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Gas and Propane Combustion from Stoves Emits Benzene and Increases Indoor Air Pollution

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ABSTRACT: Exposure pathways to the carcinogen benzene are well-established from tobacco smoke, oil and gas development, refining, gasoline pumping, and gasoline and diesel combustion. Combustion has also been linked to the formation of nitrogen dioxide, carbon monoxide, and formaldehyde indoors from gas stoves. To our knowledge, however, no research has quantified the formation of benzene indoors from gas combustion by stoves. Across 87 homes in California and Colorado, natural gas and propane combustion emitted detectable and repeatable levels of benzene that in some homes raised indoor benzene concentrations above well-established health benchmarks. Mean benzene emissions from gas and propane burners on high and ovens set to 350 °F ranged from 2.8 to 6.5 μg min^{-1}, 10 to 25 times higher than emissions from electric coil and radiant alternatives; neither induction stoves nor the food being cooked emitted detectable benzene. Benzene produced by gas and propane stoves also migrated throughout homes, in some cases elevating bedroom benzene concentrations above chronic health benchmarks for hours after the stove was turned off. Combustion of gas and propane from stoves may be a substantial benzene exposure pathway and can reduce indoor air quality.

KEYWORDS: *gas stoves, benzene emissions, homes and residences, health, electric and induction stoves*

INTRODUCTION

Natural gas appliances, including furnaces, water heaters, and stoves, emit carbon dioxide through combustion and also emit methane directly into air through leaks and incomplete combustion.[15−17] Methane (CH_4), the principal component of natural gas, is the second largest greenhouse gas contributor to global climate change after carbon dioxide.[14] Gas appliances in homes further release health-damaging air pollutants, though only stoves typically release these pollutants directly into home air rather than venting them outdoors as furnaces and water heaters do.

Through combustion, gas stoves also emit toxic chemicals, including carbon monoxide (CO), which inhibits cellular respiration; formaldehyde (HCHO), a carcinogen; and nitrogen dioxide (NO_2), a respiratory irritant linked to asthma.[2−6] Under common-use conditions, indoor NO_2 from gas stoves can quickly exceed US Environmental Protection Agency (EPA) and World Health Organization (WHO) 1-h exposure benchmarks in kitchen air.[2,4,18,19] NO_x (defined here as NO + NO_2) pollution has been shown to harm human health; for instance, a meta-analysis of 41 studies on gas stoves and childhood respiratory health concluded that children who live in homes with gas stoves had a 24% higher risk of lifetime asthma and a 42% increased risk of having asthma currently.[20] Furthermore, a recent population-level analysis concluded that approximately 12% of childhood asthma in the U.S. is attributable to gas stoves.[21] More than 1/3 of U.S. households (47 million households) cook with gas,[11] and millions more do so worldwide.[12,13]

Although the formation of NO_x, carbon monoxide, and formaldehyde from gas combustion in stoves is relatively well-characterized, other hazardous air pollutants known to form in gas flames, such as benzene, have yet to be quantified indoors associated with gas combustion in stoves. Like carbon monoxide, benzene (C_6H_6) is a product of incomplete combustion. Benzene is defined as a Group 1 known human carcinogen by the International Agency for Research on Cancer[22] and as a carcinogen by the US EPA.[23] Benzene formation in methane flames is well-characterized in the laboratory,[24−27] and benzene has been observed to form from natural gas combustion in other settings such as during flaring[28] and in utility-scale boilers.[29] Other cooking fuels such as wood, charcoal, kerosene, and liquefied petroleum gas have also been shown to degrade indoor air quality in some cases by emitting pollutants such as benzene.[30] We hypothesized that

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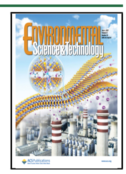

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gas and propane stoves could similarly produce benzene indoors, creating a potential human health risk.

Benzene exposure causes both cancer and noncancerous health effects. Shorter-term benzene exposure suppresses blood cell production, and chronic benzene exposure increases the risk of leukemias and lymphomas.[31,32] The carcinogenic effects of benzene exposure follow a linear dose-response curve with no lower threshold, meaning that any additional benzene exposure increases leukemia and lymphoma risk.[32] In contrast, some adverse noncarcinogenic effects, such as the suppression of blood cell production, likely occur only when benzene exposure exceeds a threshold.[32]

The carcinogenic and noncarcinogenic effects of benzene are reflected in current benzene exposure benchmarks. These benchmarks generally fall between 2.0 $\mu g/m^3$ (~0.63 ppbv), established in France, and 5.0 $\mu g/m^3$ (~1.6 ppbv), the most common global benchmark adopted by the European Union, India, South Korea, and other jurisdictions.[9] The World Health Organization, however, concluded that for cancer effects, "there is no known exposure threshold for the risks of benzene exposure" and that "it is expedient to reduce indoor exposure levels to as low as possible".[22] Some jurisdictions have also set indoor exposure benchmarks associated with different time horizons based solely on benzene's noncarcinogenic risk. For example, the California Office of Environmental Health Hazard Assessment (OEHHA) under the California Environmental Protection Agency (CalEPA) established an 8-h reference exposure level of 3 $\mu g/m^3$ (~1 ppbv) in 2014, equal to the state's chronic exposure level.[8]

Previous research on indoor benzene exposure concluded that various factors in homes can elevate benzene concentrations, particularly tobacco smoke, fossil-fueled appliances such as kerosene stoves, attached garages, and proximity to roads with gasoline-vehicle traffic.[1,10,33−37] Two studies found that benzene concentrations were elevated above the baseline when a gas or propane cooking unit was on but did not quantify benzene emission rates.[36,37]

To our knowledge, ours is the first study to quantify combustion-based benzene emission factors from gas and propane stoves. We also measured benzene emissions from alternative stove types (electric coil, radiant, and induction). To isolate benzene emissions from fuel use from any emissions from cooking food, we quantified benzene emissions from all cooktop elements by boiling water using the same pot in all homes and by measuring emissions in empty ovens. Beyond the fuel, another potential source of benzene associated with cooking is the food, particularly emissions from heating fats and oils. Maximum recommended frying temperatures are approx. 190 °C,[38] and previous research suggested that common frying oils produce not more than 2.5 ng benzene per g oil per minute at 200 °C.[39] We tested whether food itself is a source of benzene by cooking two meal types using nonbenzene-emitting induction cooktops.

