, had the next event not taken place; and (4) finding the difference between the two, in order to estimate what the current event would have looked like in the absence of other events.

Fig. 1 shows an example. The measured data is shown in solid blue. Here we wished to find the concentration profile for the second of the three cooking events shown (hood and FAU on). We projected the profile (dashed purple line) from the preceding cooking event (hood and FAU off), past the start of the event being analyzed. Similarly, we projected the profile (dashed orange line) of the second event to remove the effects of the following experiment. The difference between the orange and purple projections gives the disentangled profile for second experiment with hood and FAU operating (solid green line).

In selecting intervals for fitting data, we considered the movement of pollutants across zones of the house, as indicated by the dynamics of the two measured locations. In general, we waited until the two series “came together” – ideally by reaching or approaching the same concentration, and alternately by starting to follow the same downward trend. For the distant location, the decay interval often extended past the start of the simulated cooking event.

The decay rate was estimated in a three-step process using a least-squares approach to fit the measured data to a pseudo-first

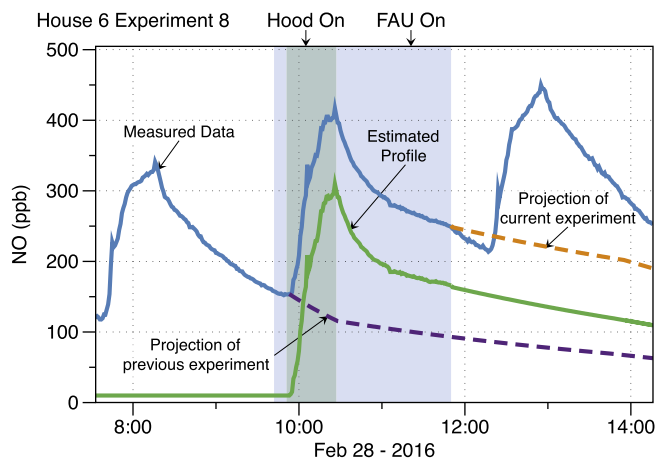


Fig. 1. Disentangling kitchen NO data.

order decay model. The first step was to fit the data to a model that had both the decay rate and the background concentration as variable parameters. The background represents the indoor concentration that would result from outdoor pollutant entry and indoor losses (including air exchange for all pollutants, and deposition for NO₂ and PN). In the second step we estimated a constant background concentration taking into account the estimated background parameters from all experiments that had the same mechanical airflow conditions (e.g. central mixing fan on or off) during the same day as well as any other pertinent concentration measurements (e.g. at the beginning or end of the day). In the third step, we again fit the decay rates to the data, but this time using the estimated constant background concentration. For the experiment shown in Fig. 1, the background was set at 10 ppb and the resulting fit gave a decay rate of 0.17 h⁻¹. The appendices to the project report [27] include plots of measured concentrations for all experiments and show the data intervals that were used to fit the decay rates.

This extrapolation and subtraction procedure was applied separately for CO₂, NO, NO₂, and PN, in the kitchen and bedroom, as data were available. The measurements and extended concentration profiles were analyzed to determine the highest 1 h average concentration of each pollutant at each location, and also to find the integrated concentration over 4 h.

2.7. Estimating emission factors from ambient concentrations

We used the highest 1 h concentrations to estimate fuel-normalized emission factors for NO₂, NO_x, and PN, using the method described previously [25]. Briefly, this approach calculates the emission rate of CO₂ from natural gas combustion, and uses the measured concentration ratios for each pollutant to CO₂ to calculate an emission factor. For NO_x and NO₂, the calculation proceeds according to Equation (2), where the first term on the right side is the ratio of NO or NO_x to CO₂, and the second term is a property of the fuel; we used 1.1 mol CO₂/MJ fuel based on Singer et al. (ibid). By convention, NO_x emissions are calculated using the molar mass of NO₂, 46 g/mol.

$$E_i \left[\frac{ng}{J} \right] = \left(\frac{\frac{mol\ i}{mol\ air}}{\frac{mol\ CO_2}{mol\ air}} \right) \left(\frac{mol\ CO_2}{MJ\ fuel} \right) \left(\frac{g\ i}{mol\ i} \right) \left(\frac{MJ}{10^6 J} \right) \left(\frac{10^9 ng}{g} \right) \quad (2)$$

Equation (3) was used to calculate PN emission factors.

$$E_i \left[\frac{\#}{J} \right] = \frac{\left(\frac{\#}{\text{cm}^3 \text{ air}} \right) \left(\frac{23600 \text{ cm}^3 \text{ air}}{\text{mol air}} \right)}{\left(\frac{\text{mol CO}_2}{\text{mol air}} \right)} \left(\frac{\text{mol CO}_2}{\text{MJ fuel}} \right) \left(\frac{\text{MJ}}{10^9 \text{ J}} \right) \quad (3)$$

3. Results

3.1. Base case measured pollutant concentrations

To elucidate some of the major themes seen in the data, sample results for base conditions with no mechanical ventilation are presented in Fig. 2 and Fig. 3. These plots present data for NO₂, NO, PN, and CO₂, in both the kitchen and bedroom area monitoring locations. Fig. 2 presents data from Day 1 in H3. As expected, kitchen CO₂, NO, NO₂ and PN increased quickly as burners fired at maximum settings: cooktop burners set to the highest flame, or oven or broiler burner firing continuously. Concentrations remained elevated throughout the simulated cooking events as the cooktop burners were adjusted to medium-low, oven burners cycled to maintain temperature, and broiler burners fired continuously. The kitchen traces show more short-term variability owing to their proximity to the source. After the burners were switched off at the end of each simulated cooking event, concentrations in the

kitchen started to decay as pollutants mixed throughout the house and were removed by ventilation and infiltration, and NO₂ and PN were removed by deposition. With each burner use, concentrations in the bedroom started to rise after a short delay, indicating the transport time from the kitchen to the distant location. Bedroom concentrations increased over a time interval that was similar in duration to burner use, but shifted later in time. For NO and CO₂, concentrations decayed slower in the bedroom than in the kitchen, presumably due to lower rates of outdoor air directly entering the bedroom, compared to the kitchen. For PN and NO₂, each of which deposit indoors at rates that are fast enough to compete with air exchange as a removal process, the kitchen and bedroom concentrations decayed at approximately the same rate in the two rooms.

Day 1 in H5 shows a somewhat different pattern (Fig. 3). In this house, the distant bedroom had less indoor air exchange with the kitchen. Following the start of an emission event without the FAU operating, the bedroom concentrations increased more slowly, and reached peak levels that were much lower than in the kitchen. Concentrations in the kitchen and bedroom converged for NO and almost converged for CO₂ only in the first and fourth experiments. Decay rates in the kitchen and bedroom were similar.

The absolute concentrations, relative dynamics and peak concentrations, and the effect of FAU operation on the relative dynamics of pollutants in the kitchen and bedroom varied widely across homes. In general, as the delay increased, so did the difference between the kitchen and bedroom area peak concentrations.

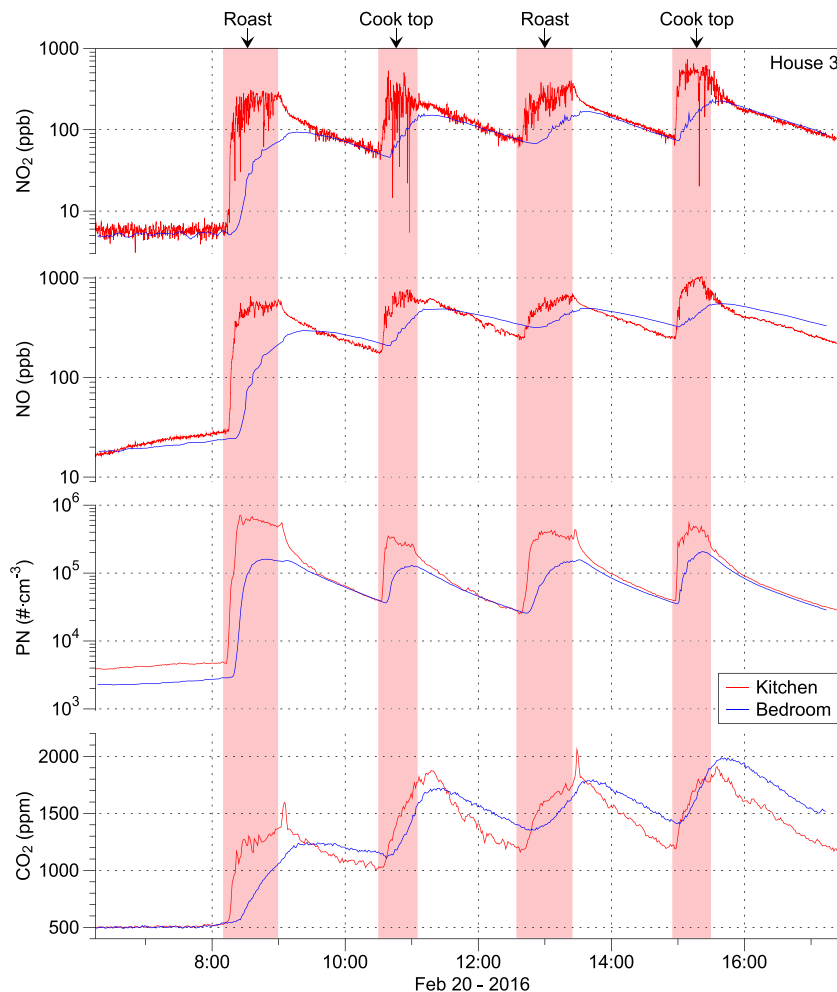


Fig. 2. Air pollutant concentrations on Day 1 in H3.

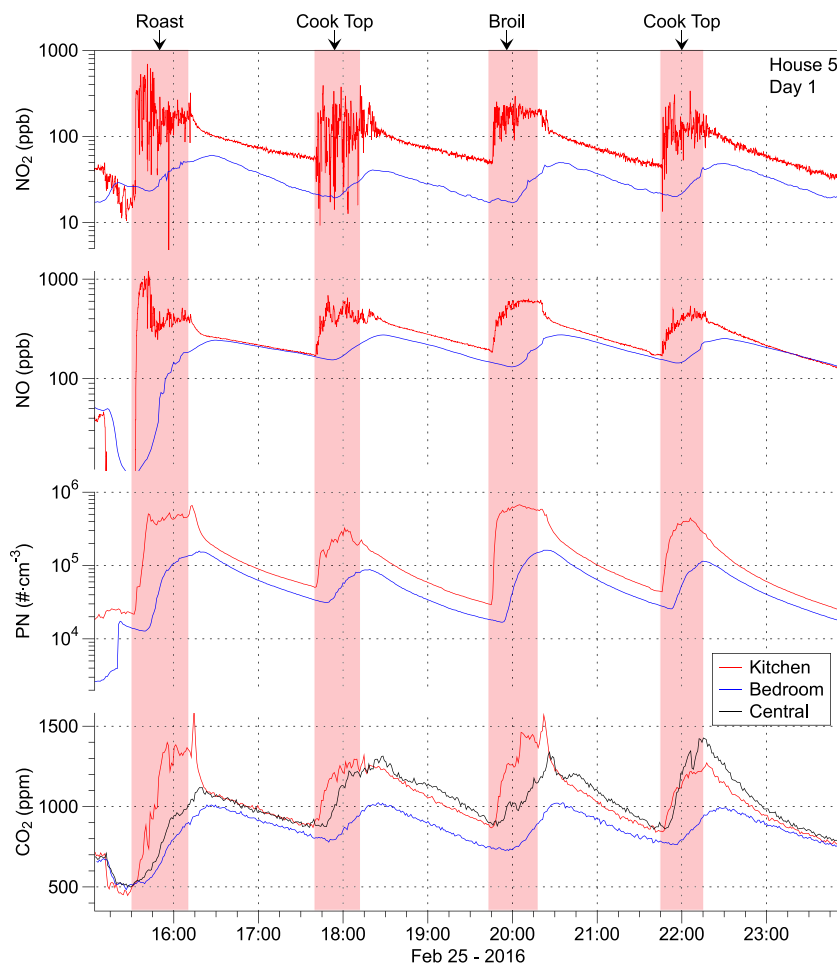


Fig. 3. Air pollutant concentrations on Day 1 in H5.

The closest coupling without FAU use occurred in H4, H7, and H8. The coupling in H4 is explained by it being a small, 2-room apartment. H7 and H8 are newer homes with open floor plans that resulted in closer connections between spaces. FAU operation substantially increased PN decays in H9, which had a high-performance (MERV13) filter installed in the FAU. In the single FAU experiment in H6 (on Day 2), NO₂ decays were much faster than they were in the experiments without the FAU operating; this suggests removal of NO₂ in the air handler.

3.2. Time-integrated pollutant concentrations under base conditions

Summary results for all experiments with base conditions of no mechanical mixing or exhaust (no FAU or range hood use) are provided in Figs. 4 and 5. These figures show the highest 1 h and 4 h time-integrated concentrations in the kitchen and bedroom locations following each simulated cooking event. The homes are grouped according to the opening between the kitchen and the rest of the home, and ordered by increasing home size. Data are presented for NO, NO₂, and PN.

