Supporting Information for Kashtan et al. (Gas and Propane Combustion from Stoves Emits Benzene and Increases Indoor Air Pollution)

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Supplementary Tables

Table S1. p values comparing pairs of cooktop element and oven categories calculated using the two-sided Mann-Whitney U test

	Gas burners on high	**Coils and radiant on high**	**Induction on high**	**Propane burners on high**	**Zero**
Gas burners on high	1.0				
Coils and radiant on high	$4.5x10^{-6}$	1.0			
Induction hobs on high	$1.35x10^{-7}$	0.031	1.0		
Propane burners on high	0.73	0.075	$3.1x10^{-3}$	1.0	
Zero	$1.7x10^{-10}$	$9.6x10^{-5}$	0.21	$1.6x10^{-5}$	1.0
	Gas burners on low	**Coils and radiant on low**	**Induction on low**	**Propane burners on low**	**Zero**
Gas burners on low	1.0				
Coils and radiant on low	$1.7x10^{-3}$	1.0			
Induction on low	$5.8x10^{-3}$	0.79	1.0		
Propane burners on low	0.20	$2.9x10^{-3}$	$4.8x10^{-3}$	1.0	
Zero	$1.6x10^{-7}$	0.58	0.58	$3.1x10^{-5}$	1.0
	Gas ovens at 350ºF	**Electric ovens at 350ºF**		**Propane ovens at 350ºF**	**Zero**
Gas ovens at 350ºF	1.0				
Electric ovens at 350ºF	$2.2x10^{-8}$	1.0			
Propane ovens at 350ºF	0.66	$1.8x10^{-3}$		1.0	
Zero	$7.0x10^{-11}$	0.012		$6.4x10^{-5}$	1.0

Table S2. Summary of stove types and locations sampled.

Homes	87
States	California and Colorado
Counties	14
Cooktop elements tested	175
Ovens tested	74

Stove age range (years)	3 to 75 (upper end approx.)
Largest gas burner power (kBTU h^{-1})	9.5 to 17
Gas oven power (kBTU h^{-1})	16 to 19
Largest electric coil power (kBTU h^{-1})	7.2 to 15.7

Table S3. Benzene emission rates from propane burners (µg C_6H_6 min^{-1}), grouped by relative power (high or low) and absolute power (> 1kW or < 1kW). Gas burners are not shown because the power output of gas stoves measured on high and on low did not overlap (i.e., all gas burners on high were > 1 kW and all on low were <1kW), so both grouping methods yield the same outcomes. Electric cooktop elements are not shown because they do not emit CO_2 and their power was therefore not measured directly. Mean and 95% confidence interval from 2.5% to 97.5%; median and 95% confidence interval from 2.5% to 97.5%, calculated with a 25,000 replicate bootstrap (see Methods).

	Median	Lower Bound	Upper Bound	Mean	Lower Bound	Upper Bound
Propane burners on high	1.91	0.14	7.08	5.48	1.20	11.0
Propane burners >1kW	1.55	0.29	3.90	4.95	2.14	8.36
Propane burners <1kW	1.96	0.33	18.3	6.46	0.47	18.0
Propane burners on low	0.47	0.18	1.02	3.98	0.28	11.0

Supplementary Methods

Correction for air exchange

Because it is impossible to seal kitchens perfectly, we corrected for air exchange between the chamber and the air outside the chamber. We calculated the air exchange constant for each kitchen by injecting 500-ml volumes of ethane and measuring changes in concentration through time as described in the Methods and in Lebel et al.[1].

Kitchen volume is calculated using Eq. S1:

$$V_k = \frac{V_i}{C_{e,peak}} \qquad \text{S1}$$

where V_k is the kitchen volume, V_i is the volume of injected ethane, and $C_{e,peak}$ is the peak ethane concentration following injection.

The concentration of ethane after injection follows an exponential decay attributable to air exchange and is described by Eq. S2:

$$C_{e,t} - C_{e,b} = C_{e,0}\, e^{-\lambda t} \tag*{S2}$$

where $C_{e,t}$ is the concentration of ethane at time t, $C_{e,b}$ is the background concentration of ethane, $C_{e,0}$ is the concentration of ethane in the kitchen prior to injection (typically very close to background), t is time, and λ is the air exchange constant.

Rearranging, we can calculate the air exchange constant λ using Eq. S3:

$$\lambda = \frac{\ln\left(\frac{C_{e,0}}{C_{e,t} - C_{e,b}}\right)}{t} \tag*{S3}$$

Then, the corrected gas concentration $\hat{C}_{g,t}$ for the nth datapoint collected is given by Eq. S4:

$$\hat{C}_{g,t} = \; C_{g,0} + \Sigma_{i=1}^{n}\left(C_{g,t} - C_{g,b}\right)e^{-\lambda(t_i - t_{(i-1)})} \tag*{S4}$$

where $\hat{C}_{g,t}$ is the corrected gas concentration,$C_{g,b}$ is the background gas concentration, $C_{g,t}$ is the gas concentration at time t, and $C_{g,0}$ is the initial gas concentration.

The flowrate of the gas can then by calculated using the linear model given by Eq. S5:

$$f_g = \frac{V_k\left(\hat{C}_{g,t} - C_{g,0}\right)}{t} \tag*{S5}$$

where f_g is the gas flowrate (expressed as volume per time).

We used Eq. S5 to calculate flowrates for CH_4, CO, and CO_2. This method is the same as that used by Lebel et al.[1].

Using the decay constant for a given kitchen derived using the ethane tracer gas, we calculated corrected benzene flowrates and a correction for the measured benzene concentration appropriate for three or four data points collected by the AROMA analyzer during each measurement:

The instantaneous decay rate of benzene in the kitchen chamber follows Eq. S6:

$$\frac{dC_{b,t}}{dt} = \; -\lambda\left(C_{b,t} - C_{b,b}\right) + r_b \tag*{S6}$$

where $C_{b,t}$ is the concentration of benzene at time t, λ is the kitchen chamber's air exchange constant derived above, $C_{b,b}$ is the background benzene concentration outside the kitchen chamber, and r_b is the benzene emission rate from the stove, expressed as concentration per time (for instance, if the emission flowrate were 1mL/hour and the kitchen chamber were 100,000L, r_b would be 10^{-8} hr^{-1}).