MATERIALS AND METHODS

Definitions. We define a "cooktop" as a flat surface with 2−6 individual cooking elements, "burners" as cooking elements using a gas or propane flame, "coils" as those with an exposed metal coil that is heated via electric resistance, "radiant hobs" as elements with a metal coil covered by a smooth ceramic or glass surface, and "induction hobs" as elements that heat cookware directly using electromagnetic induction. We therefore define a "cooktop element" broadly as a single burner, coil, or hob.

For ovens, we define gas, propane, and electric ovens as enclosed spaces heated by gas, propane, and electric resistance, respectively (there is currently no oven equivalent of an induction hob). We define a "stove" (also called a "range") as a freestanding unit that contains both a cooktop and an oven. Stoves typically use the same fuel for the cooktop and oven, although some have a gas cooktop and an electric oven. We grouped electric coils and radiant hobs together in our sampling because they both transfer heat to a pot via conduction. Throughout the paper, we use the term "concentration" for its accessibility in place of the more strictly correct term "molar mixing ratios." We assume a temperature of 25 °C and a pressure of 1 atm when converting between true concentrations and molar mixing ratios, which yields the conversion 1 ppbv benzene = 3.19 μg benzene m^{-3}.

To promote real-world comparisons (using vocabulary that home cooks use), we refer to burners set to "high" and "low" in homes. Although these categories are qualitative, the least powerful gas burner that we tested on high operated at >1 kW of power (3.4 kBTU h^{-1}), whereas the most powerful gas burner that we tested on low operated at <1 kW (Figure S11). For gas stoves, we therefore define burners on "high" as >1 kW and "low" as <1 kW. In contrast, propane stoves had some overlap in power output across stoves between "high" and "low" settings. Grouping benzene emission results from propane stoves by power output (>1 kW and <1 kW) rather than by "high" and "low" on the burner dials yields similar results to our "high" and "low" groupings (Figure S12). Because electric coils and induction stoves do not emit CO_2, we used the qualitative values of "high" and "low" from the stoves to describe their outputs.

As described below, when quantifying benzene emission factors (rates of benzene emitted per unit time), we closed kitchen doors and windows and sometimes sealed parts of the kitchen with plastic to limit the kitchen volume in order to measure the emission rates more accurately (see Figure S8). Throughout the paper, we refer to kitchens partitioned with any plastic as "sealed" kitchens and to kitchens with doors and windows closed but with no plastic sealing as "unsealed" kitchens. When we conducted our long 8-h time course measurements of concentrations in kitchens and bedrooms, we opened interior doors. We refer to these as "open kitchens".

Measurement Instrumentation. We measured benzene using an Entanglement Technologies, Inc. AROMA VOC analyzer (model ARVOC-191E, RSO), a cavity ring-down analyzer that quantifies benzene concentrations based on the molecule's infrared spectroscopic absorbance signature.[40] The AROMA analyzer collects one data point every 855 s (14.25 min) and collects sample air for 5 min of each measurement cycle. Thus, each AROMA data point represents a concentration average over 5 min taken once every 15 or so minutes.

To normalize benzene emissions by the amount of gas burned and to calculate the room volume through an ethane injection of known volume, we measured carbon dioxide and ethane using a Picarro cavity ring-down spectrometer (model G-2210i). To confirm results from the Picarro, during portions of our sampling campaign, we also measured ethane using a duplicate Aeris methane/ethane MIRA Ultra Mobile LDS analyzer and CO_2 using an Aeris carbon dioxide/nitrous oxide MIRA Ultra Mobile LDS analyzer. We measured carbon monoxide (CO) using a U_MCEA Los Gatos Research

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analyzer. The calibration of the AROMA VOC analyzer was regularly checked (typically once at the beginning of each sampling day). The calibrations of the other analyzers were checked regularly (typically weekly) and after being transported.

Site Selection. We sampled 87 stoves in 14 counties in California and Colorado between January and December 2022 (see Figure S6 for a map of sampling locations). Our set of sample residences included a range of kitchen sizes in private homes, apartments, and several Airbnb rentals, where we could measure longer—including overnight—time courses (see Figure S7). We selected homes through an online participant sign-up page and neighborhood and community associations (see Table S2 for a summary of the characteristics of stoves that we sampled).

Emission Rate Calculations from Combustion. We calculated benzene emission rates attributable to gas combustion by measuring the increase in benzene concentration through time in sealed and unsealed kitchens (see above), an approach analogous to that used by Lebel et al. to measure methane and NO_x emissions from stoves.[2] We converted measured concentrations into emission rates using eq 1:

$$f_{\mathrm{b}} = V_{\mathrm{k}}\frac{1}{n}\,\Sigma_{i=0}^{n}\lambda\left(\frac{C_{\mathrm{b},t_i} - C_{\mathrm{b},t_{i-1}}\,\mathrm{e}^{-\lambda(t_i - t_{i-1})}}{1 - \mathrm{e}^{-\lambda(t_i - t_{i-1})}} - C_{\mathrm{b,b}}\right) \quad (1)$$

where f_b is the mean benzene emission rate over the course of a measurement (in volume per time), V_k is the kitchen volume, λ is the kitchen chamber's air exchange constant (in reciprocal time), n is the number of benzene concentration data points collected in a given measurement, $C_{b,t}$ is the benzene concentration at time t (in parts per billion volume), t_i is the timestamp of the ith data point, and $C_{b,b}$ is the background benzene concentration outside the kitchen chamber (in parts per billion volume; see Supplementary Methods for the derivation of eq 1). We assume that $C_{b,b}$ is equal to the benzene concentration that we measured inside the kitchen immediately after airing it out with fresh outdoor air. We converted volumetric emission rates into gravimetric emission rates using the temperature measured in the kitchen.