Overall, concentrations of each pollutant varied widely both across and within homes. Concentrations of all analytes were higher in H4, the small apartment, than in other homes. As expected, bedrooms had substantially lower pollutant concentrations than kitchens, across all homes except for the two-room apartment, H4, and the open floor plan house, H8. For NO and NO₂, there was

no obvious trend across homes associated with the burner used in the simulated cooking event (cooktop, oven or broiler). For PN, there were several homes that had much higher concentrations when using the oven or broiler burner compared to the cooktop, but no homes in which the cooktop produced substantially higher PN than the oven or broiler. Unsurprisingly given its very small size, H4 had the highest concentrations of NO and NO₂. That H4 did not have higher PN concentrations than other homes suggests that the variance in PN emission rates had a larger impact than variations in home size. The data also indicate a trend of higher kitchen concentrations in homes with enclosed kitchens (H1, H3, H4, and H5) relative to homes with semi-open kitchens (H6 and H9) or open floor plans (H2, H8, H9). The trio with open floor plans included the two largest homes, which also contributed to the generally lower concentrations observed in those homes.

The plot of highest 1 h concentrations shows the EPA acute threshold concentration of 100 ppb NO₂ over 1 h. Four of the nine homes had kitchen levels exceed this benchmark, and two other kitchens had 1 h NO₂ concentrations of at least half this value. Three of the nine homes had bedroom NO₂ levels exceed 50 ppb. This suggests significant exposures may occur for anyone at home when natural gas burners are used for even a single, substantial cooking event. It is important to note that these concentrations are attributed only to the cooking event; NO₂ entering the home with outdoor air would add to occupant exposure.

For the vast majority of experiments, there were negligible increases in CO and PM_{2.5}. We limited quantitative analysis for these

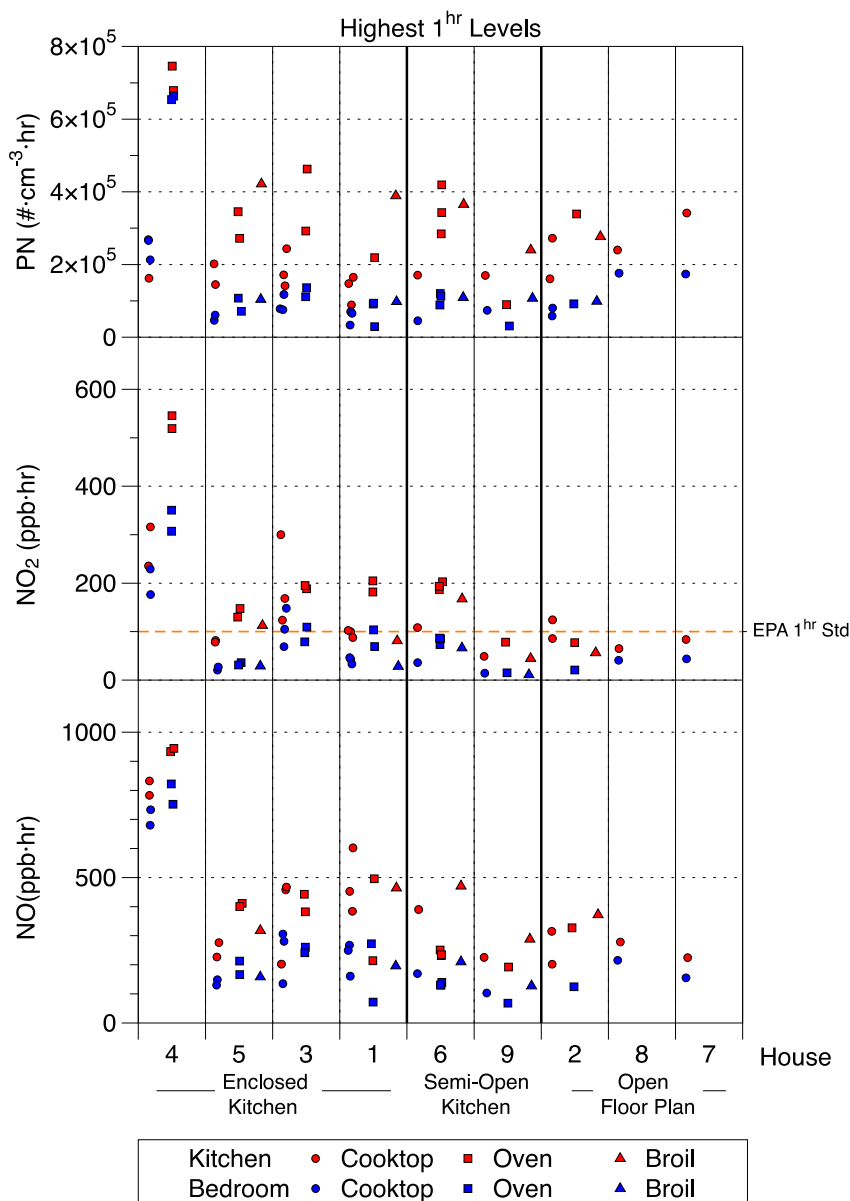


Fig. 4. Highest 1 h time-integrated concentrations in kitchen and bedroom resulting from use of natural gas burners in simulated cooking activities.

pollutants only to those cases in which concentrations were observed to increase by a substantial amount, set arbitrarily as 9 ppm for CO and 20 $\mu\text{g m}^{-3}$ for PM_{2.5}. These thresholds were exceeded only in the kitchen and during only three experiments each for CO and PM_{2.5}. Results for these experiments are provided in Table 5. Elevated PM_{2.5} occurred only with oven/broiler use, not cooktops. The assumption is that the PM_{2.5} was formed from organic deposits in the oven that were volatilized during oven use and condensed into or onto particles.

3.3. Repeatability

Summary results for replicate experiments are provided in Table 6 (Full results are in Supplemental Tables S3 and S4.). Replicates were most consistent for NO and least consistent for PN. For NO, NO₂, and PN, replicates showed similar consistency for 1 h and 4 h results. By contrast, CO₂ had much more variability in 4 h vs. 1 h results.

3.4. Effect of FAU use

Table 7 presents summary results for the experiments in which the air handler of the forced air heating system (FAU) was operated starting approximately 10 min before cooking and through the decay period. Full results are available in Supplemental Table S5. The experiments with FAU use are compared to experiments with the same cooking activities with no FAU use. From the basic physical consideration that it increases mixing, we expect the FAU to reduce kitchen concentrations and increase bedroom concentrations relative to the same cooking activity with no mixing – unless there are losses in the forced air system (e.g. removal by a furnace filter). The trend of a larger increase in the bedroom relative to the kitchen is evident in the highest 1 h CO₂, NO, and NO₂ in two H1 experiments, the experiment in H6, and the CO₂ and NO₂ data in H8. Concentrations in the kitchen either decreased or increased very slightly (which is expected from variability in emissions), whereas concentrations in the bedroom were dramatically higher

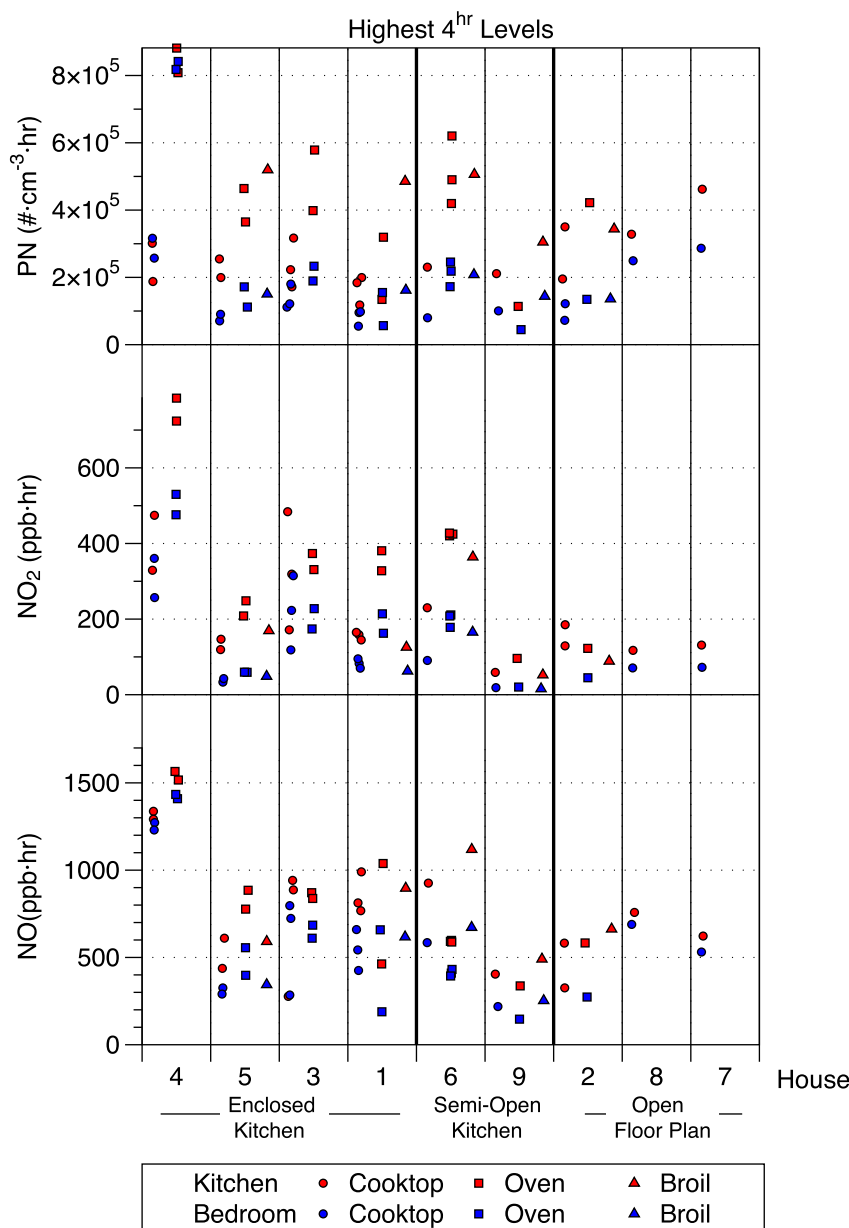


Fig. 5. Highest 4 h time-integrated concentrations in kitchen and bedroom resulting from use of natural gas burners in simulated cooking activities.

Table 5

CO and PM_{2.5} time-integrated concentrations in experiments meeting criteria of CO rising at least 9 ppm and PM_{2.5} rising at least 20 $\mu\text{g m}^{-3}$.

Expt	Burner	FAU	Parameter [units]	Highest 1 h	Highest 4 h
H402	Oven	No	PM _{2.5} [$\mu\text{g m}^{-3}\text{ h}$]	238	378
H404	Oven	No	PM _{2.5} [$\mu\text{g m}^{-3}\text{ h}$]	86	131
H603	Broiler	No	PM _{2.5} [$\mu\text{g m}^{-3}\text{ h}$]	24	53
H402	Oven	No	CO [ppm h]	9.5	21.7
H403	Cooktop	No	CO [ppm h]	8.6	17.7
H405	Oven	No	CO [ppm h]	7.4	11.5

with FAU operation. The PN results for FAU use in H1 don't follow this trend, but in H6 there was still a much larger increase in bedroom PN vs. kitchen PN when the FAU operated. The trend was not apparent in the FAU experiments in H7 and H9. The experiment in H2 had only CO₂ and PN data to compare. Overall, there was not a consistent trend of FAU impacts in this study.

Table 6

Summary Statistics for relative deviations ($n = 2$) and relative standard deviations ($n = 3$) of highest concentrations for replicated conditions.

	CO ₂		NO		NO ₂		PN	
	B	K	B	K	B	K	B	K
<i>1h time-integrated concentrations</i>								
Mean	19%	13%	12%	12%	23%	17%	30%	29%
SD	18%	14%	8%	15%	11%	20%	11%	19%
N	6	7	5	7	5	7	6	7
<i>4h time-integrated concentrations</i>								
Mean	26%	18%	13%	17%	18%	20%	31%	27%
SD	32%	21%	12%	20%	15%	15%	16%	19%
N	6	7	5	7	5	7	6	7

The lack of a clear trend across all the data collected for this study does not mean that FAU use does not impact pollutant spatial

Table 7
Percent change in highest 1 h pollutant concentration from baseline with no forced air to conditions with FAU mixing fan on.

Cooking		CO ₂		NO		NO ₂		PN	
Burner	Experiments	B	K	B	K	B	K	B	K
BR	H102/H113	28%	–15%	13%	–27%	63%	8%	–31%	–45%
OV + CT	H106/H114	19%	1%	16%	–14%	24%	3%	–40%	–32%
CT	H202/H204/H207	24%	6%	–	17%	–	9%	13%	25%
CT	H602/H604	81%	–20%	67%	–18%	64%	–22%	139%	40%
CT	H701/H703	–21%	–5%	–15%	2%	5%	9%	–23%	–29%
CT	H801/H804	–23%	–6%	–24%	–22%	–31%	–7%	–29%	–
OV	H901/H908	–9%	–12%	6%	–16%	37%	–4%	–83%	–67%
CT	H902/H903	34%	19%	33%	22%	29%	42%	–45%	3%

distributions in homes. Rather, the results suggest that the mixing effect was of the same order of magnitude or less than variations caused by other factors, including emission rate and variability of non-mechanical mixing, for the homes studied.