This differential equation has solutions of the form expressed in Eq. S7:

$$C_{b,t}(t) = \frac{C_{b,b}\lambda + r_b}{\lambda} + c_1 e^{-\lambda(t-t_0)} \tag{S7}$$

where c_1 is an integration constant and t_0 is the start time of the measurement. Letting $C_{b,0}$ be the concentration of benzene at the beginning of the measurement and applying the initial condition $C_b(t_0) = C_{b,0}$ to Eq. S7 yields $c_1 = C_{b,0} - \frac{C_{b,b}\lambda + r_b}{\lambda}$ and thus Eq. S7 becomes Eq. S8:

$$C_{b,t}(t) = \frac{C_{b,b}\lambda + r_b}{\lambda} + (C_{b,0} - \frac{C_{b,b}\lambda + r_b}{\lambda})e^{-\lambda(t-t_0)} \tag{S8}$$

We measure $C_{b,t}$ but want to know the true emission rate, r_b. Re-arranging to isolate r_b yields Eq S9:

$$r_b = \lambda(\frac{C_{b,t} - C_{b,0}e^{-\lambda(t-t_0)}}{1-e^{-\lambda(t-t_0)}} - C_{b,b}) \tag{S9}$$

The overall benzene emission rate for a given measurement with n AROMA data points (typically 4 and not fewer than 3) is then calculated by averaging each of the r_b values calculated between each data point, according to Eq. S10:

$$r_{b_{overall}} = \frac{1}{n}\ \Sigma_{i=0}^{n}\lambda(\frac{C_{b,t_i} - C_{b,t_{i-1}}e^{-\lambda(t_i-t_{i-1})}}{1-\ e^{-\lambda(t_i-t_{i-1})}} - C_{b,b}) \tag{S10}$$

For a perfectly sealed chamber with λ = 0, note that:

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

We can then multiply by volume to get the benzene flowrate (in volume per time), analogous to the flowrate for CH_4, CO, and CO_2 expressed in Eq. S5:

$$f_b = V_k r_{b_{overall}} \tag{S11}$$

where V_k is kitchen volume, as above.

We can also use Eq. S10 to derive an equation for corrected kitchen chamber benzene concentrations, analogous to the corrected concentrations for CH_4, CO, and CO_2 calculated using Eq. S4.

We assume that the benzene emission rate is constant after the first data point, once the cooking element has reached a constant temperature. The change in corrected benzene concentration between two measurements is $r_b(t_i - t_{i-1})$. The corrected benzene concentration at the end of the measurement is the sum of these differences plus the initial benzene concentration, $C_{b,0}$ This is given by Eq S11:

$$\hat{C}_{b,t} = C_{b,0} + \Sigma_{i=1}^{n} \lambda \left(\frac{C_{b,t_i} - C_{b,t_{i-1}} e^{-\lambda(t_i - t_{i-1})}}{1 - e^{-\lambda(t_i - t_{i-1})}} - C_{b,b} \right) (t_i - t_{i-1}) \qquad \text{S11}$$

where $\hat{C}_{b,t}$ is the corrected benzene concentration.

Benzene Emissions from Cooking 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 reported here and from previous research tracking burner and oven usage in 70 California homes[2]. The usage dataset tracks the number of a minutes that the cooktop or the oven was on during different periods of the day over 6 – 8 days (depending on the home). The dataset does not record the number of burners used or the burner intensity, so we assumed that when the cooktop was noted as "on," one burner was on high and one burner was on low. We assumed that all ovens noted as "on" were set to 350ºF. We assumed that stove use between 7am – 11am was associated with breakfast, between 11am – 1pm was associated with lunch, and between 5pm – 9pm with dinner. In the dataset, cooktops were "on" for an average of 28 minutes during each of these three time periods and ovens were on for an average of 29 minutes. Combining these averages with our assumptions about burner number and intensity and oven temperature, we define cooking a "meal" as having one burner on high and one burner on low for 28 minutes and setting the oven to 350ºF for 29 minutes. We then used per Joule benzene emissions reported here to calculate per-meal benzene emissions. Uncertainty in benzene emissions dominated uncertainty in usage (given our above assumptions), so we calculated our final estimate based on the 95% CI of benzene emissions.

Supplementary Figures

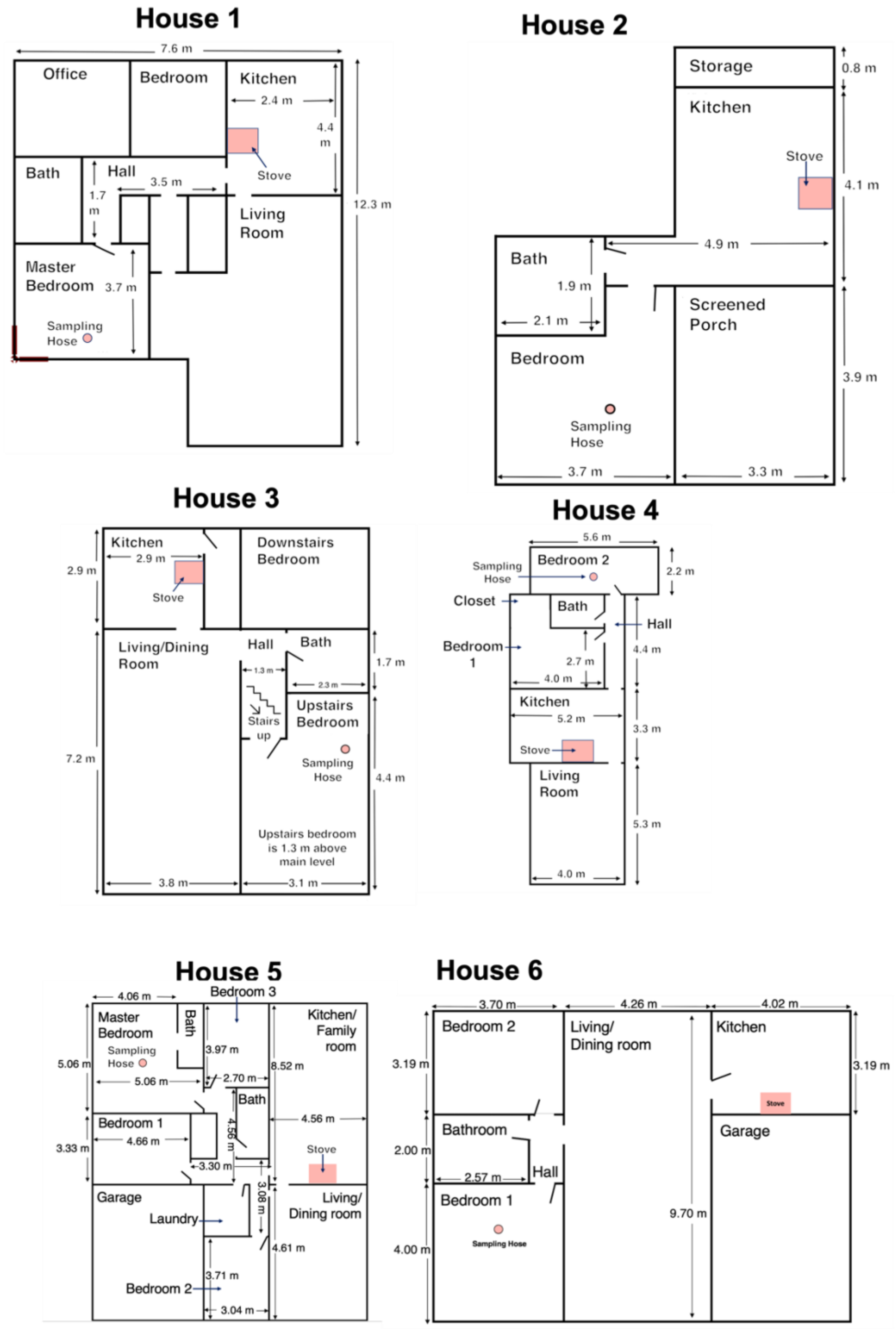

Figure S1. Floorplans of the houses in which we took the six 8-hour time course benzene measurements: House 1 (90 m^2), House 2 (85 m^2), House 3 (70 m^2), House 4 (75 m^2), House 5 (140 m^2), and House 6 (85 m^2). The location of the kitchen stove is marked with a pink square and the bedroom sampling location is marked with a pink dot.