For a perfectly sealed kitchen with $\lambda = 0$, note that

$$\lim_{\lambda \to 0} V_{\mathrm{k}}\frac{1}{n}\,\Sigma_{i=0}^{n}\lambda\left(\frac{C_{\mathrm{b},t_i} - C_{\mathrm{b},t_{i-1}}\,\mathrm{e}^{-\lambda(t_i - t_{i-1})}}{1 - \mathrm{e}^{-\lambda(t_i - t_{i-1})}} - C_{\mathrm{b,b}}\right) = V_{\mathrm{k}}\frac{1}{n}\,\Sigma_{i=0}^{n}\left(\frac{C_{\mathrm{b},t_i} - C_{\mathrm{b},t_{i-1}}}{t_i - t_{i-1}}\right)$$

which is the average rate of the benzene concentration rise multiplied by the kitchen volume. We measured the energy output from gas burners and ovens using the flow rate of CO_2 and the enthalpy of combustion of methane to calculate the joules (J) of energy emitted per unit time.

We calculated the kitchen volume (V_k) and air exchange constant (λ) using 500 mL injections of ethane as a tracer, using fans to mix the kitchen air. The estimated kitchen volume is the injected ethane volume divided by the peak ethane concentration immediately following injection, and the air exchange constant is the ethane's decay constant through time after the peak (see Supplementary Methods). We also measured the volume of four kitchens using a laser distance measurer and obtained good agreement with the ethane injection method (slope = 1.1, standard error of the slope = 0.12, r^2 = 0.97, and p value of the line fit: 0.01).

Measurement Setup. To sample air in the kitchen or other rooms of interest, we connected polytetrafluoroethane (PTFE) tubing to a stand approximately 1.5 m off the ground, typically in the center of the kitchen or other rooms (and not immediately next to the stove, see Figure S8). We drew air from the kitchen to the analyzers located outside the kitchen using a 7 L min^{-1} pump.

To measure some benzene emission rates (but never benzene concentrations), we created enclosed kitchen partitions with well-mixed air by closing the kitchen's doors and windows, closing off some open spaces with plastic in some houses, and placing one or two fans in each kitchen to mix the air while being careful not to disturb the burner flame (see Figure S8). In other cases, we measured benzene emission rates in unsealed kitchens with doors closed but without plastic partitions (see above). We accounted for gaps under doors, in windowpanes, and around any plastic sealing with the measured air exchange constant λ (see above). We used plastic sealing for measuring benzene emission factors only; we never used plastic when reporting benzene concentrations in any rooms. We verified that the plastic we used did not emit detectable benzene by placing approximately 10-fold the amount of plastic used to seal a large kitchen in a closed kitchen at 95 °F and measuring the benzene concentration over 45 min, our typical measurement time. This quantity of plastic did not emit detectable amounts of benzene.

We aimed to eliminate all potential sources and sinks of benzene in the kitchen chamber other than the stove itself. We kept the range hood (a potential sink) off if one was present except when testing hood-on and hood-off comparisons as described below. When measuring emission rates from ovens, we ensured that the oven was empty. When measuring burners, we placed the same stainless-steel pot with tap water on every burner analyzed across all houses (i.e., the same pot for every burner or hob measurement). The pot contained only water and, to avoid introducing potential volatiles from detergents, was not washed between emissions factor measurements. Using a nonemitting induction cooktop, we verified that tap water emits no detectable benzene when boiled. No people were present in kitchens during measurements except for <30 s to turn on the burner or oven or for scenarios that involved cooking food. When we measured emissions from cooking food, we typically had someone in the kitchen for the duration of the measurement to stir and prevent the food from burning. People do not exhale amounts of benzene that would be detectable in kitchen air.[43]

Statistics. We calculated mean and median 95% confidence intervals from a 25,000 replicate bootstrap as described previously (Lebel et al.).[2] We generated each replicate sample in the bootstrap by randomly sampling with replacement of the set of emission factors in question to form a random sample of equal size to the set of emission factors in question. For instance, the bootstrap for gas burners on high consisted of 25,000 replicates of size 54, generated by randomly sampling (with replacement) the set of 54 emission rates for gas burners on high. We then calculated 95% CIs for the means and medians of these bootstraps.[44,45] We used an analogous methodology to measure benzene emissions from electric, induction, and propane stoves, with replicate sizes equal to our sample size in each category. For stove emissions, we

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calculated statistical significance between groups using the two-sided Mann−Whitney U test (also known as the Wilcoxon rank-sum test). For emissions from food, we calculated significance relative to zero using the two-sided Student's t-test (because measurement uncertainty could lead to negative values). Sample sizes for each category are summarized in Table 1.

Table 1. Summary of Sample Sizes for Each Cooktop Element and Oven Type

	burners on high	burners on low	ovens
gas	54	43	47
propane	11	10	9
coil/radiant/electric	14	9	18
induction	13	9	N/A

Controlled-Release Validation. Prior to our sampling campaign, we validated our benzene measurement methods using controlled-release experiments. We injected 1.5 and 10 ppmv benzene standards at known flow rates ranging from 150 mL min^{-1} to 5 L min^{-1} of the standard into two unoccupied chambers with volumes of 27,200 and 30,050 L. This process produced benzene concentrations typical of values that we observed in kitchens during combustion by gas stoves. The slope of our calculated emission rate divided by the known emission rate was statistically indistinguishable from one (slope = 0.992, standard error of the slope = 0.08, r^2 = 0.94, p value of the line fit $\ll$ 0.01; see Figure S9).

Benzene Concentration Measurements in Air. In addition to measuring emission rates as described above, we also measured benzene concentrations in kitchens and bedrooms for various stove-use scenarios. The kitchen benzene concentrations described in the section "Benzene Concentrations in Home Air" were measured in 17 "unsealed" kitchens (see Definitions above). Because we sometimes tested more than one burner or oven in each kitchen (always separately), we have a total of 33 kitchen benzene concentration measurements. Additionally, the long 8-h time course benzene concentrations reported in the section "Benzene Migration to Bedrooms" were measured in open kitchens (see Definitions above) in 6 different houses. We did not use fans or other means of active air circulation during the long 8-h time courses. In all six houses for which we did the 8-h time courses, we set the oven to 475 °F, a temperature commonly used to bake bread, for 1.5 h with the hood off. We continued monitoring benzene concentrations for 6.5 more hours in the farthest bedroom from the kitchen after turning the oven off.