3.5. Spatial variations

The calculated time-integrated concentrations in the kitchen and bedroom were compared for each experiment to quantify the

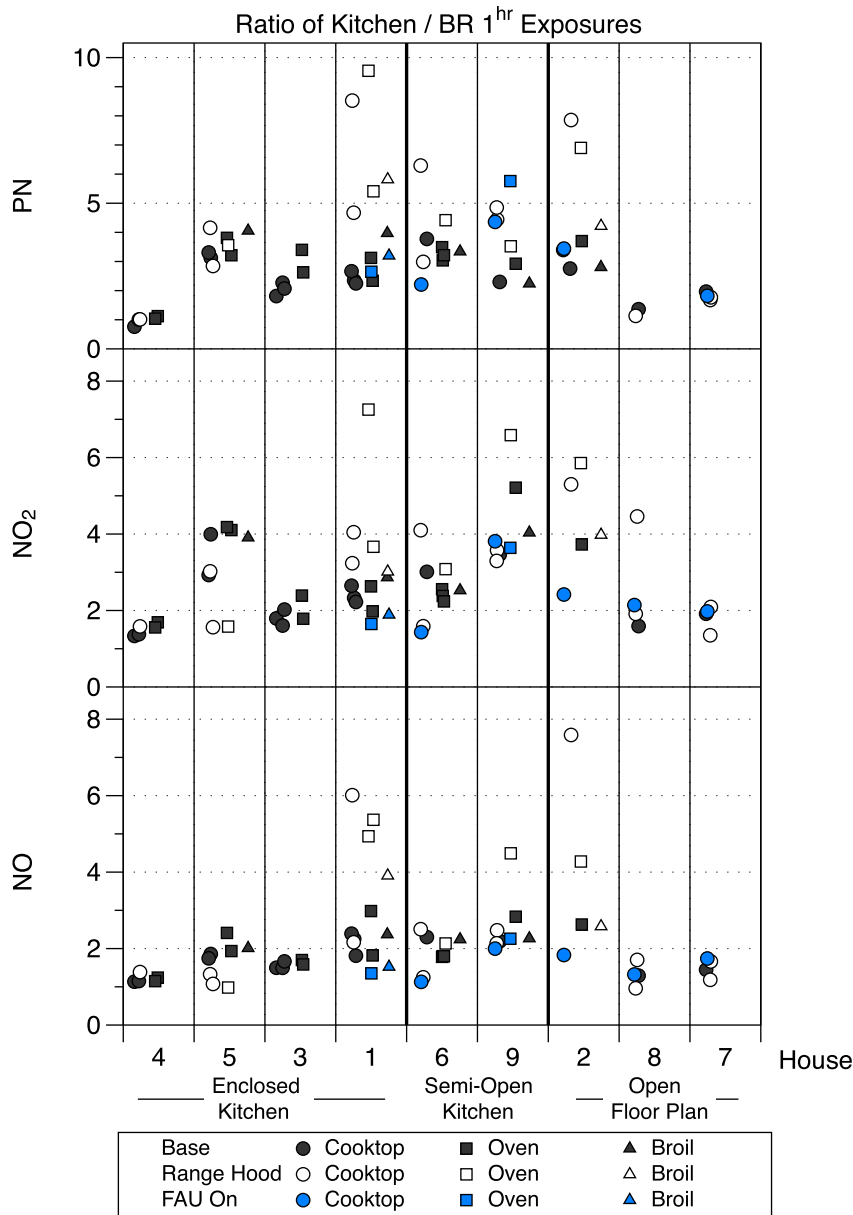


Fig. 6. Kitchen-to-bedroom ratios of highest 1 h time-integrated concentrations.

potential spatial variations in cooking pollutant exposures in homes; results are shown in Fig. 6 and 7. Across the sample, the least pronounced spatial variations occurred in the small two-room apartment (H4) and two of the homes with open floor plans (H8 and H7). Spatial variations of highest 1 h NO in these three homes were almost all in the range of 1.3–2.1 (there was one outlier in H8). The greatest spatial variations for 1 h NO₂ were in H9, the two-story 1990s home retrofitted for deep energy savings: K/BR ratios for NO₂ in this home were 3.3–6.6. For many of the homes, the kitchen to bedroom ratios were somewhat higher for the highest 1 h compared to the 4 h time-integrated concentrations. This is consistent with the difference between the two locations being largest during the period during and just after cooking. Within each home, spatial variations for NO (and CO₂, not shown in the figures) were smaller than for NO₂ and PN. This occurred because NO₂ and PN were removed during transport.

3.6. Emission factors

Calculated emission factors for NO₂, NO_x, and PN are shown in Fig. 8. The emission factors derived from the simulated cooking experiments can be compared with those presented for samples of burners in two earlier reports: (1) a compilation of data from studies completed prior to 1990, mostly from the 1970s and early 1980s [34]; and (2) a study that reported emission factors for previously used appliances including 13 cooktops, 12 oven burners and 6 broiler burners that were first sold in 1992–2007 and tested at ages 2–17 years [25]. Note that NO₂ emission factors from the 25 gas ranges reported by Ref. [16] are included in the compilation [34].

NO_x emission factors determined in the current study ranged from 28 to 64 ng/J, with most of the data between 30 and 45 ng/J, and a geometric mean of 40. These are a bit higher than the

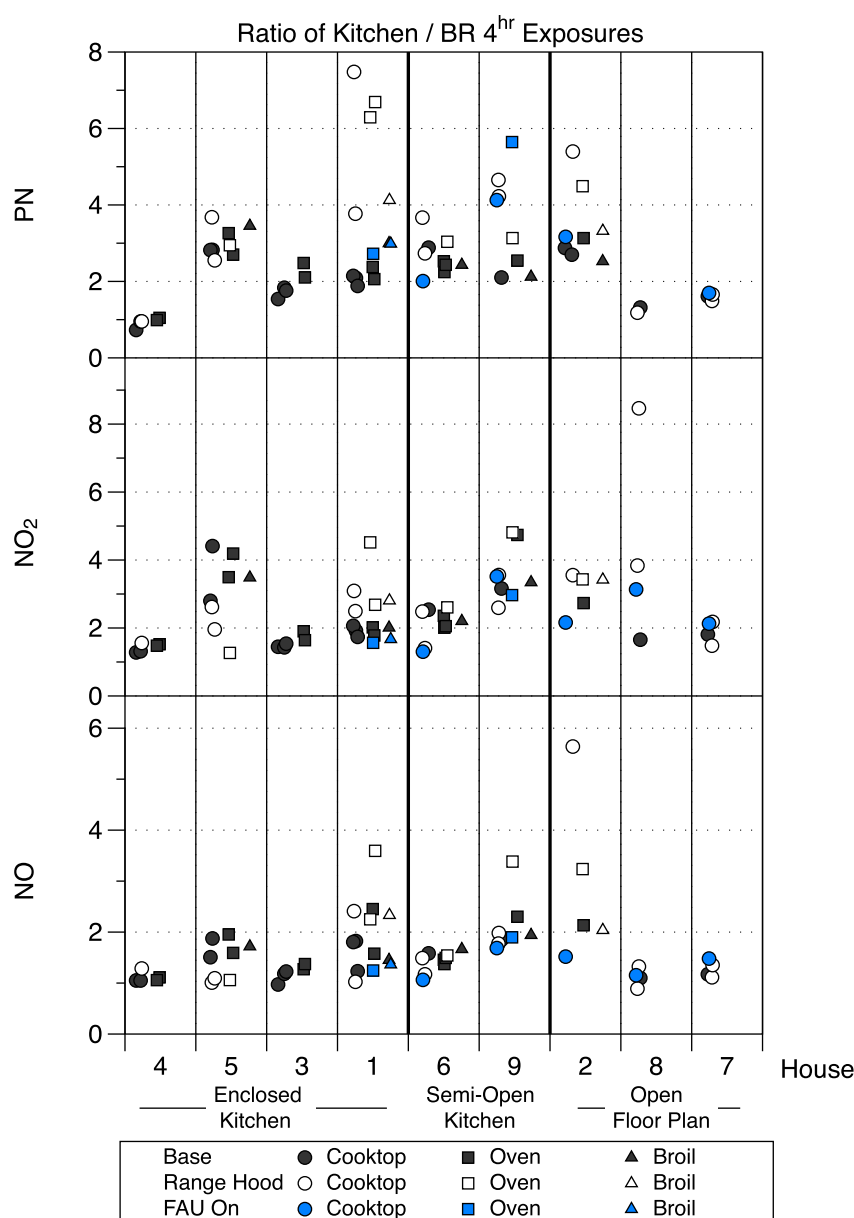


Fig. 7. Kitchen-to-bedroom ratios of highest 4 h time-integrated concentrations.

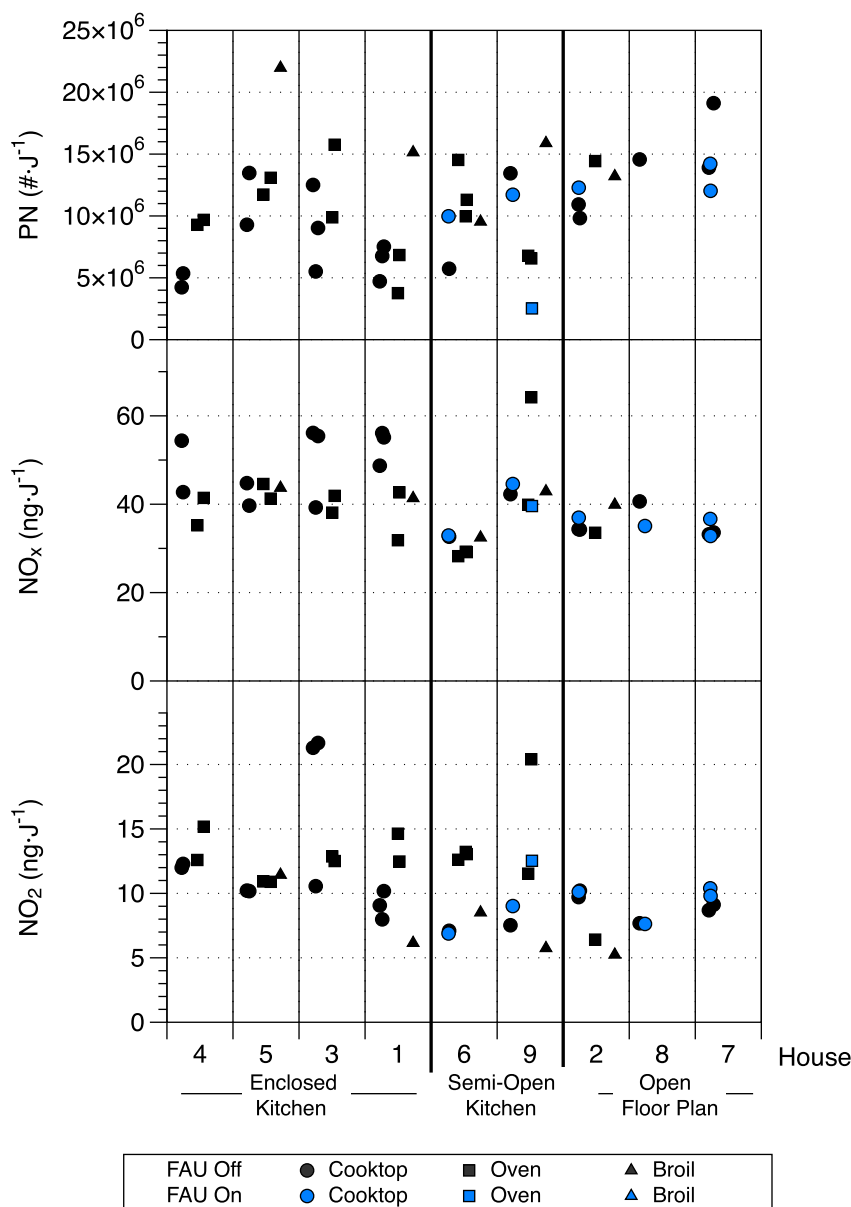


Fig. 8. Emission factors calculated from ratios of highest 1 h NO_2 , NO_x , and PN to highest 1 h CO_2 . Mass emission rates for NO_x calculated using a molecular mass of 46 g/mol by convention.

emission factors reported by Singer et al. [25], which ranged from 17 to 47 ng/J with most of the data between 30 and 36 ng/J, and the results reported by Traynor et al. [34], which had a geometric mean of 32 ng/J. Most of the NO_2 emission factors calculated in the current study were between 5 and 15 ng/J, with a geometric mean of 10.3 ng/J. The prior study [25] reported similar results with 28 of the 31 burner sets having NO_2 emissions within the range of 5–15 ng/J.