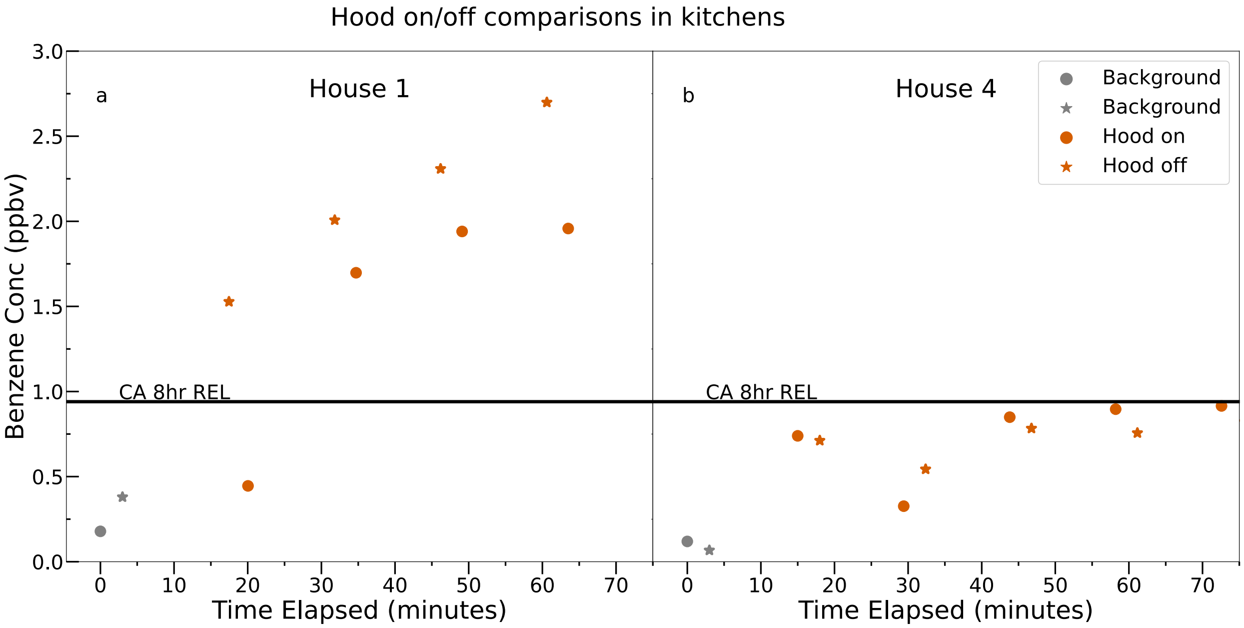

Figure S2. Benzene concentrations (ppbv) over time in un-sealed (no plastic) kitchens with gas stoves and their hoods on and off. Benzene concentrations measured in the kitchens (> 1 meter away from the stove) of House 1 (a) and House 4 (b) with three gas burners on high and the gas oven set to 350ºF. The black line at 0.94 ppbv benzene is the California OEHHA 8-hour REL for non-cancer effects [3]. Grey circles and stars represent background benzene concentrations in kitchens for hood on and off runs, respectively; orange circles represent measurements taken with the hood on and stove lit; orange stars represent measurements taken with the hood off and stove lit. All concentrations were recorded in real-time using the AROMA VOC analyzer. During measurements, all interior doors of the houses were open. House 1 had a hood with no discernable make, model, or serial number. House 4 had a General Electric Co. integrated microwave oven/range hood, model PVM 1970DR1CC.

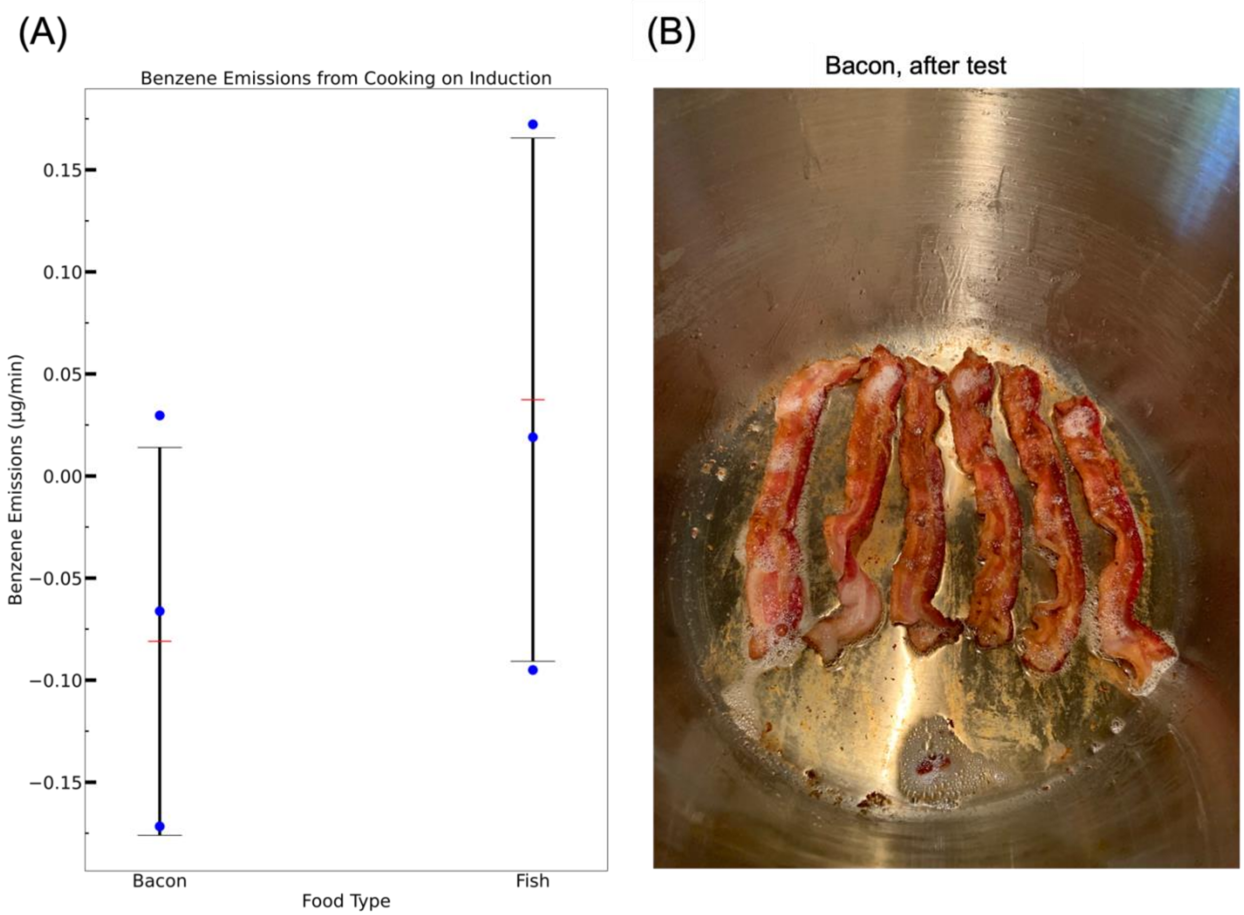

Figure S3. A) Benzene emissions in µg C_6H_6 min^{-1} from two foods, bacon (left) or fish (right) (n=3 for each food type). Blue points represent individual reps, red bars represent the means, and black error bars represent one standard deviation above and below the means. (B) Image of bacon after the frying test.