Benzene Emissions from Fuel Burned to Cook a Single Meal on a Gas Range. We estimated the amount of benzene emitted by gas combustion associated with cooking a single meal based on median benzene emission factors from gas stoves reported here and from previous research tracking usage of cooktops (burners) and ovens in 70 California homes[7] (see the Supporting Information).

National Scale-Up of Emissions. To estimate the amount of benzene emitted by gas combustion in all stoves annually in the United States, we calculated the mean benzene emission factor (ng C_6H_6 J^{-1} gas burned) for ovens and burners on high using the flow rate of CO_2 and the enthalpy of combustion of methane to calculate the joule (J) of energy emitted per unit time. We calculated the 95% CI using a 25,000 replicate bootstrap (see above). We then multiplied this emission factor and 95% CI by the 2015 Residential Energy Consumption Survey estimate[11] of annual natural gas burned for residential cooking in the United States to reach our annual estimate of benzene emissions from gas combustion in stoves. This estimate is based on the assumption that most gas used for cooking is consumed by ovens or by burners on high. If we instead assumed that more gas is consumed by burners on low, our mean estimate would increase by 30% because we found that burners on low emit more benzene per joule of natural gas consumed than the amount that burners on high emit.

Emissions from Cooking Food and Leakage of Unburned Gas. We measured benzene emissions from cooking meals on two induction cooktops that we determined had zero detectable benzene emissions (see Figure S3). Any benzene emissions could then be attributed to cooking food rather than to the cooktop or fuel used. We cooked two meals, pan-fried fish and bacon, selected because previous research showed particulate and polyaromatic hydrocarbon emissions from frying these foods to be relatively high.[46] We cooked fish with approximately one tablespoon of olive oil as well as bacon in its own grease, both on medium heat in a stainless-steel pan with regular flipping. We cooked three replicates of each meal for 20−35 min until they were "well done" (see Figure S3) to understand the maximum potential emissions of benzene. We calculated p values relative to zero using the two-sided Student's t-test.

We calculated benzene emission rates attributable to leakage of uncombusted gas using published data on gas stove leakage rates,[2] data on benzene concentrations in natural gas in California,[41] and the following equation from Lebel et al.[2] (adapted from Marrero et al.[42]):

$$E_{\mathrm{benzene}} = E_{\mathrm{methane}} \times C_{\mathrm{benzene}}\left(\frac{\mathrm{MW}_{\mathrm{benzene}}}{\mathrm{MW}_{\mathrm{methane}}}\right)$$

where E_{benzene} is the emission rate of benzene, E_{methane} is the emission rate of methane, C_{benzene} is the mole fraction of benzene in the leaked gas, $\mathrm{MW}_{\mathrm{benzene}}$ is the molecular weight of benzene, and $\mathrm{MW}_{\mathrm{methane}}$ is the molecular weight of methane.

RESULTS

Benzene Emissions from Combustion. Benzene emissions from gas and propane burners and ovens were substantial (Table 2), nonzero (p < 0.01 for each; Table S1), and repeatable (coefficient of variation = 11% for replicates of gas ovens at 350 °F and gas burners on high up to three months apart). Mean benzene emission rates from gas burners on high and from ovens set to 350 °F were 2.8 [95% confidence interval (CI): 1.7, 4.6] and 5.8 [95% CI: 3.3, 9.7] μg C_6H_6 min^{-1}, respectively (Table 2). Mean benzene emissions from propane burners on high and ovens set to 350 °F were 5.5 [95% CI: 1.2, 11.0] and 6.5 [95% CI: 2.1, 12.6] μg min^{-1}, respectively (Table 2). Flames from higher-molecular-weight propane produce more benzene in the laboratory than flames from methane per joule,[25] so it would not be surprising if future stove studies with larger sample sizes find that propane stoves emit more benzene than gas stoves. With our dataset, however, we find that while the central estimates of mean benzene emissions from propane burners and from ovens were higher than from their gas counterparts, benzene emissions from gas and propane stoves were nevertheless statistically indistinguishable (p > 0.20 for each pairwise comparison). Median benzene emissions were lower than mean emissions

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Table 2. Benzene Emission Rates per Stove Type ($\mu g\ C_6H_6\ min^{-1}$)[a]

	median	lower bound	upper bound	mean	lower bound	upper bound
propane ovens at 350 °F	3.89	0.40	12.5	6.46	2.05	12.5
propane burners on high	1.91	0.14	7.08	5.48	1.20	11.0
gas ovens at 350 °F	1.91	1.33	3.76	5.82	3.26	9.70
gas burners on high	1.34	0.94	2.01	2.78	1.65	4.55
propane burners on low	0.47	0.18	1.02	3.98	0.28	11.0
coils and radiant on high	0.24	0.06	0.38	0.28	0.13	0.44
gas burners on low	0.21	0.16	0.27	0.47	0.26	0.73
electric ovens at 350 °F	0.11	−0.02	0.18	0.23	0.03	0.52
induction hobs on high	0.04	−0.06	0.17	0.01	−0.13	0.12
coils and radiant on low	0.00	−0.10	0.08	0.01	−0.05	0.09
induction hobs on low	−0.02	−0.16	0.11	−0.02	−0.23	0.23

[a]Median and 95% confidence interval from 2.5 to 97.5%; mean and 95% confidence interval from 2.5 to 97.5%, calculated with a 25,000 replicate bootstrap (see methods). Mean benzene emission rates for induction hobs on low and high as well as electric coils and radiant hobs on low were statistically indistinguishable from zero.

for both gas and propane stoves (Table 2) because emission distributions tended to be long-tailed (Figure 1). Grouping stoves by absolute power output (>1 kW and <1 kW) yielded similar results to grouping by relative power output shown on burners (high vs low, Table S3 and Figure S12).