The biggest difference between the current results and previously reported emission factors is for PN. In this study, the calculated PN emission rates ranged from $2.5 \times 10^9 \text{ J}^{-1}$ to $2.2 \times 10^{10} \text{ J}^{-1}$ with a geometric mean of $1.0 \times 10^{10} \text{ J}^{-1}$. In the prior study [25], PN emissions were much more variable. The highest emission factors for each burner type were in the same range as those determined in the current study: 1.4×10^{10} , 0.5×10^{10} , and $2.6 \times 10^{10} \text{ J}^{-1}$ for CT, OV and BR burners respectively. But for each burner type reported in the earlier study, there were many more burners with PN emission factors below the lowest values reported for the current study. This

difference may result from the cleaning, pre-conditioning, and more repeat experiments in the earlier study. This hypothesis is based on the idea that particles were formed by volatilization of organics compounds deposited on cooktop, oven or broiler burner surfaces, as reported in a study or particle emissions from electric burners and hot surfaces [40].

3.7. Measured range hood performance

Measurements of range hood performance parameters are presented in Table 8. Consistent with a prior in-home study [28], the measured airflows were substantially below rated values for five of the six installed hoods. Interestingly, the estimated sound ratings (in sones) were lower than the rated values for many of the hood settings.

Table 9 presents the estimated capture efficiencies measured with the new field test method described in Section 2.5. Consistent with a prior field study (ibid), the performance of several of the

Table 8
Measured performance parameters of range hoods in study homes.

Home ID	Type	Measured flows [L/s] (% of rated flow)		Measured sound [dBA] (Calculated Sones)		Bkg [dBA]
		Low speed	High speed	Low speed	High speed	
H1	Hood	66 ^b	148 (59%)	57.3 (3.0)	59.2 (3.7)	30.1
H2	Microwave	66 (78%)	76 (54%)	63.9 (4.6)	72.1 (6.2)	36.8
H5	Hood	135 (98%)	153 ^b	58.9 (4.0)	66.7 (4.8)	28.9
H6	Microwave	43 ^b	49 (45%)	59.2 (3.6)	62.5 (4.6)	30.7
H8 ^a	Hood	20 (40%)	30 (40%)	54.0 (2.2)	58.2 (3.7)	32.9
H9 ^a	Hood	39 (79%)	19 (64%)	54.1 (2.0)	61.4 (3.7)	39.8

^a The hoods in H8 and H9 are the same product, sold under different nameplates.^b Performance information not provided with product literature.**Table 9**
Measured capture efficiency of range hoods in study homes.

Home ID	Hood type	Low speed		High speed	
		Front burners	Back burners	Front burners	Back burners
H1	Hood	NM ^a	NM ^a	NM ^a	NM ^a
H2	Microwave	25%	>95%	35%	>95%
H5	Hood	61%	68%	72%	84%
H6	Microwave	31%	88%	31%	93%
H8 ^a	Hood	59%	68%	65%	80%
H9 ^a	Hood	25%	74%	36%	75%

^a Not measured; there was no way to access the range hood exhaust duct without aesthetic damage.

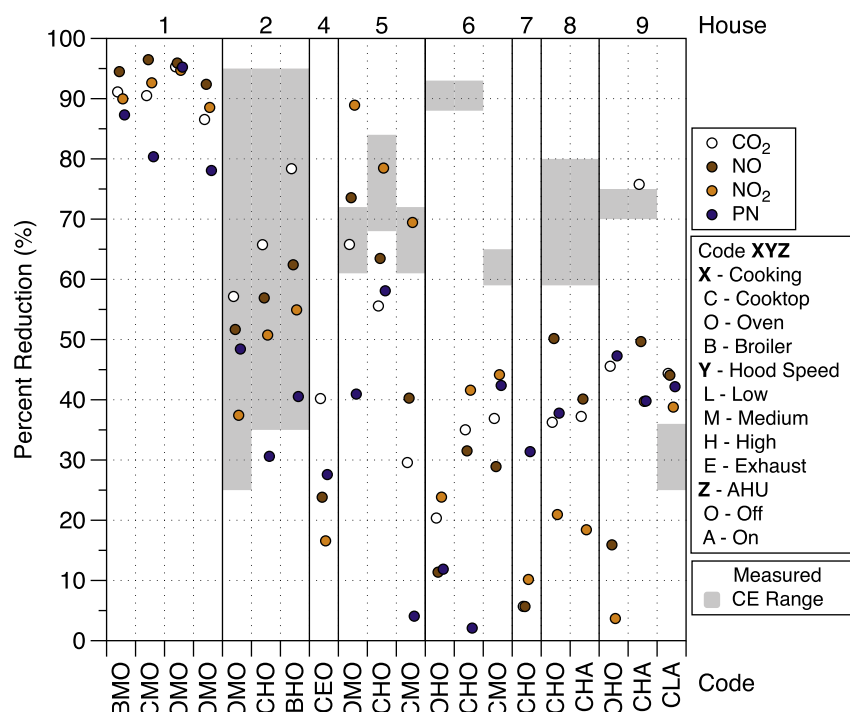
hoods was dramatically different for the front and back cooktop burners. For most ranges, the performance of the back cooktop burners is a good indicator of capture for oven emissions.

3.8. Effect of range hood use

The effects of operating a venting range hood during cooking are presented in Fig. 9. This figure presents the percentage reduction in

the highest 1 h concentration, calculated as the difference between experiments with range hood use and analogs without range hood use. Included in this figure are the calculated reductions from using the bath fan as the only available exhaust device in H4, and the recirculating range hood in H7.

Broadly, these results indicate that use of range hoods can yield substantial reductions in cooking burner pollutant concentrations both in the kitchen and throughout the house. The impact of using a

**Fig. 9.** Percent reductions in highest 1 h kitchen concentrations calculated by comparing experiments with range hood use to analogous experiments without range hood use. Shaded grey areas show the range of capture efficiencies measured for the hood at the flow rate used in the experiment, as presented in Table 9.

venting range hood was larger than the variability of the experimental method, producing net reductions in all cases. The most benefit was seen in H1, which had a range hood with large capture volume and a measured airflow of 108 L/s. This hood, which produced reductions mostly in the range of 80–95%, was of similar design to hoods that showed very high capture efficiency in prior studies [3,28]. The next most effective hood, in house H5, also had characteristics shown to enhance performance [28]: it fully covered the front burners and had airflow substantially above the benchmark of 95 L/s.

The only other hood with reductions mostly exceeding 50% was the over-the-range microwave in H2, which had an exhaust fan that moved 76 L/s at high speed. Prior studies [3,15,28] have found over the range microwave exhaust fans to vary widely in their capture efficiency, in large part explained by variations in airflow but likely also relating to disadvantageous geometry. The reductions observed for H5 were consistent with those measured for hood H2 operating at similar airflow in Singer et al. [28]. The range hoods in H6, H8 and H9 had calculated reductions of 0–50% with more than half of the results falling in the top half of that range. These hoods had lower airflows and did not cover front burners fully. The bath fan in H4 reduced concentrations by 15–40% across the measured species, suggesting a modest benefit that could have been caused by method variability. The recirculating range hood in H7 showed small net reductions ($\leq 10\%$) for NO₂, NO, and CO₂, and a larger reduction for PN ($\sim 30\%$). All were within the variability of replicate experiments.

The effects of airflow, burner position, and particle size on range hood effectiveness for PN have been discussed [15,20]. In measuring particles down to 2 nm, Rim et al. found that removal effectiveness was lower for 2–6 nm particles than for particles > 6 nm. Since a large number of particles are in the lower size range, that effect is expected to reduce the overall effectiveness reported by Rim et al. relative to what would be reported for 6 nm and larger particles, as measured in this study. The “A” hood in the Rim et al. study was similar in design and airflow to the hood in H8 and H9 in this study. The roughly 40% reductions in cooktop-emitted PN calculated with range hood operation in those homes is roughly midway between the 31 (6%) for front burners and 54 (9)% for back burners reported by Rim for Hood A. The “B” hood tested by Rim et al. was similar in design to the hood in H5 in this study and the measured airflows of the hood in H5 were between the medium and high flows measured for Hood B in Rim et al. Yet the effectiveness for PN was substantially lower in H5 than reported by Rim et al. for their Hood B. The apparent difference may simply result from the greater variations in PN associated with the in-home experiments conducted in this study.

4. Concluding recommendations

Based on the findings of this field study and the related, prior work referenced herein, the authors offer the following policy recommendations. Efforts should be made to increase awareness (a) that natural gas cooking burners are a source of air pollutant emissions into homes, and (b) that these pollutants can be controlled with an appropriately-sized venting range hood or other kitchen exhaust ventilation.

Building standards should require that range hoods have airflows of at least 95 L/s and cover front burners or preferably demonstrate performance through a standard test [37]. Since the performance of most hoods is much better when cooking on the back cooktop burners, this practice should be encouraged to improve safety. Since cooking with electric burners also produces pollutants, kitchen exhaust ventilation should be available in all homes, and operated as a precaution whenever cooking occurs.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.buildenv.2017.06.021>.

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Composition, Emissions, and Air Quality Impacts of Hazardous Air Pollutants in Unburned Natural Gas from Residential Stoves in California

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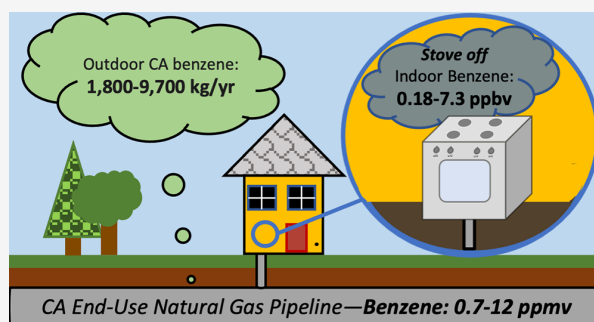
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ABSTRACT: The presence of hazardous air pollutants (HAPs) entrained in end-use natural gas (NG) is an understudied source of human health risks. We performed trace gas analyses on 185 unburned NG samples collected from 159 unique residential NG stoves across seven geographic regions in California. Our analyses commonly detected 12 HAPs with significant variability across region and gas utility. Mean regional benzene, toluene, ethylbenzene, and total xylenes (BTEX) concentrations in end-use NG ranged from 1.6–25 ppmv—benzene alone was detected in 99% of samples, and mean concentrations ranged from 0.7–12 ppmv (max: 66 ppmv). By applying previously reported NG and methane emission rates throughout California's transmission, storage, and distribution systems, we estimated statewide benzene emissions of 4,200 (95% CI: 1,800–9,700) kg yr⁻¹ that are currently not included in any statewide inventories—equal to the annual benzene emissions from nearly 60,000 light-duty gasoline vehicles. Additionally, we found that NG leakage from stoves and ovens while not in use can result in indoor benzene concentrations that can exceed the California Office of Environmental Health Hazard Assessment 8-h Reference Exposure Level of 0.94 ppbv—benzene concentrations comparable to environmental tobacco smoke. This study supports the need to further improve our understanding of leaked downstream NG as a source of health risk.

KEYWORDS: BTEX, benzene, downstream, fossil fuels, natural gas leak, cooking, hazardous air pollutants, indoor air quality, regional BTEX inventories



INTRODUCTION

In 2021, the United States consumed approximately 30.2 trillion cubic feet of processed natural gas (NG), comprising 32% of total U.S. energy consumption, of which 15% was consumed by an estimated 74.6 million residential households.^{1–3} Non-methane volatile organic compounds (NMVOCs) are naturally present in unprocessed NG;^{4–9} however, very little data exists on the chemical composition and concentrations of hazardous air pollutants (HAPs) in processed NG at the point of end use.¹⁰ Moreover, the indoor air quality impacts from fugitive NMVOC or HAP emissions associated with known NG leakage or incomplete combustion pathways are understudied, in part, due to a lack of chemical characterization of downstream NG.¹¹

While NG transmission pipelines have some NMVOC restrictions to prevent operational failures (see Federal Register Vol 71 No 120), there are no corresponding limits on HAPs in transmission pipeline gas in the United States.^{12–16} Many NMVOCs are hazardous to human health and therefore have been deemed HAPs under the U.S. Clean

Air Act, which is further exemplified by various state, federal, and international health-based guidelines for exposure to these compounds.^{17,18} For example, NMVOCs contribute to the formation of ozone, a constituent of photochemical smog, and fine particulate matter (PM_{2.5}).^{19–21} Benzene is a known human carcinogen that increases the risks of developing leukemia.²² One study detected benzene, toluene, ethylbenzene, and xylenes (BTEX) and other health-damaging NMVOCs in downstream NG;²³ however, these data were not quantitative and only included two samples. Recently, Michanowicz et al.²⁴ found extensive NMVOCs in distribution NG in Massachusetts; however, measurements were confined to a region that is geographically far from hydrocarbon