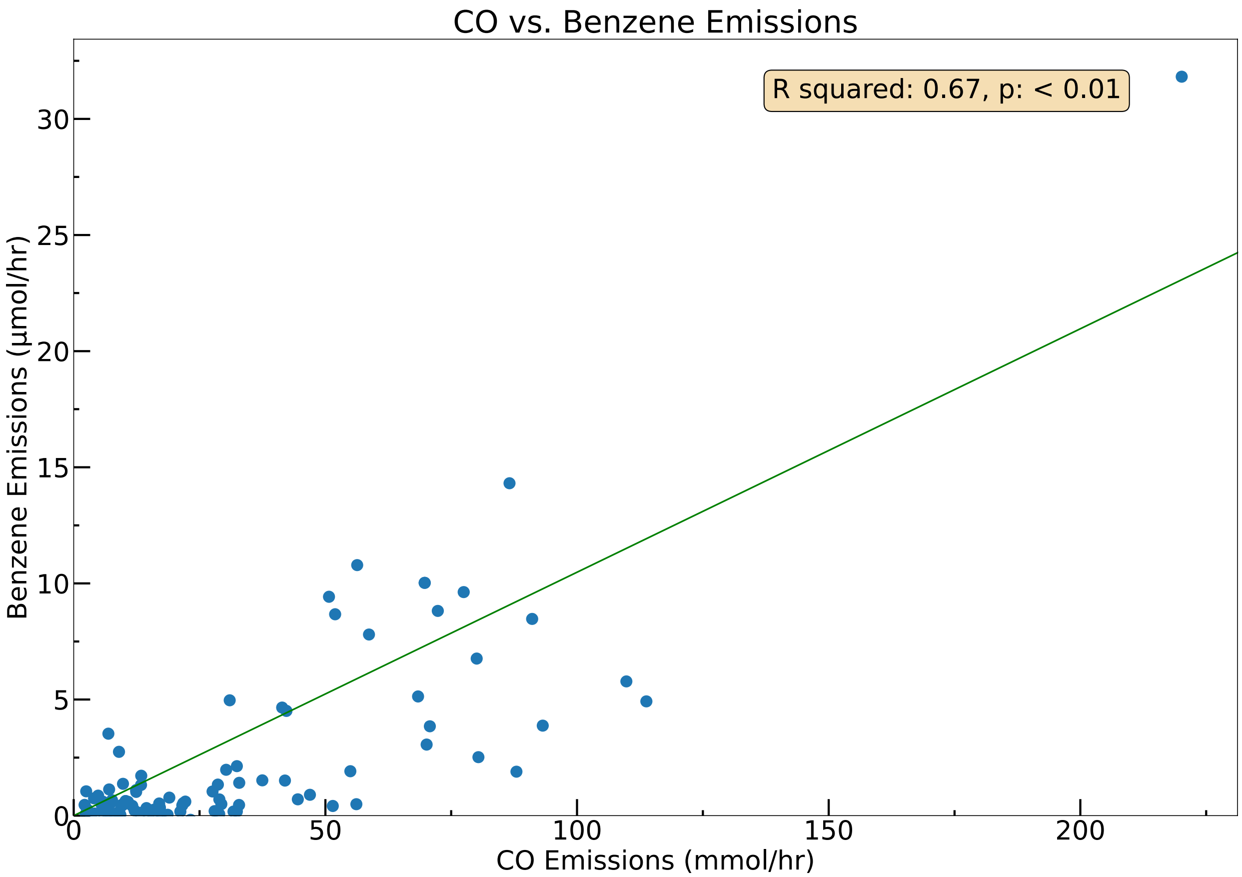

Figure S4. Benzene emission rate (µmol C_6H_6/hr) vs. carbon monoxide (CO) emission rate mmol CO/hr for 80 gas burners and ovens. Each light blue point represents an emission rate from a single burner or oven. Emissions of benzene and CO were directly measured using our kitchen-partition approach (see Methods), with concentrations measured in real-time. Benzene was measured using the AROMA analyzer and CO was measured using the Los Gatos Research analyzer (U-MCEA). We included every gas burner and oven for which we measured both benzene and CO emissions. Removing the potential outlier in the top right-hand corner still results in an R^2 of 0.49 with a p value still <0.01.