Expressed per joule (see methods), mean emissions from gas burners on high were 0.014 [95% CI: 0.009, 0.021] $\mu g\ C_6H_6\ kJ^{-1}$, roughly one quarter the emissions per joule from ovens set to 350 °F (Figure S5). Mean emissions normalized per joule for burners on low were higher on average than for both ovens and burners on high but more variable: 0.080 [95% CI: 0.017, 0.192] $\mu g\ kJ^{-1}$. These benzene emission factors correspond to approximately 100 [95% CI: 70, 190] μg benzene per meal cooked on a gas stove for a typical meal cooked using one burner on "high", one on "low", and the oven on 350 °F each for approximately 30 min (see methods). We observed no statistically significant relationship between benzene emissions and stove brand or age (see Figure S10).

Benzene emissions from gas and propane burners and ovens were significantly greater than those from electric alternatives (Table 2 and Figure 1; $p < 0.01$ for all gas-electric comparisons, propane vs induction, and propane vs electric oven; $p = 0.075$ for propane vs coil/radiant on high; see Table S1). Benzene emissions from induction hobs on high were statistically indistinguishable from zero ($p > 0.2$; Table 2 and Figure 1a). Mean benzene emissions from coils and radiant hobs on high (grouped together) and from electric ovens were 0.28 [95% CI: 0.15, 0.42] and 0.23 [95% CI: 0.04, 0.52] $\mu g\ C_6H_6\ min^{-1}$, respectively, or 10 and 4% of the emissions from their gas counterparts (Table 2). Benzene emissions from electric coils, radiant stoves, and electric ovens may occur because of organic matter burning off the red-hot (> 500 °C) surface.

Benzene Concentrations in Home Air. We quantified kitchen benzene concentrations after stove use in a 17-home subset of our emission factor sample (using 33 distinct burner and oven measurements; see methods). A single gas or propane burner on high or oven set to 350 °F for 45 min raised kitchen benzene concentrations above the baseline in every kitchen that we tested (Figure 2). This result suggests that gas stoves can contribute substantively to elevated benzene concentrations indoors. Additionally, in 9 of the 33 cases (29%), a single gas burner on high or an oven set to 350 °F raised kitchen benzene concentrations above the upper range of indoor benzene concentrations attributable to secondhand tobacco smoke (0.34−0.78 ppbv)[10] and above the median indoor benzene concentration measured in the US, Canada, Western Europe, Japan, South Korea, Hong Kong, and Australia (0.78 ppbv).[47]

Benzene Migration to Bedrooms. To quantify the dispersion of pollutants from kitchens to bedrooms, we measured ambient benzene concentrations in bedrooms farthest from open kitchens of six houses (without using fans to mix the air) under a scenario with the oven set to 475 °F for an hour and a half and then turned off while measurements continued for another 6.5 h. During these measurements, we kept interior doors open (see Figure S1 for floorplans of the houses). House 1 (90 m^2) had a gas oven with the highest emissions that we measured, houses 2 (85 m^2) and 3 (70 m^2) had ovens with emissions between the mean and maximum, fueled by propane and gas, respectively, houses 4 (75 m^2) and 5 (140 m^2) had gas ovens with near-mean emissions, and house 6 (85 m^2) had a gas oven with below-average benzene emissions. In all six cases tested, burner or oven use elevated peak bedroom benzene concentrations between 5 and 70 times above baseline levels and in some cases beyond the California OEHHA acute and chronic RELs (Figure 3).[8]

In house 1, bedroom benzene concentrations peaked at 8.9 ppbv, slightly below the maximum value observed in the kitchen of 11.6 ppbv, and remained above the California OEHHA acute REL of 27 $\mu g/m^3$ (8 ppbv) for >20 min (Figure 3). By comparison, in 2020, there were two separate incidents in which benzene concentrations near schools in Greater Los Angeles and the Colorado Front Range reached between 8.84 and 15.3 ppbv, respectively.[48,49] These benzene measurements prompted local and state news coverage and investigations by authorities.[50,51] Benzene concentrations in house 1 also remained above the California OEHHA 8-h and chronic REL of 3 $\mu g/m^3$ (1 ppbv) for more than 5 h (Figure 3), and the 8-h time-averaged bedroom benzene concentration (2.8 ppbv) in house 1 was 23 times higher than the baseline bedroom benzene concentration prior to stove use.

Peak benzene concentrations that we measured in other houses were also substantially elevated over background but lower than in house 1. In house 2, bedroom benzene concentrations peaked at 4.2 ppbv and remained elevated above the California OEHHA 8-h REL for the entire duration of the 8-h measurement (Figure 3); the time-averaged bedroom benzene concentration over the 8-h time course was 2.4 ppbv, 26 times higher than the baseline benzene concentration. In house 3, bedroom benzene concentrations peaked at 2.9 ppbv and remained elevated above the California OEHHA 8-h REL for more than 5 h (Figure 3). In houses 4, 5, and 6, with lower benzene emission rates, benzene