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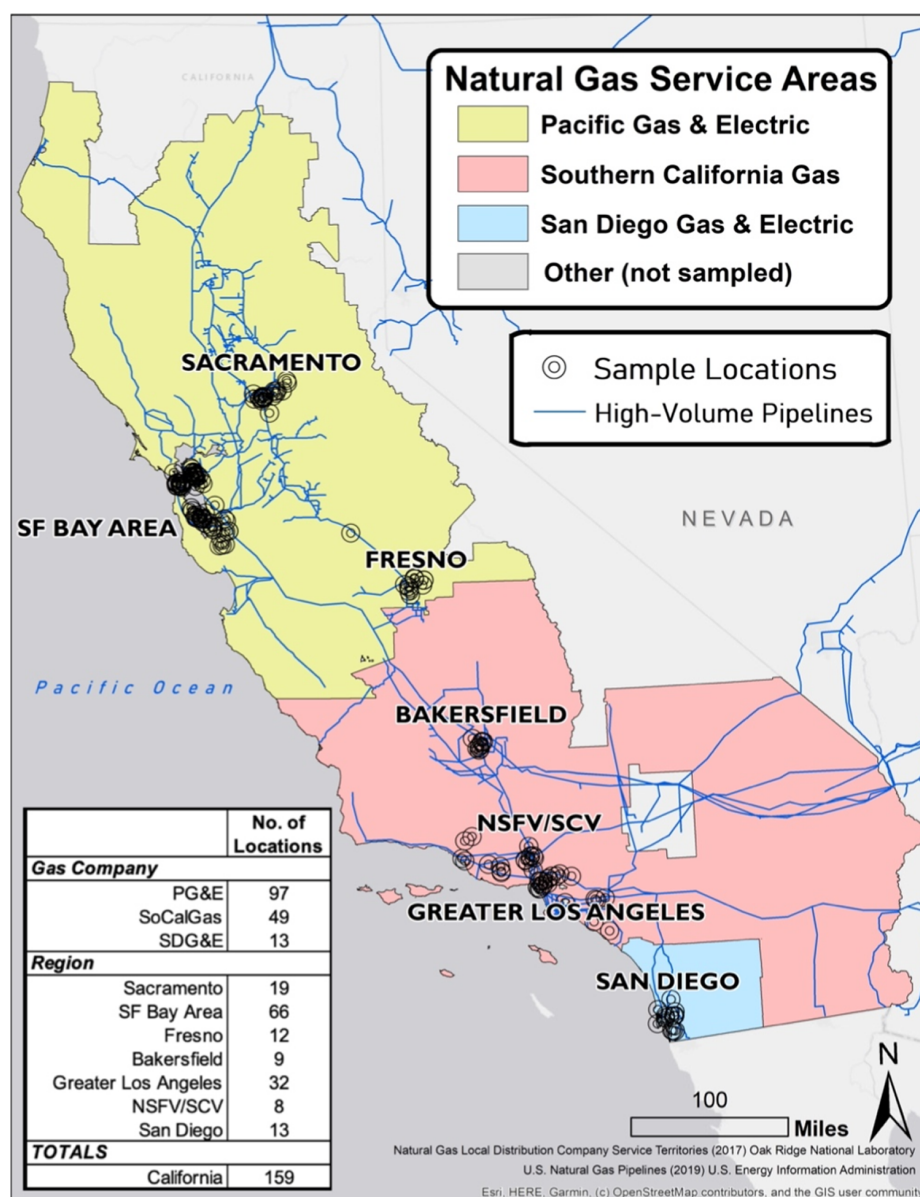


Figure 1. Spatial distribution of the 159 households sampled in California. The points show the location of each of the samples. The shading shows gas utility service territories in each region, and the blue lines represent high-volume pipelines traversing California and neighboring states. Measurements were taken in the following gas companies' territories: Pacific Gas and Electric (PG&E)—yellow; Southern California Gas Company (SoCalGas)—red; and San Diego Gas & Electric (SDG&E)—blue. Samples were grouped into one of 7 different regions which we used throughout this study: San Francisco (SF) Bay Area, Sacramento, Fresno, Bakersfield, North San Fernando/Santa Clarita Valleys (NSFV/SCV), Greater Los Angeles (excluding NSFV/SCV), and San Diego.

production or underground NG storage in depleted oil reservoirs—two features present throughout California that may impact NMVOC concentrations in NG. There are also limited publicly available NMVOC composition data for the transmission and storage sector,²⁵ and it is unclear the degree to which these trace gases persist in the end-use gas distribution system.

Residential NG saturation in California is the second highest in the United States, with 88% of all households (11.5 million in total) having NG service in 2020. Typically, 70% of these homes cook with an NG-powered stove or oven, representing the highest percentage of gas stove users in the country.¹ Driven in part by concerns regarding the climate-forcing nature

of methane, multiple studies in California have quantified NG emissions from buildings and appliances. Fischer et al.²⁶ measured NG appliance leakage in 75 homes in California and found that chronic, low-level leaks can persist undetected directly in residential homes and that 15% of total NG methane emissions in California could be attributed to fugitive post-meter (i.e., in-home) emissions. Lebel et al.^{27,28} observed that at least 75% of total emissions from gas stoves and storage water heaters originate while they are off.^{27,28} These leakage profiles, in combination with uncertainties related to the effectiveness of NG odorants as exposure deterrents,^{24,29–34} support the need for further study of potential risks of exposure

to health-damaging compounds contained in end-use NG in California.

In this study, we adopted canister collection methods with gas chromatography–mass spectrometry (GC–MS) NMVOC analysis from Michanowicz et al.²⁴ to determine the chemical composition of unburned NG from downstream NG supply in seven distinct geographic locations in the state of California.²⁴ Our goals for studying the gas stream at the point of end use were twofold: (1) To understand the contribution of NMVOCs to urban air quality associated with leakage throughout the downstream NG supply chain, and (2) To understand potential indoor exposures from known leakage pathways. To begin to address goal 1, we combined these NG composition data with existing California-specific NG leakage data to estimate unaccounted-for BTEX emissions for the entire state and for certain individual source types within the downstream infrastructure. To address goal 2, we estimated indoor residential ambient benzene concentrations attributable to published rates of NG leakage from household gas stoves and ovens while off across various scenarios.^{35,36} We compared these indoor air concentrations of benzene to the California Environmental Protection Agency Office of Environmental Health Hazard Assessment (OEHHA) benzene Reference Exposure Level (REL) to evaluate health risks from this understudied environmental exposure.³⁷

METHODS

Data Collection. We collected 185 unburned NG samples from 159 unique residential NG stoves in California. We used passivated 1 L SUMMA or Silonite-lined Entech canisters with Teflon tubing to form a direct in-line connection with a single-stove burner NG outlet orifice. NG samples were analyzed by two labs based in California with Environmental Laboratory Accreditation Program (ELAP) certification. We used two labs to accommodate canister availability (Lab 1: $n = 54$; Lab 2: $n = 131$).

NMVOCs for all samples were analyzed using U.S. Environmental Protection Agency Method TO-15, which calls for sample separation and analysis by gas chromatography and mass spectroscopy.³⁸ Samples were analyzed by the lab within 30 days of collection—the maximum hold time specified by the TO-15 method during which concentrations are considered stable. The TO-15 analyte list included 69 compounds from Lab 1 and 63 compounds from Lab 2. We quantified a total of 76 unique compounds using TO-15 across the two laboratories, including 56 compounds analyzed by both labs (see Table S1 for a complete list). Each sample was also tested using Method ASTM D1946 to quantify methane and ethane content in unburned NG as well as elemental nitrogen and oxygen for all but 27 samples.³⁹ These data were used for quality control to ensure pure NG sample collection and to verify minimal ambient air intrusion during sampling.

We collected samples using a similar methodology as described in detail in Michanowicz et al.²⁴ and briefly summarized here. First, we removed gas stove flame spreader plates and metal grates to expose the NG outlet orifice. We then applied Teflon-lined tubing of 1/4" inner diameter to tightly enclose the NG outlet orifice, bypassing any ignition sources and minimizing the potential for NG leakage or ambient air entrainment. We then turned the gas flow knob between medium and high for the entirety of the sampling event. After flushing the hosing, we immediately connected it to the sampling canister provided by the lab via a cone washer

and screw nut or quick release connection. We filled canisters until the vacuum on the canister was between -5 and 0 in Hg, taking approximately 20–30 s. For safety, we deployed a Bascom Turner Gas Rover or similar device near the stove to ensure the concentration of leaked NG did not approach the lower explosive limit of methane (5% v/v gas), and we turned on range hoods when present during and after the sampling process to reduce indoor air concentrations of NG.

We intentionally designed this measurement campaign to sample NG in major urban areas serviced by the three main NG utilities in California: Pacific Gas and Electric (PG&E), Southern California Gas Company (SoCalGas), and San Diego Gas & Electric (SDG&E). In all, we collected samples from 16 counties across California from these three utilities and grouped samples into seven regions we defined: the San Francisco Bay Area (San Francisco, San Jose, Oakland, and nearby cities); Sacramento and Fresno [PG&E]; Greater Los Angeles; the North San Fernando and Santa Clarita Valleys; and Bakersfield [SoCalGas]; and San Diego [SDG&E]. We collected samples between February and September 2021 (Figures 1 and S2).

Quality Control and Data Analysis. Data from our 185 samples are reported as pure gas mole fractions (concentrations) in parts per billion by volume (ppbv). Lab 1 reported directly in ppbv, while Lab 2 required conversion from mass concentrations to volumetric concentrations (ppbv) using the molecular weight of the compound and assuming normal temperature (298 K) and pressure (101.3 kPa). For values below the lab's internal limit of detection, we took a conservative approach and assigned these data as zero.⁴⁰

We took several quality control measurements throughout the study, in addition to blanks and other quality checks performed by the labs. First, to compare our NG samples to background air collected by the same method, we collected one field blank by attaching the hosing to the stove and not turning the gas on and one ambient air blank by opening the can in the kitchen space with no hosing attached, both of which did not detect target analytes. Second, we used both methane and elemental nitrogen (N_2) as source signature tracers—methane as proxy for NG, and nitrogen as proxy for ambient air. Using these fixed gas measurements, we conducted a k-means cluster analysis to exclude samples that entrained a substantial amount of ambient air. Based on our analysis, we identified two k-clusters and excluded the cluster with the lowest methane and highest nitrogen values (Figure S1). To control for samples that did not have a paired nitrogen, nine samples were additionally excluded that had a methane measurement within the range of the excluded k-cluster. The mean of benzene and total BTEX of the excluded samples were less than the mean of the included samples; this is unsurprising, given the assumed dilution effect from ambient air intrusion (Figure S1).

We also collected a series of samples to quantify variability across our study. These included duplicate samples (collected consecutively at a single location) analyzed by different labs; duplicate samples analyzed by the same lab; and repeated samples that were collected in the same households several months apart (Note S1). In our analysis, duplicates collected on the same day were treated as replicate samples; therefore, we selected only the first of the two samples to be used in our analyses. For the times we collected repeat samples in different months, we treated them as independent samples for regional

and statewide means as household-level factors should not influence gas composition.

Regional and Statewide Emissions Estimates. To evaluate the contribution of downstream NG emissions to regional and statewide benzene and BTEX budgets, we used methane emission estimates from previous studies of the downstream NG sector in California to scale bootstrapped regional and statewide mean concentrations of benzene and total BTEX measured in our study.^{26,28,41–44} Emissions rates of benzene and total BTEX were calculated using the following equation adapted from Marrero et al.⁶

$$E_{\text{NMVOC}} = E_{\text{methane}} \times C_{\text{NMVOC}} \left(\frac{MW_{\text{NMVOC}}}{MW_{\text{methane}}} \right) \quad (1)$$

where E_{NMVOC} represents the NMVOC mass flow rate (kg yr^{-1}); E_{methane} represents the methane mass flow rate (Gg yr^{-1}) derived from existing literature;^{26,28,41–44} C_{NMVOC} represents the state or regional concentration of NMVOCs in the pipeline NG (ppmv); and MW_{NMVOC} and MW_{methane} represent the molecular weights of the NMVOC and methane, respectively (g mol^{-1}). This relationship assumes that the concentration of methane in pipeline NG is 100%, and as such, we are slightly underestimating total NMVOC emissions. Scaled benzene emissions (E_{NMVOC}) assume a benzene-to-methane ratio that is conserved throughout the distribution pipeline network and likely underestimates NMVOC emissions using methane flux rather than NG flux. The emissions estimates also assume no differential mechanisms of transport and fate between methane and benzene emissions (e.g., soil adsorption)—a limitation of this methodology.

Regional and distribution pipeline methane emissions were taken from available data sources in the literature to scale our NMVOC emissions. NMVOC emissions for the North San Fernando and Santa Clarita Valleys specifically were estimated using distribution pipeline NG emissions from SoCalGas for zip codes in this region (Figure S4) and NMVOC measurements from our samples in the North San Fernando and Santa Clarita Valleys.

Indoor Air Quality Modeling. To better understand the potential indoor air quality implications of benzene content in end-use NG leaked from gas stoves while turned off, we modeled ambient benzene concentrations in the kitchen and compared the results to California's Reference Exposure Levels (REL). We modeled 140 scenarios that varied unburned NG leakage rates (median: 24 mg hr^{-1} and 95th percentile: 282 mg hr^{-1} , from Lebel et al.²⁸), benzene concentrations in unburned gas (median and 95th percentiles calculated from this study), and household parameters (Table S2). We used CONTAM—a multizone, whole-building model developed by the National Institute of Standards and Technology—that can be used to estimate contaminant concentrations in the indoor environment using information about the structure, ventilation, airflow, and contaminants.^{35,36,45}

Here, as an introductory assessment of the impact of leaked NG on indoor air quality, we conservatively focused only on leakage from a single gas stove/oven while off (steady-state off) and for only one pollutant: benzene. This modeling approach almost certainly underestimates the true quantity of behind-the-meter NG emissions which also may include leaks from other gas appliances and emissions of natural gas when the stove/oven is on.