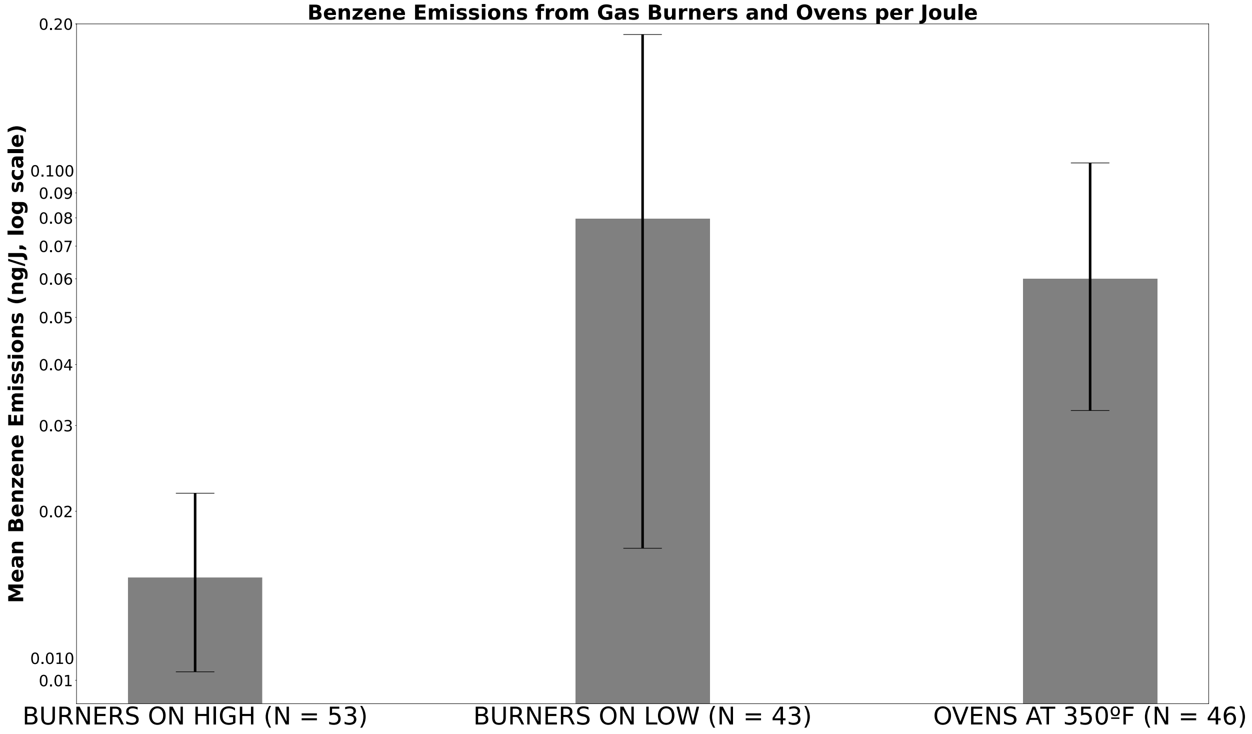

Figure S5. Mean benzene emissions from gas burners and ovens normalized by Joules of energy released (ng/J) on a log scale. Joules consumed were calculated based on the amount of CO_2 emitted and the enthalpy of combustion of methane (see Methods and Lebel, et al. 2022)[1].

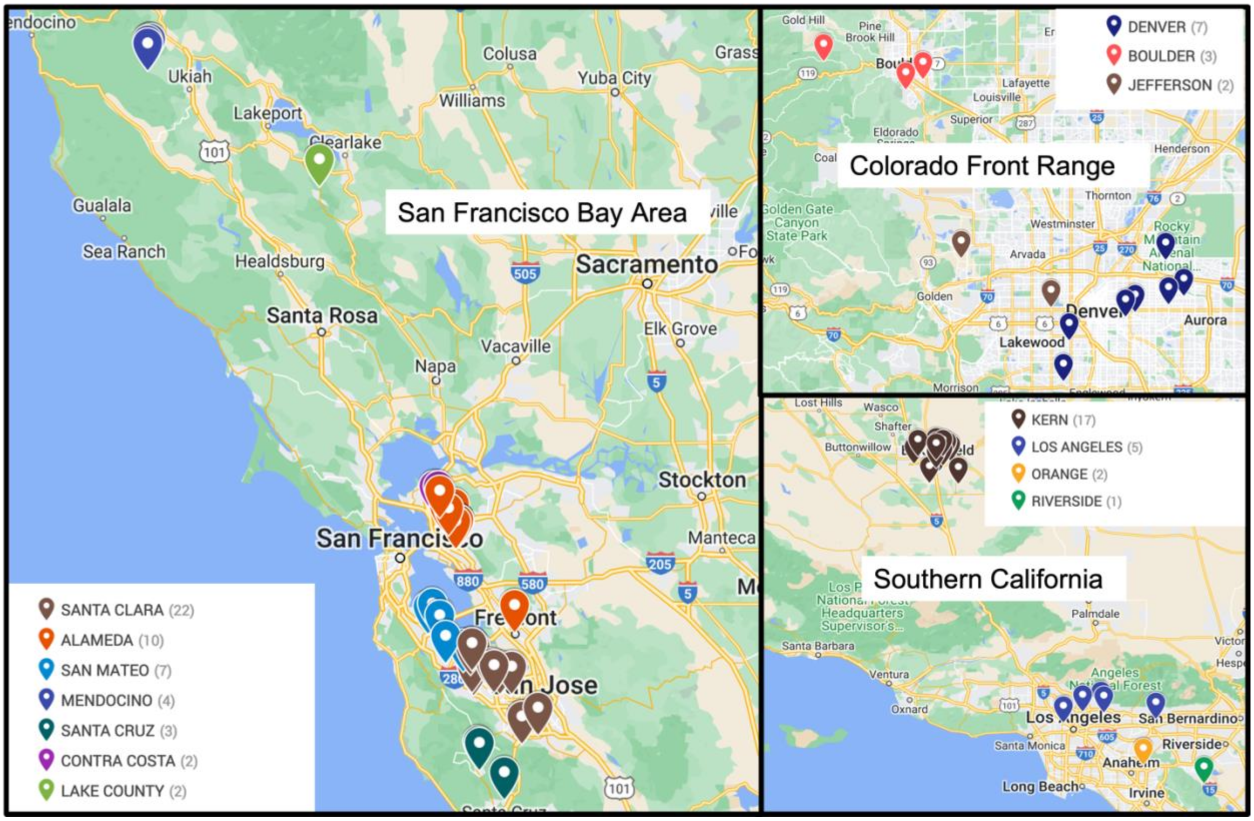

Figure S6. Geographical distribution of homes sampled for stove emissions, with house totals in parentheses and color-coding by county. We typically measured one burner on high, one burner on low, and one oven set to 350ºF in each home. Some houses lacked an oven, in which case we measured only burners. Map produced using Google MyMaps.