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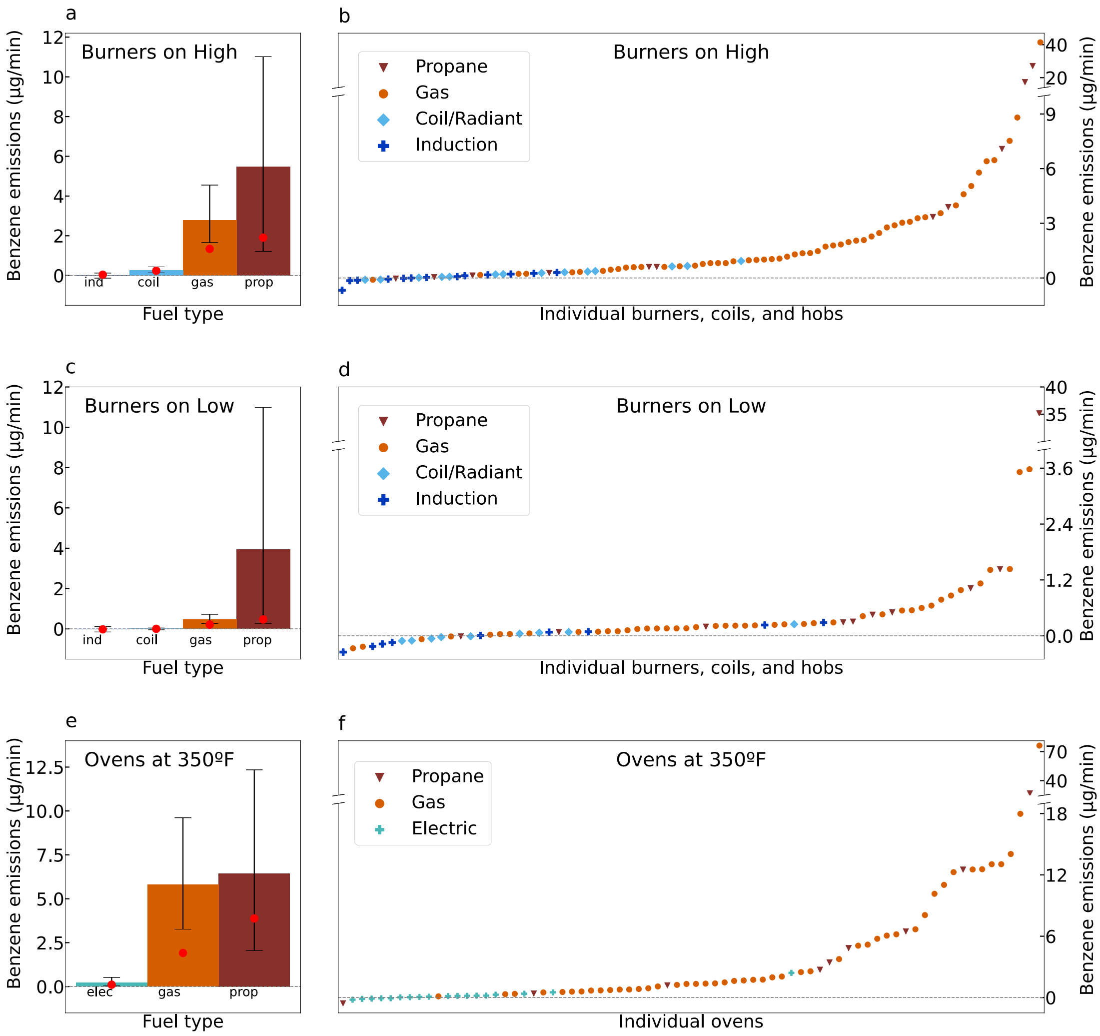

Figure 1. Mean and median benzene emissions by fuel type (left panels; induction, coil, natural gas, and propane) and emissions from individual cooktop elements and ovens (right panels) in $\mu g\ C_6H_6\ min^{-1}$ by appliance type (gas, coil/radiant, or induction) for burners on high (a,b) or on low (c,d) and for ovens set to 350 °F (e,f). The red points inside the bars in plots a, c, and e are the median values. Bar heights are the mean values, and black error bars are the 95% confidence interval of the mean (calculated as described in methods; see Table S1 for p values). Benzene emission rates were measured directly using the AROMA analyzer (see methods). Burners on "High" in panels a and b refer to the highest-power cooktop element on each stove set to its highest setting; "Low" refers to the lowest-power cooktop element on each stove set to its lowest functional setting. Oven emission rates include the oven preheat and oven cycling (to maintain its temperature) over the 45 m sampling interval. Note the y-axis breaks in panels b, d, and f.

concentrations in bedrooms during the oven test peaked at 0.69, 0.78, and 0.70 ppbv, respectively (Figure 3).

Ventilation and hood use affect benzene concentrations. Surveys show that ventilation hoods are used by residents only 25−40% of the time.[52] We tested the efficacy of kitchen range hoods by comparing concentrations with two outside-venting hoods on and off in houses 1 and 4. In house 1, kitchen benzene concentrations exceeded the California OEHHA 8-h and chronic REL, 3 $\mu g/m^3$ (1 ppbv), within 1 h of stove use whether the hood was off or on high (Figure S2). The comparison of hood-on high vs hood-off demonstrates that residential range hoods are not always effective at reducing pollutant concentrations, even if they vent outdoors, as highlighted in previous research.[52]

It is useful to compare other benzene sources to the benzene emissions that we observed attributable to combustion from gas stoves. With stricter emission control measures, benzene concentrations have fallen in many parts of the world over recent decades. For instance, average outdoor benzene concentrations in California fell from 2.5 ppbv in 1990 to 0.3 ppbv in 2012.[53] The indoor benzene concentrations that we measured resulting from combustion by gas stoves are thus substantial relative to ambient concentrations in many parts of the world, especially Europe and the Americas.[9] Unfortunately, however, high benzene concentrations persist in other parts of the world; throughout the 2000s and 2010s, benzene concentrations exceeding 10 ppbv were often measured in several East and South Asian cities, and concentrations exceeding 100 ppbv have been reported in Mumbai, India.[9] In places with extremely high benzene pollution, other larger benzene sources (such as from burning solid fuels[30]) may be more important than pollution from gas stoves.