We calculated benzene steady-state-off leakage rates from stoves using the following relationship

$$V_{\text{benzene}} = V_{\text{methane}} \times C_{\text{benzene}} \quad (2)$$

where V_{benzene} represents the volumetric emissions of benzene from the leaking stove (nL hr^{-1}); V_{methane} represents the volumetric methane emissions rate from the leaking stove (mL hr^{-1}); and C_{benzene} represents the concentration of benzene in the the pipeline gas (ppmv), again assuming that the concentration of methane in pipeline NG is 100%. This enables us to directly convert the volumetric flow of methane, V_{methane} , to a volumetric flow of benzene, V_{benzene} , at a given concentration of benzene. Data on methane emission rates from leaking stoves were obtained from Lebel et al.,²⁸ who measured methane emission rates from 53 gas stoves in California using a chamber-based approach. Benzene concentrations are from this study. This relationship assumes that the emission rate of methane is equivalent to the emission rate of NG. This is a conservative assumption as distribution system NG was measured to be 90–95% methane in this study (Figure S5).

We modeled a range of household parameters and emissions rates; a complete list is in Table S2. We chose a range of households that reflects California's housing stock from the 2019 Residential Appliance Saturation Survey.⁴⁶ This included a mobile/manufactured home, an attached home, a detached single-family home, and a six-unit apartment building. Building designs were selected from the "Suite of Homes Representing the U.S. Housing Stock."^{47,48} Within each household type, we varied the air change rate, the methane emission rate, and the benzene concentration in the pipeline gas. For comparison's sake, we used only two air change rates—the buildings' natural ventilation air change rate (i.e., infiltration only) and the minimum recommended air change rate set by the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE 62.2). The ASHRAE standard was achieved by parameterizing HVAC system settings to input the appropriate volume of air changes per hour. The methane emission rate and benzene concentration rate were used together to calculate the benzene emissions rate via eq 2 (Table S2). All simulations were run under steady-state airflow conditions at normal temperature (298 K) and pressure (101.3 kPa).

Across our simulations, we assumed that benzene was not reactive because the atmospheric lifetime of benzene is several days and the indoor concentrations of the hydroxyl radical ($\cdot\text{OH}$) are similar to outdoors.^{49,50} Following previous studies, we also assumed that surface deposition would not substantially impact indoor concentrations.^{51,52} We assumed that there was no demand-controlled mechanical kitchen hood active during simulations, since our source was only fugitive emissions while off.

■ RESULTS AND DISCUSSION

Overview. We collected 185 individual samples of unburned NG from 159 unique residential stoves across seven distinct geographic locations spanning 16 counties throughout California (Figure 1). After eliminating blanks, duplicate samples, and low-methane samples that did not meet QC standards (see Materials and Methods), we retained 160 samples for analysis. Of the 76 NMVOCs analyzed, 21 unique constituents were detected in end-use NG in California (see Table S1 for a full list of pollutants). Of these, 12 carried a

HAP designation and therefore are known to cause short-term and chronic adverse health impacts.⁵³ Six NMVOCs were detected in more than 98% of NG samples, four of which are HAPs: benzene, toluene, m,p-xylene, and hexane (Figure 2).

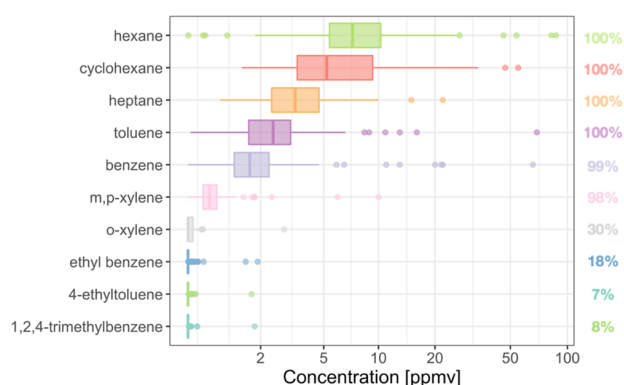


Figure 2. Summary of the top 10 non-methane volatile organic compounds (NMVOCs) detected in residential NG. Percent of values above the level of detection are listed on the right side of the figure. Note: the x-axis is a “pseudo-log” scale, which smoothly transitions to a linear scale around zero.

Median concentrations of BTEX were all within 0.1 ppmv of the means in the following regions: Greater Los Angeles, Bakersfield, Fresno, and Sacramento. This suggests regionally normal distributions of BTEX, further supported by a Kolmogorov–Smirnov test for benzene in these regions ($p > 0.05$; Figure 3 and Table S1). Notably, samples taken in the North San Fernando and Santa Clarita Valleys were not normally distributed and contained several very high benzene concentrations, including the maximum benzene concentration observed in the study—66 ppmv—which is approximately 66 times greater than the highest benzene level recorded in end-use NG in Massachusetts (max: 1.0 ppmv, mean: 0.17 ppmv).²⁴

Benzene and BTEX by Gas Company. We compared the chemical composition of NG across all three major gas utility territories in California: PG&E, SoCalGas, and SDG&E (Figures 1, 3, S2, and S3 and Table S3). Total BTEX content differed significantly by gas company (Kruskal Wallace: $\chi^2 = 25.5$, $p < 0.0001$, $n = 3$), driven in part by the significantly lower BTEX observed in the SDG&E territory compared to PG&E ($p < 0.0001$) and SoCalGas ($p < 0.0001$) territories (Wilcoxon Rank Sum Test). Samples taken in SoCalGas service areas showed the highest variability and the highest overall concentrations—over 22% of samples in SoCalGas were above the maximum benzene concentrations of 3.8 ppmv from PG&E and 3.5 ppmv from SDG&E. Differences in benzene and BTEX concentrations between SoCalGas and SDG&E were particularly unexpected, given that SDG&E is a wholesale customer of SoCalGas and purchases NG directly from SoCalGas for distribution.^{54,55} Further, both utilities reported sourcing NG from similar distribution pipeline systems and underground NG storage facilities (Note S2 and Figure S4), with the majority of gas originating from either the El Paso Pipelines sourced from basins in the Southwestern United States (i.e., the San Juan, Permian, or Anadarko Basins) or the Kern River Pipeline that sources NG from the Rocky Mountain Basin.⁵⁶ A fraction of gas comes from “California Sources,” but we did not find publicly available data that

specifies the exact source within the state (Figure S4). While BTEX concentrations did not significantly differ between SoCalGas and PG&E ($p = 0.12$), the vast majority of NG distributed by PG&E comes from Gas Transmission North-western pipeline, which sources NG from Western Canada,⁵⁶ indicating that while NG distributed by PG&E and SoCalGas is likely sourced from different basins, their C6+ content (NMVOCs with six or more carbons) was not significantly different.

Regional Benzene and BTEX Concentrations. Regional differences in BTEX concentrations (Kruskal Wallace: $\chi^2 = 46.7$, $p < 0.0001$, $n = 7$) were more pronounced than our observed differences by gas company. We observed exceptionally high NMVOCs collected in the North San Fernando and Santa Clarita Valleys (near the Aliso Canyon underground NG storage facility) and therefore analyzed these samples independently from the remainder of the samples in the Greater Los Angeles region. Notably, the 14 samples collected in this region—including 8 unique sites and 5 repeated samples in different months—exhibited some of the highest benzene and BTEX content observed throughout the entire study, with an overall mean benzene of 12 ppmv and a mean BTEX of 25 ppmv. We observed the maximum benzene concentration in the study (66 ppmv) in this region. The remaining samples collected in the Greater Los Angeles region had the most variability of the remaining regions, with a BTEX 95% CI of 7.5–24 ppmv, compared to the next highest BTEX 95% CI of 4.7–5.4 ppmv for the San Francisco Bay Area. Sacramento and Fresno had mean BTEX concentrations of 5.7 and 4.7 ppmv, respectively (Table S3). San Diego had relatively low BTEX concentrations compared to the other regions. Both Fresno and Bakersfield are located in the Central Valley but are serviced by different gas companies (PG&E and SoCalGas, respectively). This could partially account for their significantly lower BTEX content; however, these two areas represented the lowest sample counts with 12 and nine, respectively, and therefore required additional sampling to verify observed differences.

Overall, NMVOC and HAP concentrations reported in this study agree with the limited available data on NMVOC and HAP content observed in the midstream NG sector, including NG flowing through transmission pipelines, suggesting that the NG undergoes little additional processing.²⁵ For example, a study of HAPs present in transmission NG throughout the United States noted a mean benzene value of 5.9 ppmv (σ : ± 6.0 ppmv), suggesting the composition of NG in transmission pipelines may not differ substantially from that of distribution gas at the point of end use.^{25,57} Additionally, three preexisting NG composition samples associated with the Aliso Canyon underground NG storage facility (near Porter Ranch, CA) reported mol % benzene ranging from 0.0003–0.001 (3–10 ppmv),⁵⁸ which falls within the 25th–75th quartiles observed in samples in the North San Fernando and Santa Clarita Valleys from this study (1.8–20 ppmv).

While determining potential explanatory factors of trace gas variability was beyond the scope of this study, trace gas variability observed indicates that California’s NG supply chain is complex and likely reflects the multiple hydrocarbon sources both from in-state production and imports in addition to the variability of the efficacy of NG processing systems that support end-use consumption. This is further evidenced by the five samples in the North San Fernando/Santa Clarita Valleys, where we sampled during different seasons. On average, the

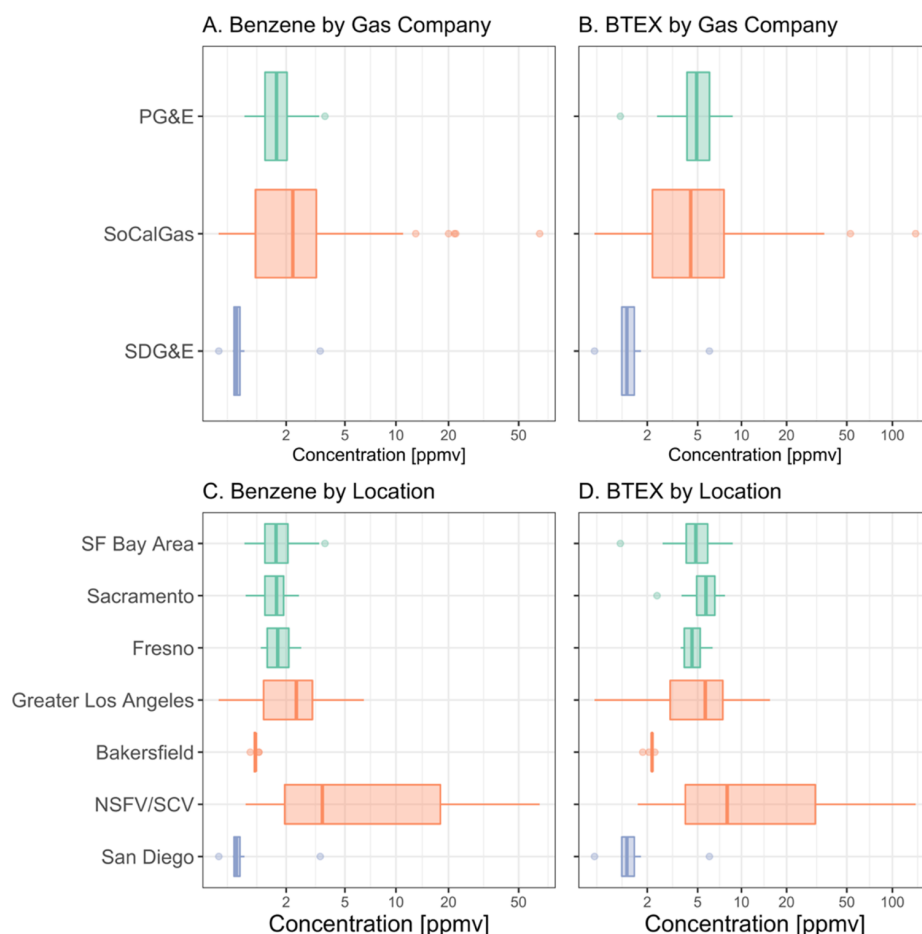


Figure 3. Concentrations of benzene and BTEX (sum of benzene, ethylbenzene, toluene, *m,p*-xylene, *o*-xylene) detected in residential NG by gas company and by location. Note that samples by location (C and D) are color-coded based on their respective gas company (A and B). Some samples collected in the North San Fernando and Santa Clarita Valleys (NSFV/SCV) had exceptionally high concentrations of BTEX in the gas; therefore, we analyzed these samples as a separate region from the remainder of the samples taken in the Greater Los Angeles region. No samples from the NSFV/SCV were included in the Greater Los Angeles region. Note: x-axis are a “pseudo-log” scale, which smoothly transitions to a linear scale around zero.

concentration of benzene in samples taken in the fall was 56% lower than samples taken in the winter/spring at the same location. Michanowicz also observed increased NMVOC concentrations during the winter heating season, suggesting that seasonal variability may be predictable and caused by source switching and/or influences from ambient temperature.²⁴

Regional and Statewide Benzene and BTEX Emission Estimates. Assuming that the NMVOC content in unburned NG observed in our study is representative of California’s transmission and distribution segments of the supply chain, we estimated annual statewide benzene and BTEX emissions using NG methane emission estimates of indoor appliances by Fischer et al.²⁶ and Lebel et al.²⁸ Our study suggests that there is an estimated 310 (95% CI: 140–980) kg benzene yr^{−1} emitted statewide from whole-house residential leaks, with an estimated 63 (95% CI: 32–150) kg benzene yr^{−1} emitted from gas stove steady-state-off leakage alone. For comparison, the 2017 NEI estimates benzene emissions from residential NG combustion (not unburned leakage) in California to be 381 kg benzene yr^{−1}.⁵⁹ Adding together benzene from combustion and our estimate of benzene from unburned leaks, our results imply that the California residential sector emits an estimated

690 kg benzene yr^{−1} from NG combustion and leakage. This suggests that the current emissions inventory underestimates benzene emissions for the residential sector by approximately 44%. This suggests that our estimate increases the existing inventory for benzene emissions from the residential sector by approximately 81%.