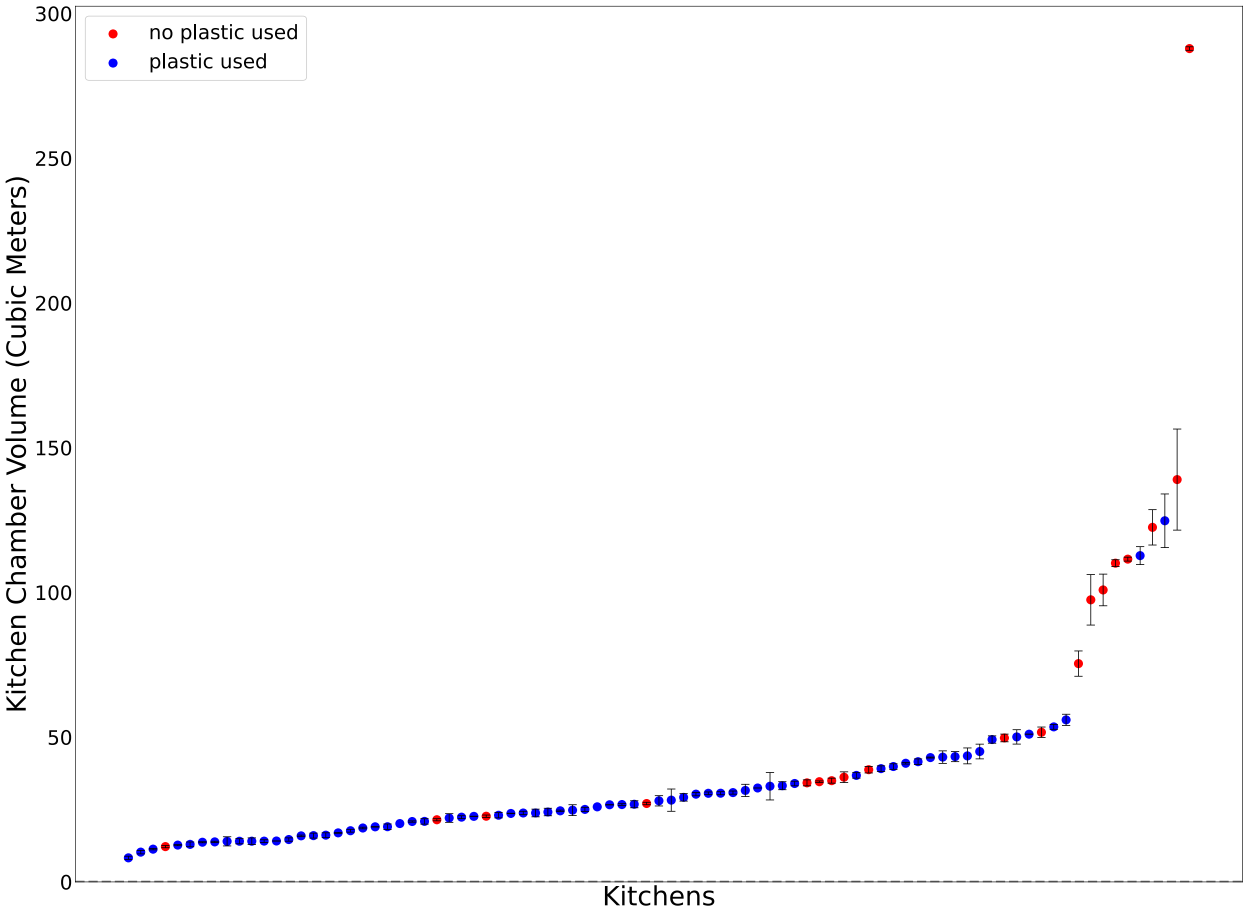

Figure S7. Tented kitchen chamber volume for each house we sampled, calculated with a 500mL ethane tracer gas injection (see Methods and Lebel et al.[1]). Points represent the mean estimated volume for each kitchen based on 2-4 injections (typically 3). Blue points are sealed kitchens and red points are unsealed kitchens (see Methods). Black error bars represent the standard deviation of measurements for a given kitchen.

Figure S8. Image of a typical setup for emission factor measurements. In this home plastic sheeting seals off openings outside of the kitchen to yield a known kitchen volume, and all windows and doors to the outside are closed. Two fans are placed > 1m away from the stove, pointed upwards, and placed on the "low" setting to circulate air in the chamber but not interfere with the burner combustion. A stainless-steel pot filled with tap water is placed on top of the burner. The PTFE sampling hose is placed >1m away from the stove at head-height and attached to a 7L/min pump (off screen). This pump draws kitchen chamber to the analyzers outside the kitchen. Plastic sheeting was not used in the ambient concentration measurements reported in Figures 2 and 3.

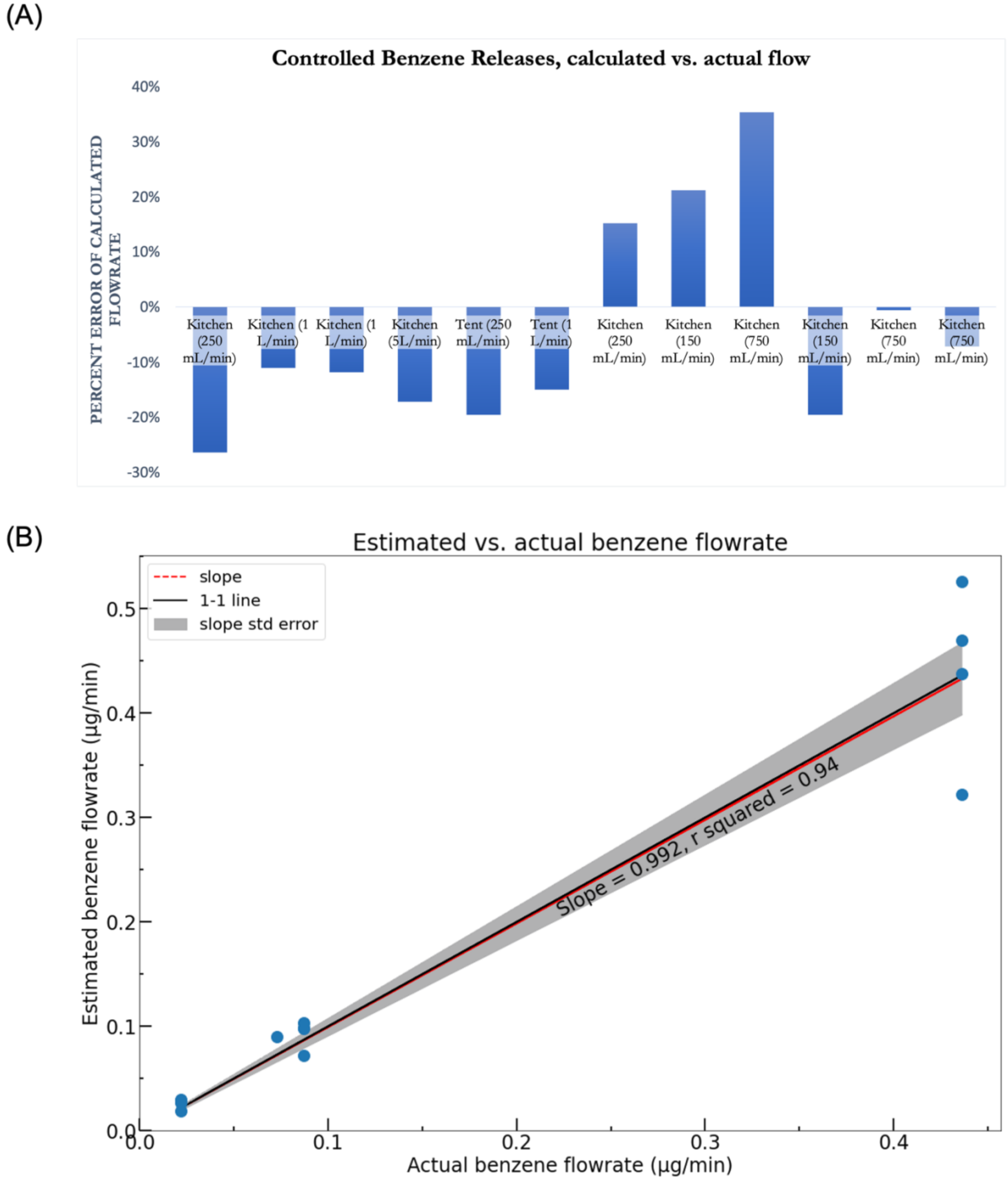

Figure S9. Results of methods validation using controlled releases of a known benzene standard (slope = 0.992, standard error of slope = 0.08, $r^2 = 0.94$, $p = 1.6 \times 10^{-7}$). The setup of the validation tests was identical to typical emission factor measurements, except that benzene emissions from the stove were replaced by a mass flow controller emitting a known flowrate of 1.5ppmv or 10ppmv benzene in nitrogen. To validate the methodology we used a 30,050-L tent and a kitchen with a volume of 27,200 L. (A) Percent error between actual and estimated benzene flowrates. Error was calculated as $(r_{calc} - r_{real})/(r_{real})$, where r_{calc} is the calculated emission rate and r_{real} is the actual emission rate from the mass flow controller. (B) Estimated vs. actual benzene flowrate.