Benzene Emissions from Food and from Unburned Gas. Benzene emissions from cooking include combustion-related emissions from the stove, potential emissions from food as it cooks, and any emissions from unburned gas leaking from or near the stove (because gas entering homes has been shown to often contain benzene).[41,57] To isolate emissions produced

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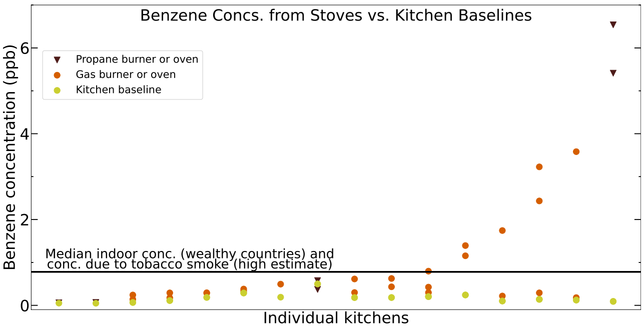

Figure 2. Benzene concentrations in unsealed kitchens as measured immediately after being aired out with outdoor air (mustard circles) and after 45 min with one burner on high or oven set to 350 °F fueled by propane (brown triangles) or gas (orange circles). Each point represents a single measurement; each *x*-value represents a single kitchen; multiple dots in the same column represent measurements of different burners or ovens in the same kitchen. These measured kitchen concentrations are compared with the upper estimate of benzene concentrations attributable to secondhand tobacco smoke[10] (0.78 ppbv) and the median indoor benzene concentration in the US, Canada, Western Europe, Japan, South Korea, Hong Kong, and Australia derived from a literature survey[47] (0.79 ppbv).

from food while cooking from those associated with fuel combustion, we cooked two meal types on induction cooktops that we determined had benzene emissions indistinguishable from zero. We chose these two foods, fish and bacon, because a previous study found that pan-frying these foods on a gas cooktop produced more polycyclic aromatics (which, like benzene, are partial-combustion products) than any other foods or cooking conditions that the authors tested.[46] Benzene emissions from the foods that we evaluated were indistinguishable from zero ($p > 0.35$; Figure S3). Our results for these foods suggest that the benzene produced when cooking with gas and propane comes from the fuel burned and not from the food being cooked, at least for these two common meal types. Future research on a broader suite of meal types is warranted.

Benzene emissions from gas use also come from any unburned gas that leaks into the kitchen and that contains trace levels of benzene.[41] Recent studies that sampled uncombusted gas directly from residential stoves in Boston and California measured the concentrations of benzene and other hazardous air pollutants in the gas delivered to homes.[41,57] Assuming that gas is 95% methane, a stove leaking gas with a median benzene concentration for California (1.6 ppmv)[41] at the median rate (24 mg methane h^{-1})[2] would emit 0.003 μg min^{-1} of benzene. Such a steady-state off emission of benzene in the smallest kitchen partition that we measured (8.3 m^3) with no air exchange would result in an undetectable concentration increase over our 45 min measurement period using our AROMA analyzer. In contrast, we measured median benzene emissions from gas combustion in burners on high and on low and from ovens that are 450, 70, and 640 times greater than this median steady-state off leak rate (Table 2). Thus, typical benzene emission rates attributable to such gas leakage are small relative to the emission rates we measured attributable to gas combustion.

Benzene Emissions as Partial-Combustion Products. When gas burns in a fuel-rich (oxygen-poor) flame, it does not combust fully to water and carbon dioxide. Some partial-oxidation products known to form under these conditions include carbon monoxide, formaldehyde, polycyclic aromatics, and monocyclic aromatics such as benzene.[25] We hypothesized that benzene and carbon monoxide emissions would therefore be correlated, which turned out to be correct ($r^2 = 0.67$, $p < 0.01$, Figure S4). This relationship suggests that both benzene and carbon monoxide may be produced in similar partial-

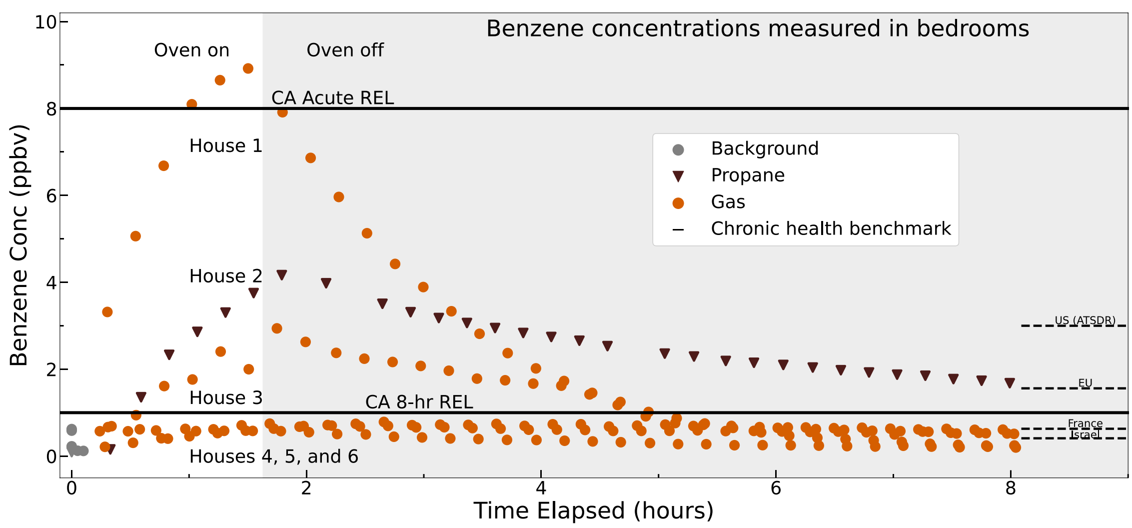

Figure 3. Benzene concentrations (ppbv C_6H_6) measured in bedrooms furthest from open kitchens with the oven set to 475 °F for 1.5 h and then turned off for 6.5 h. Each point represents a single reading from the AROMA analyzer, and all data points collected are illustrated in the plot. Air-sampling hoses in houses 1, 3, 4, 5, and 6 were placed in bedrooms ~8 meters down the hall from the kitchen; the sampling hose in house 2 was in a bedroom ~4 meters down the hall from the kitchen. The solid black line at 1 ppbv benzene represents the California OEHHA 8-h REL for noncancer effects;[8] the solid black line at 8 ppbv benzene represents the California OEEHA acute REL.[8] Several chronic benzene standards are plotted for comparison at the right with dashed black lines for Israel[54] and France[55] at the bottom (0.41 and 0.63 ppbv, respectively), two of the lowest chronic benzene standards globally; the European Union chronic health benchmark (1.56 ppbv), which is the most common chronic health benchmark globally;[9] and, at the top, the chronic standard adopted by the US ATSDR (3.0 ppbv), a department affiliated with the Centers for Disease Control and Prevention.[56] We measured all concentrations in real time using the AROMA VOC analyzer. During the measurements, all interior doors of the houses were open, and no fans were used to mix the air.