Using transmission and distribution methane estimates from Jeong et al.,⁴¹ the results of our study indicate that an estimated 4,200 (95% CI: 1,800–9,700) kg benzene yr^{−1} is emitted from leaks in NG distribution and transmission systems across California (Table 1).⁴¹ For context, this is a similar amount of benzene as emitted by 58,800 cars annually.^{59,60} It is also equal to nearly half of the estimated benzene emissions (~47%) from all on-road diesel light-duty vehicles in California and would rank 24th out of 49 categories of California’s benzene emissions tracked by the U.S. National Emissions Inventory (NEI).⁵⁹

Given that statewide variability in BTEX concentrations is largely driven by the Greater Los Angeles Area, we also estimated regional benzene and BTEX emissions using regional NG emission estimates by Jeong et al.,⁴² He et al.,⁴³ and SoCalGas⁴⁴ where available (Table 1). We estimated that 8.8 (95% CI: 4.5–19) kg benzene yr^{−1} is emitted from

Table 1. California Statewide and Regional Benzene and BTEX Emission Estimates Based on NG Methane Emissions from Previous Studies^{a,b,c}

study	region	sector	methane emissions (Gg/yr) (95% CI)	implied benzene emissions (kg/yr) (95% CI)	implied BTEX emissions (kg/yr) (95% CI)
Jeong et al. ⁵⁵	Statewide	NG transmission and distribution emissions	317 (179–455)	4,200 (1,800–9,700)	11,000 (5,200–26,000), 500 (5,200–25,900)
Fischer et al. ²⁵	Statewide	residential appliances steady-state operation/combustion	13.3 (6.6–37.1)	170 (67–790)	480 (192–2,100)
	Statewide	residential whole-house quiescent leaks (i.e., pilot lights, pipes, etc.)	23.4 (13.7–45.6)	310 (140–980)	850 (400–2,600)
	Statewide	residential whole-house total emissions	35.7 (21.7–64.0)	470 (220–1,400)	1,300 (630–3,600)
Lebel et al. ²⁷	Statewide	residential NG stove steady-state-off leaks	4.8 (3.1–7.0) ^d	63 (32–150)	170 (90–400)
Jeong et al. ⁵⁶	San Francisco Bay Area	whole-region emissions (primarily attributable to distribution)	(23–38) ^e	(180–370)	(590–1,200)
He et al. 2019 ⁵⁷	Greater Los Angeles	whole-region emissions based on residential and commercial NG consumption	72.8 (67.6–78.0) ^f	850 (590–1,100)	2,200 (1,600–3,000)
SoCalGas ⁵⁸	North San Fernando/Santa Clarita Valleys	utility distribution emissions	0.15 ^g	8.8 (4.5–19)	20 (9.5–50)

^aBTEX emissions are calculated by summing the emissions of the individual compounds—benzene, toluene, ethylbenzene, and all xylenes. ^b95% confidence intervals are calculated by scaling the upper and lower values of the methane estimate from the reported study and the confidence interval of the concentration of the compound from this study. ^cRegional estimates were calculated using regional concentrations of compounds from this study. ^dBased on mean steady-state-off stove emissions of 57.9 (36.3–84; 95% CI) mg CH₄ hr⁻¹. ^eBased on the assumption that NG distribution emissions are proportional to consumption. Represents a leakage rate of 0.3–0.5%. ^fBased on residential and commercial NG consumption in the South Coast Air Basin in 2020 and a leakage rate of 1.4 ± 0.1%. ^gDistribution pipeline emissions in 2020 for zip codes within an ~5 mile radius of Porter Ranch.

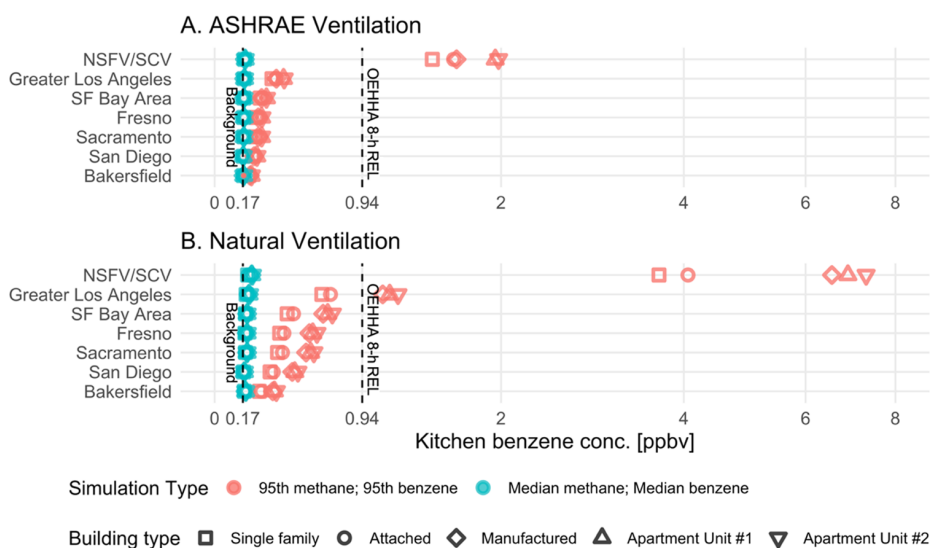


Figure 4. Kitchen indoor air benzene concentration estimates from steady-state-off CONTAM model simulations. Results are for two scenarios of ventilation: ASHRAE standard mechanical ventilation of 0.35 air changes per hour (ACH) and natural ventilation (0.05 to 0.11 ACH). The vertical dashed line shows the California Environmental Protection Agency Office of Environmental Health Hazard Assessment (OEHH) 8-h and chronic Reference Exposure Level (REL) for benzene (3 $\mu\text{g m}^{-3}$; 0.94 ppb). Median and 95th percentile methane emissions from gas stoves while off (24 and 282 mg hr⁻¹, respectively) are taken from Lebel et al.²⁸ Median and 95th percentile benzene concentrations are taken from this study (Figure 3C,D and Table S3). Building types are shown by various shapes and also have different-sized kitchens ranging from 29–33 m². Apartments 1 and 2 are the same except that Apartment 1 is on the ground floor and Apartment 2 is on the upper floor. Note: Greater Los Angeles does not include samples from the North San Fernando/Santa Clarita Valleys (NSFV/SCV).

SoCalGas distribution pipelines in the North San Fernando and Santa Clarita Valleys alone, where BTEX concentrations were the highest observed (Table S4). These estimates assume that the composition of NG for all sources is relatively stable and do not account for NMVOCs that may be produced or destroyed during the combustion process.^{61–63}

While previous studies have estimated HAP emissions from NG production and development and their implications on air quality,^{64–67} to our knowledge, HAP emissions from leaking

downstream infrastructure and NG appliances are rarely measured and are not included in current inventories. Our regional estimates of benzene emissions in the San Francisco Bay Area (Table 1) are similar to what Michanowicz et al.²⁴ estimated in the Greater Boston Area (120–356 kg benzene yr⁻¹) but are 3–5 times higher in the Greater Los Angeles region.²⁴ Moreover, these inferred NMVOC emissions have likely been misclassified or unaccounted for as sources of

secondary formation of ozone and $\text{PM}_{2.5}$, particularly in nonattainment areas.

Modeled Benzene Kitchen Concentrations. Most model simulations—including all median value simulations—did not result in ambient benzene concentrations attributable to emissions of NG from gas stoves that are off above the California Environmental Protection Agency Office of Environmental Health Hazard Assessment (OEHHA) 8-h REL of 0.94 ppbv (also equal to the chronic REL). However, we found that certain parameter combinations have the potential to lead to exceedances of the OEHHA benzene REL from gas stoves while off (Figure 4).³⁷ Modeled indoor ambient benzene concentrations ranged from 38–126% of the 8-h REL in the Greater Los Angeles region and reached over 700% the REL in the North San Fernando/Santa Clarita Valleys. Notably, all 95th percentile model runs for the NSFV/SCV region resulted in REL exceedances irrespective of ventilation or building type or other indoor sources of benzene. These exceedances produce indoor concentrations of benzene similar to concentrations to environmental tobacco smoke (i.e., secondhand smoke).^{68,69}

Based on model results, an elevated leakage rate of benzene and a low ventilation rate are both requisite for indoor concentrations to exceed the OEHHA 8-h REL for benzene. Using multiple linear regression, we found that the benzene emission rate (benzene concentration times methane flow rate) was the most important factor for determining the kitchen benzene concentration ($p < 0.0001$), followed by the air changes per hour ($p < 0.0001$)—a measure of the air tightness of the space. Interestingly, building size had an insignificant effect on benzene concentration ($p = 0.19$), possibly due to a small number of modeled home sizes. Modeled air changes per hour (ACH) for the natural ventilation simulations ranged from 0.11 for the single-family home to 0.05 for apartments, which has been verified empirically elsewhere (See the [Supporting Information](#) for more details).⁷⁰

There are several reasons our estimates of indoor concentrations of benzene could underestimate true concentrations indoors. First, our calculations were exclusively focused on the contribution of benzene from NG leakage from gas stoves while off and do not include leakage from gas stoves during combustion or incomplete combustion,⁶² other gas appliances (including but not limited to quiescent leakage from other appliances, see Table 1), gas pipes in residence, or other non-NG benzene sources. Second, we included a background ambient benzene concentration of 0.174 ppb for all model runs—the average outdoor ambient concentration in California in 2019.⁷¹ This background benzene concentration may be a conservative measure particularly for some outdoor regions within our study area, and indoor VOCs are generally higher than outdoors to begin with. For example, the South Coast Air Quality Management District (SCAQMD) notes a background outdoor benzene concentration of 0.25 ppb.⁷² We designed our modeling to identify a possible hazard (gas stoves while off only) and recognize that our parameters could lead to underestimating indoor benzene concentrations.

Future Research Directions. Our sampling effort was designed to capture geographic variability rather than temporal variability. While several of our samples were collected in different seasons, our study did not systematically investigate seasonal variability and its associated impacts (e.g., NG demand, withdrawal from underground NG storage facilities,

etc.) on concentrations of NMVOCs in distribution gas. Future work could investigate temporal trends in California in greater depth, as temporal variability was evident in Boston.²⁴ Additionally, while we can infer the various high-volume transmission lines supplying NG regionally in California, we were unable to ultimately source NMVOC composition by upstream source or any potential modifying factors. The highly elevated NMVOC concentrations in distribution NG in the North San Fernando/Santa Clarita Valleys warrant further study to better understand factors contributing to these high concentrations.

Our CONTAM modeling only included emissions of unburned NG leaking from one source: stoves while off. Future work should consider air quality impacts and human health risks of exposure to multiple HAPs and other air pollutants (beyond benzene), emissions of unburned NG from appliances beyond stoves, and indoor emissions from burning NG to build a more complete picture of the potential indoor air quality and health impacts of household NG usage. Additionally, fugitive VOCs from downstream NG systems are likely an under-appreciated contributor to the secondary formation of $\text{PM}_{2.5}$ and ozone—contributing to outdoor air pollution—and should be studied further. As transportation and industry-related VOC emissions continue to decline in urban areas, NG-sourced VOCs may increase in their relative importance as $\text{PM}_{2.5}$ and ozone precursors.