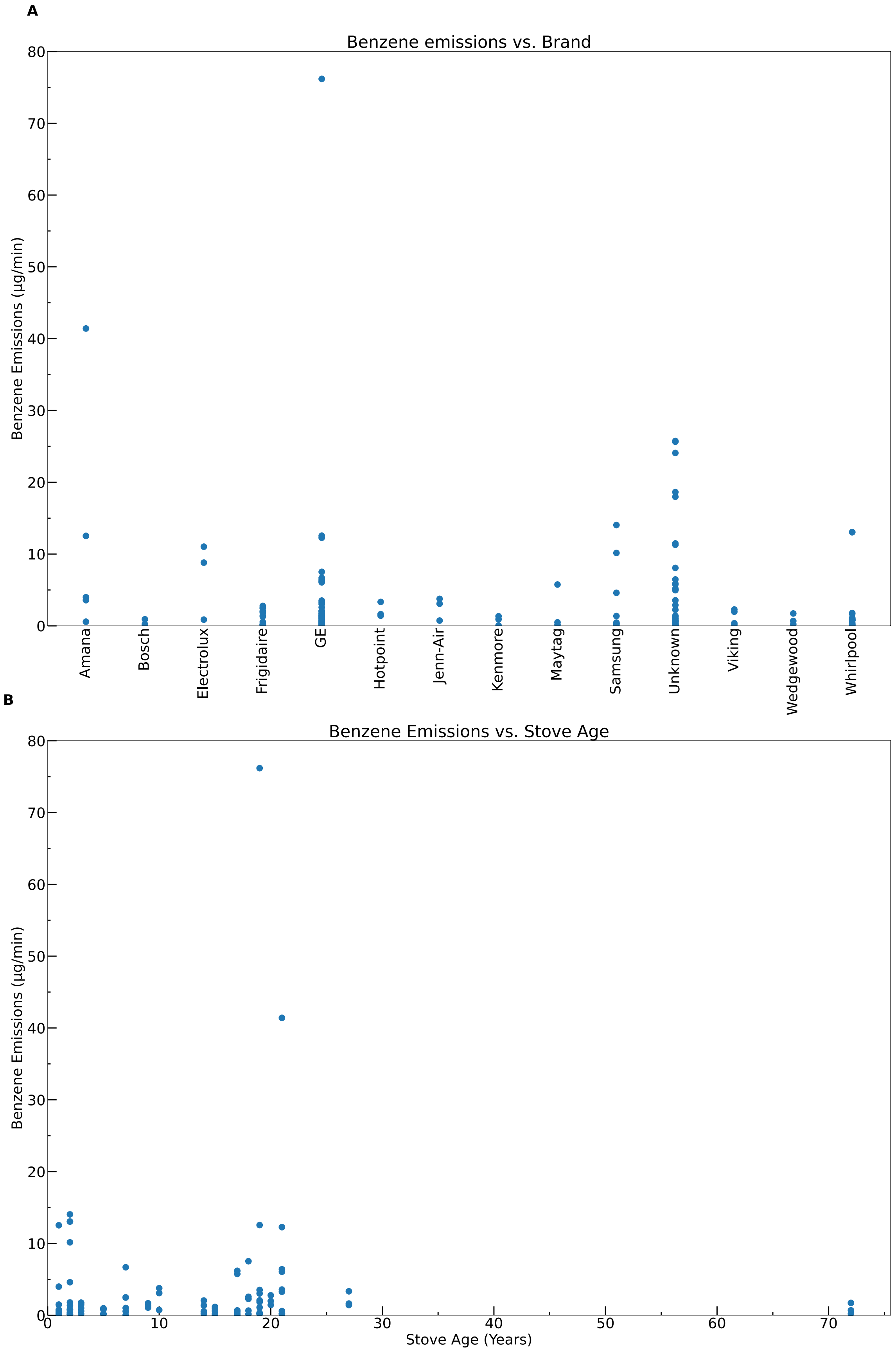
A
Benzene emissions vs. Brand
Benzene Emissions (µg/min)
0
10
20
30
40
50
60
70
80
Amana
Bosch
Electrolux
Frigidaire
GE
Hotpoint
Jenn-Air
Kenmore
Maytag
Samsung
Unknown
Viking
Wedgewood
Whirlpool
B
Benzene Emissions vs. Stove Age
Benzene Emissions (µg/min)
0
10
20
30
40
50
60
70
80
Stove Age (Years)
0
10
20
30
40
50
60
70

Figure S10. Benzene emissions from gas stoves (µg/min) plotted against (A) stove brand and (B) stove age. There is no clear relationship between benzene emissions and stove age ($r^2 = 0.002$, $p = 0.64$) or stove brand. Stoves for which we were unable to ascertain brand or age are omitted. When a stove serial number corresponded with multiple possible manufacture dates (typically ≥ 10 years apart), we estimated age based on visual inspection and/or consultation with the resident.