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combustion conditions in gas and propane stoves and ovens. Similarly, other unstudied partial-combustion pollutants could also be produced, a possibility that warrants future research.

Benzene Emissions from Gas Combustion in Stoves Compared with Current EPA Estimates and Other Benzene Sources. Our results suggest that the U.S. EPA substantively underestimates benzene emissions from total residential gas combustion. Our results show that gas burners on high and gas ovens set to 350 °F (grouped together) emit 0.059 [95% CI: 0.037, 0.088] μg benzene kJ^{-1} gas burned. Scaling up our measured emission factor, we estimate that ~7200 [95% CI: 4200, 12,800] kg of benzene is emitted by gas combustion by stoves alone in the U.S. annually (see methods). In contrast, the EPA's National Emissions Inventory estimates that all residential gas appliances (including furnaces and water heaters) emit 4300 kg of benzene in total annually,[58] slightly more than half of our estimate for stoves alone. This discrepancy likely arises because the EPA measured benzene emission factors from utility-scale boilers rather than from home appliances and used these values to calculate residential emissions.[29,58,59] The EPA's boiler-derived benzene emission factor for natural gas combustion of 2.1×10^{-3} lb benzene per 10^6 standard cubic foot of gas (0.0009 μg benzene kJ^{-1} gas burned)[29] is 65 times lower than our estimate. The EPA relies on the same boiler-derived benzene emission factor to estimate emissions from water heaters, furnaces, and stoves. If the benzene emission factors from water heaters and furnaces are similar to those from stoves, then the discrepancy between current EPA estimates and true benzene emissions due to residential gas combustion may be even higher than presented here.

The total quantity of benzene emitted attributable to gas combustion in stoves is small relative to some other large benzene sources but could have a disproportionate effect on health because the benzene from combustion is emitted directly indoors. According to the EPA, the largest sources of benzene in the U.S. are wildfires (45 million kg $year^{-1}$), gasoline-powered light-duty vehicles (37 million kg $year^{-1}$), and oil and gas production (25 million kg $year^{-1}$).[60] Large sources of benzene should certainly be minimized wherever possible, but benzene emitted outdoors can quickly dissipate and degrade.[61] As a result, outdoor and indoor benzene emissions are not directly comparable for their health effects on a kg-for-kg basis.

Study Implications and Suggestions for Future Research. Gas stoves are common in U.S. homes and around the world, and their emissions can alter the indoor air quality of people who live and work around them.[2,7,11−13,20,21,62] We quantified benzene emission factors from combustion in gas and propane stoves, to our knowledge for the first time. Our findings suggest that the concentrations of benzene produced by combustion from gas stoves and ovens indoors may increase health risks under some conditions. Further research is needed to assess actual exposures and the full health impacts of benzene emitted indoors from combustion by gas stoves. We also showed that using a gas burner or oven may increase kitchen and bedroom benzene concentrations above chronic exposure guidelines, depending on ventilation conditions and home size. By measuring benzene concentrations in bedrooms during and after gas burner and oven use, we also showed that benzene produced from gas stoves migrates well beyond the kitchen. People outside the kitchen can be exposed to elevated levels of benzene for hours after the stove is turned off.

Our work also shows that gas and propane fuels appear to be the dominant source of benzene produced by cooking. We found that benzene emissions from induction stoves and from cooking food were undetectable (i.e., statistically indistinguishable from zero), at least for the two meals that we evaluated. We further found that mean emissions from electric coils and radiant hobs on high and electric ovens on 350 °F were 10- to 25-fold smaller than emissions from gas and propane stoves. Average emissions from coils and radiant hobs on low were statistically indistinguishable from zero.

To understand and model benzene exposure indoors, researchers need emission rates, as we provide here, and better data for stove use, hood use, and hood type. For instance, researchers have very little data on how often multiple burners are used simultaneously, how often burners and ovens are used together, and how frequently the oven is used to heat the home (a practice that is not recommended but that we nevertheless encountered multiple times during our home surveys).

A better understanding of indoor pollutants and air quality is also needed, in part because people spend much of their time indoors. One study of 1760 California residents, for instance, determined that people spent 87% of their time indoors.[63] Our results show that gas and propane combustion by stoves emits benzene directly into indoor air. These results highlight the importance of combustion by gas stoves for indoor air quality and human exposure in future policies designed to protect people from air pollution, particularly people in lower-income neighborhoods with smaller home sizes.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c09289.

Additional details for air exchange calculations and assumptions underlying calculations of benzene emissions from a single meal; table of *p*-values for pairwise comparisons of benzene emissions from gas and propane burners on high, on low, and from ovens; summary of attributes of all stoves measured and locations sampled for the study, including the map of sampling locations; floorplans of houses in which bedroom benzene concentrations were measured; kitchen benzene concentrations measured with a hood on and off; photo showing the setup and plot showing results for measurement of benzene emissions from food cooked on an induction stove; benzene emissions from gas stoves plotted against accompanying carbon monoxide emissions; benzene emissions expressed per joule of gas burned; kitchen chamber volumes used in emission rate measurements; image of the sampling setup for emission rate measurements; calculated and actual benzene emissions rates from controlled-release tests; benzene emissions by stove age and brand; gas and propane burner power output; benzene emissions from propane stoves grouped by absolute power output (PDF)

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Author Contributions

R.B.J., S.B.C.S., and Y.S.K. conceived the project. Y.S.K., M.N., C.F., Z.O., R.B.J., and E.D.L. developed the methods for the project. Y.S.K., M.N., C.F., and Z.O. performed most of the field measurements. E.D.L. and D.R.M. assisted with methods and data interpretation. Y.S.K. wrote the manuscript with assistance from R.B.J., and all authors edited the final version. R.B.J. supervised the project.

Notes

The authors declare no competing financial interest.

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