Policy Implications. While there has been substantial research on the health cobenefits of greenhouse gas emission-reduction strategies in transportation, electricity generation, buildings, and industry that has helped inform policy, there is limited research on the health benefits of policies that reduce NG emissions and coemitted air pollutants. This is particularly true in urban areas with many emission sources and in indoor environments where people may be much more frequently exposed to NG-related emissions at higher concentrations. Thus, our study adds to the body of literature that suggests that downstream NG emissions are an unaccounted-for source of both indoor and outdoor emissions of NMVOCs and HAPs. California is already starting to take action to reduce urban pollution from NG: in September 2022, California air regulators approved a commitment to phasing out the sale of NG-fired furnaces and water heaters by 2030. The state has also moved to phase out subsidies for connecting new homes to the NG system, and is considering ending rebates for gas appliances to incentivize the transition to electric alternatives.^{73,74}

In August 2021, the California Energy Commission adopted its 2022 Building Energy Efficiency Standards, which, in part, included differentiated demand-controlled ventilation requirements for electric stoves vs NG-fired stoves.⁷⁵ While this could lead to important improvements in indoor air quality related to combustion-related pollutants (e.g., NO_x , $\text{PM}_{2.5}$, CO_2), this ventilation requirement likely does not impact indoor air quality associated with fugitive, steady-state-off cooking appliance NG emissions—the source-exposure pathway characterized and modeled in this study.

While operating a continuous HVAC system would address nontransitory exposures, it is both cost- and energy-intensive, and retrofitting existing built environments like apartment complexes may not be feasible. Moreover, failing to address indoor pollution sources (e.g., cooking) has been shown to lead to unaccounted-for indoor air quality disbenefits following energy efficiency weatherization such as building sealing.⁷⁶ Our

study suggests that weatherization and building efficiency measures should be coupled with residential and other building electrification measures to support multiple health, air quality, and climate cobenefits by mitigating HAPs and methane emissions while appliances are both off and on.^{77–81} In California, the California Public Utilities Commission (CPUC) pilot programs, TECH and BUILD, aim to reduce the use of NG in the home and provide incentives for all-electric new housing—with the latter program focusing on low-income households.^{82,83} Given the potential air quality impacts and associated health risks from fugitive NG emissions alone—particularly those that may occur indoors—these electrification programs likely entail additional and unaccounted-for health cobenefits and associated long-term cost savings.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.2c02581>.

Details on sample analysis, locations, and pollutant data; details on CONTAM modeling, and summary of public information on California NG sources and storage facilities (PDF)

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Notes

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Population Attributable Fraction of Gas Stoves and Childhood Asthma in the United States

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Abstract: Indoor gas stove use for cooking is associated with an increased risk of current asthma among children and is prevalent in 35% of households in the United States (US). The population-level implications of gas cooking are largely unrecognized. We quantified the population attributable fraction (PAF) for gas stove use and current childhood asthma in the US. Effect sizes previously reported by meta-analyses for current asthma (Odds Ratio = 1.34, 95% Confidence Interval (CI) = 1.12–1.57) were utilized in the PAF estimations. The proportion of children (<18 years old) exposed to gas stoves was obtained from the American Housing Survey for the US, and states with available data (n = 9). We found that 12.7% (95% CI = 6.3–19.3%) of current childhood asthma in the US is attributable to gas stove use. The proportion of childhood asthma that could be theoretically prevented if gas stove use was not present (e.g., state-specific PAFs) varied by state (Illinois = 21.1%; California = 20.1%; New York = 18.8%; Massachusetts = 15.4%; Pennsylvania = 13.5%). Our results quantify the US public health burden attributed to gas stove use and childhood asthma. Further research is needed to quantify the burden experienced at the county levels, as well as the impacts of implementing mitigation strategies through intervention studies.



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Keywords: gas; stove; current; use; asthma; burden; cooking; respiratory; children

1. Introduction

Indoor gas stove use for cooking is associated with an increased risk of current asthma among children [1] and is prevalent in 35% of households in the United States (US), with certain states (e.g., California, Illinois) reaching 68% [2]. Although children's exposure to gas cooking is widespread, the population-level implications of cooking with gas are largely unrecognized [3]. With this background, we attempted to quantify the population-attributable fraction (PAF) for gas stove use and current childhood asthma in the US, including at the state level, which had never been done. The PAF can be helpful for determining the proportion of preventable disease and may guide public health interventions that aim to reduce disease risk

2. Materials and Methods

First, we sought to update the effect-size estimates by identifying peer-reviewed manuscripts published subsequent to the most recent meta-analysis [1]. We searched PubMed using keywords: “gas cooking and children” OR “gas appliance and children” OR “unvented and children” OR “gas heating and children” OR “gas heater and children”. After including only manuscripts of human studies published in English since 4 January 2013, 357 studies remained for possible inclusion. The title review identified 27 manuscripts

as potentially pertinent. Full manuscripts ($n = 27$) were independently reviewed by co-authors; none reported new associations between gas stove use and childhood asthma specifically in North America or Europe. As a result, effect sizes previously reported for current asthma in North America and Europe combined (weighted by inverse variance; $N_{\text{studies}} = 10$; Odds Ratio (OR) = 1.34, 95% Confidence Interval (CI) = 1.12–1.57) were utilized in the PAF estimations [1]. The combined effect size was based on a North America-specific effect size ($N_{\text{studies}} = 3$; OR = 1.36, 95% CI = 0.76–2.43) and Europe-specific effect size ($N_{\text{studies}} = 7$; OR = 1.34, 95% CI = 1.13–1.60), as reported in a previously published meta-analysis [1]. We combined effect sizes for North America and Europe given the similarities in housing characteristics and gas-stove usage patterns across these geographies. We estimated the proportion of children (<18 years old) exposed to gas stoves in the US and in certain states using the American Housing Survey (AHS) [2]. The AHS is a nationally representative survey of households used to characterize the residential housing stock of the US. Each year, a subset of states is oversampled, allowing for precise estimates of housing characteristics for these states. In the most recent iteration of the survey in 2019, nine states were oversampled, allowing us to estimate PAFs at the national level and for these nine states.

PAFs were estimated using a previously published approach [4]. The approach aimed to quantify our uncertainty regarding the PAF point estimates, given that the inputs to the calculation are based on samples of the population. First, we used the summary statistics of the North America and Europe combined odds ratio described above to generate a distribution of effect sizes. Similarly, we used the summary statistics of the proportion of children exposed to gas cooking to generate distributions of proportions nationally and for each state for which we have data. We then drew a random value from each of these distributions to calculate the PAF according to the following equation:

$$\text{PAF} = \frac{p \times (RR - 1)}{p \times (RR - 1) + 1}$$

where p is the proportion of households with children exposed to gas stoves and RR is the relative risk of developing asthma given exposure to gas stoves. Note that we used the combined North America and Europe odds ratio for the relative risk in this equation, as childhood asthma in the US remains relatively rare, affecting 1 in 12 children [5]). We repeated this calculation 10,000 times to create distributions of PAFs nationally and for each state, using the same combined odds ratio for national- and state-level calculations. We then took the mean of these distributions and calculated their 95% confidence intervals to estimate the PAF of current childhood asthma attributable to gas stove usage. All calculations were performed using the R programming language.

3. Results

We found that 12.7% (95% CI = 6.3–19.3%) of current childhood asthma in the US is attributable to gas stove use (Figure 1). At the state level, the proportion of childhood asthma that could be theoretically prevented if gas stove use was not present (e.g., state-specific PAFs) varied. Illinois experiences the highest burden (21.1%), followed by California (20.1%), New York (18.8%), Massachusetts (15.4%), and Pennsylvania (13.5%). Texas, Colorado, and Ohio all experience burdens around 10%. Florida experiences the lowest burden (3%). The state-level PAFs differ due to varying exposure to gas stoves among children. In Illinois, for example, approximately 79.1% of households with children cook with gas, whereas in Florida, the figure is only 9.1%. States with a higher percentage of children living in households with gas stoves have higher proportions of current childhood asthma attributable to gas stove usage.

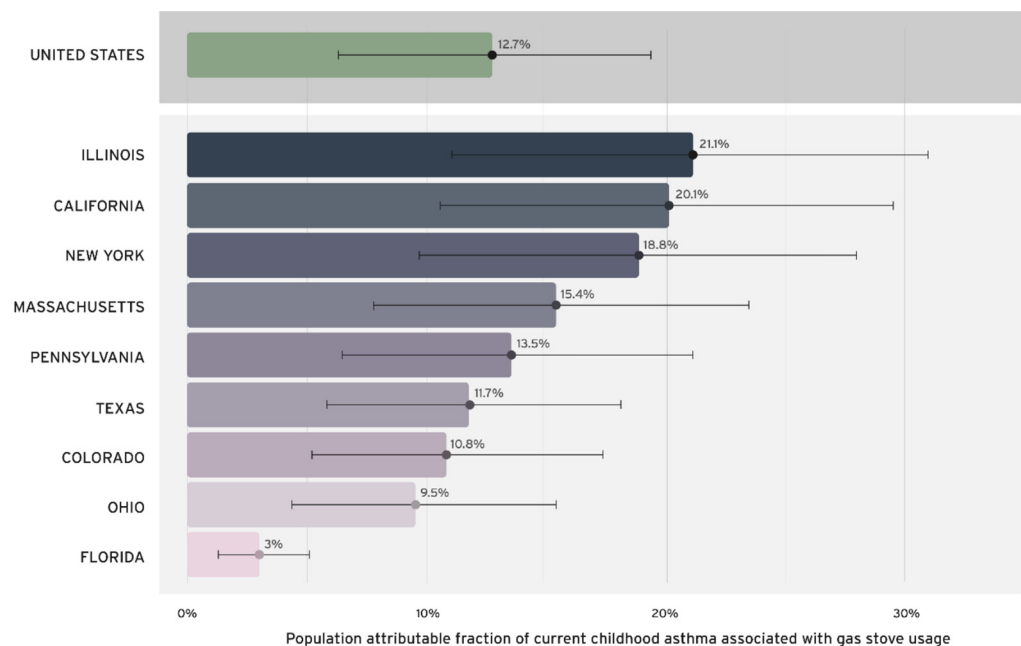


Figure 1. Population attributable fraction (PAF) of current childhood asthma associated with gas-stove use in the United States [†]. [†] PAFs calculated using methods published by Knibbs et al., 2018 [4], with the effect sizes reported by Lin et al., 2013 [1] and the proportion of children (<18 years old) exposed to gas stoves reported by the 2019 American Housing Survey (AHS) [2]. AHS only included information on gas stove use for the nine states included. No data were available for other states.

4. Discussion

Our results quantify the US public health burden attributed to gas-stove use and childhood asthma. Our study strengths include utilization of peer-reviewed effect sizes [1] and existing PAF models [4]. While we are the first to report these PAFs at national and state-specific levels using nationally representative AHS data, we were limited by the number of states ($n = 9$) with available data. Nonetheless, our results align with a cross-sectional study which found that the use of a gas stove or oven for heat was a main risk factor for doctor-diagnosed asthma in US children under age six [6].

We note that PAF analyses have limitations. For example, we rely on aggregate data, although we have quantified our uncertainty and have still found a significant public health burden. Additionally, our interpretation of the PAF as the proportion of preventable disease rests on the assumptions that (1) exposure to gas cooking among children is orthogonal to other risk factors such as exposure to tobacco smoke and (2) that we can conceive of a broad-based public health intervention to reduce the disease risk in children exposed to gas cooking to that of the unexposed, while holding all other risk factors equal.

We posit that there are two such interventions to reduce the childhood asthma disease risk attributable to gas stoves: (1) removing the source by replacing gas cooking with cleaner alternatives (e.g., electric), and (2) reducing exposure through source ventilation (e.g., range hoods). Notably, ventilation is associated with the reduction, but not elimination, of childhood asthma risk [7]. Moreover, high-efficiency range hoods are not practical in all settings (e.g., apartment buildings). In homes with range hoods, they may not vent outdoors or be effective at removing combustion pollutants, and residents simply may not use them [8]. Indeed, according to National Health and Nutrition Examination Survey data, among children living in households that use gas stoves, only 21.1% live in households where the stove's exhaust vent is always used.

5. Conclusions

In conclusion, 12.7% of current childhood asthma nationwide is attributed to gas stove use, which is similar to the childhood asthma burden attributed to secondhand smoke

exposure [9]. Gas stove usage should be considered in multi-faceted asthma prevention approaches. Given that this exposure is preventable, our study demonstrates that known mitigation strategies will lessen childhood asthma burden from gas stoves, particularly in states with elevated PAFs. Further research is needed to quantify the burden experienced at the state and county levels, as well as the impacts of implementing mitigation strategies through intervention studies.

Author Contributions: Conceptualization, B.A.S. and H.D.H.III; methodology, L.D.K. and H.D.H.III; formal analysis, T.G.; data curation, T.G.; writing—original draft preparation, T.G.; writing—review and editing, T.G., B.A.S., L.D.K. and H.D.H.III. All authors have read and agreed to the published version of the manuscript.

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STATE OF MICHIGAN

BEFORE THE MICHIGAN PUBLIC SERVICE COMMISSION

In the matter of the application of **DTE
ELECTRIC COMPANY** for authority to
increase its rates, amend its rate schedules
and rules governing the distribution and
supply of electric energy, and for
miscellaneous accounting authority

Case No. U-21534

ALJ Sally Wallace

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I, Mark N. Templeton, certify that an electronic copy of the Accompanying Exhibits DAO-211 to DAO-215 (Part 2 of 3) for the Direct Testimony of Yunus Kinkhabwala on Behalf of Soulardarity and We Want Green, Too was served on the following on July 26, 2024.

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The statements above are true to the best of my knowledge, information, and belief.

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Date: July 26, 2024

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