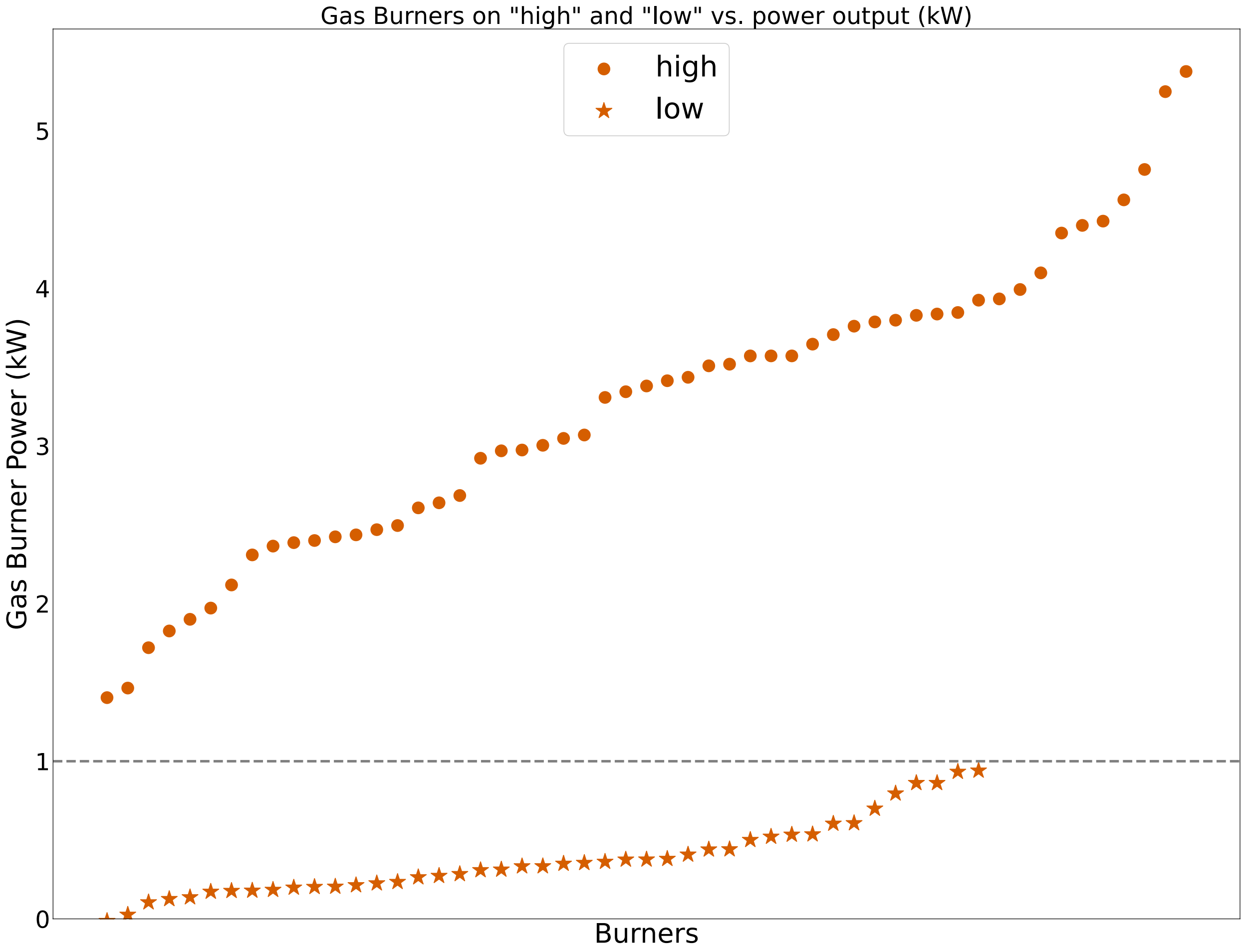

Figure S11. Gas burner power as quantified by CO_2 emissions. Orange circles represent burners set to high; orange stars represent burners set to low. The least powerful burner we measured on "high" was more powerful than the most powerful burner on low.

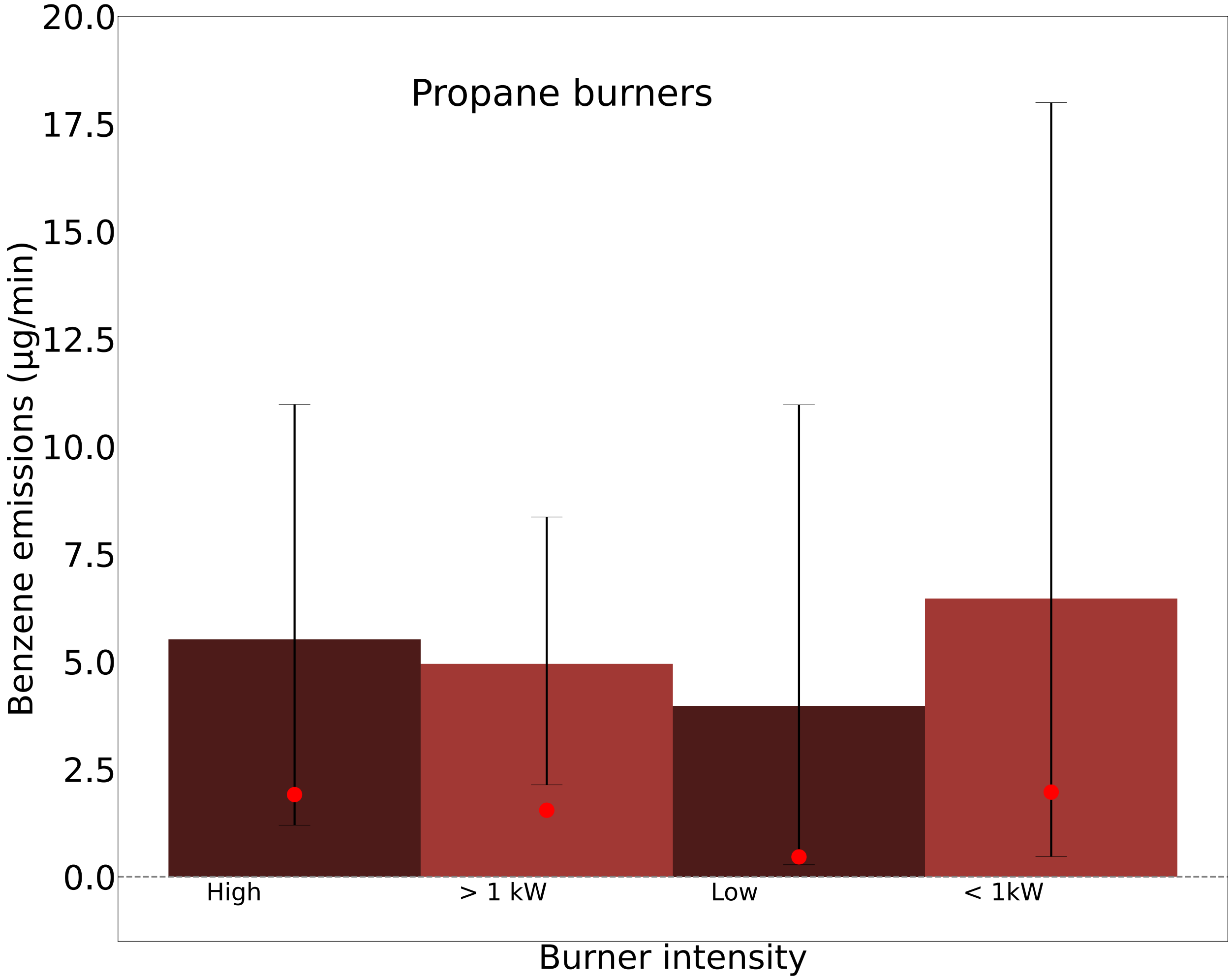

Figure S12. Mean and median benzene emissions in µg C_6H_6 min^{-1} from propane stoves on high and low and by a power threshold. The red points inside the bars represent median values. Black error bars represent the 95% confidence interval of the mean (calculated as described in Methods). Benzene emission rates were measured directly using the AROMA analyzer (see Methods). Burners on "High" refers 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; "> 1kW" and "< 1kW" refer to burners whose power output is greater than and less than 1kW, respectively. Power level was calculated based on CO_2 emissions (see Methods).

References

1. Lebel, E. D., Finnegan, C. J., Ouyang, Z. & Jackson, R. B. Methane and NOx Emissions from Natural Gas Stoves, Cooktops, and Ovens in Residential Homes. *Environ Sci Technol* **56**, 2529–2539 (2022).
2. Singer, B. C., Chan, W. R., Kim, Y. S., Offermann, F. J. & Walker, I. S. Indoor air quality in California homes with code-required mechanical ventilation. *Indoor Air* **30**, 885–899 (2020).
3. California OEHHA. TSD for Noncancer RELs Appendix D. Individual Acute, 8-Hour, and Chronic Reference Exposure Level Summaries. 139–216 (2014).