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SUMMARY
We developed a physics-based data-supported model to investigate indoor pollutant exposure distributions resulting from use of natural gas cooking appliances across households in California. The model was applied to calculate time-resolved indoor concentrations of CO, NO_2 and formaldehyde resulting from cooking burners and entry with outdoor air. Exposure metrics include 1-week average concentrations and frequency of exceeding ambient air quality standards. We present model results for Southern California (SoCal) using two air-exchange scenarios in winter: (1) infiltration-only, and (2) air exchange rate (AER) sampled from lognormal distributions derived from measurements. In roughly 40% of homes in the SoCal cohort (N=6634) the 1-hour USEPA NO_2 standard (190 $\mu g/m^3$) was exceeded at least once. The frequency of exceeding this standard was largely independent of AER assumption, and related primarily to building volume, emission rate and amount of burner use. As expected, AER had a more substantial impact on one-week average concentrations.

IMPLICATIONS
Our results suggest that using natural gas cooking burners without venting (kitchen exhaust systems) commonly leads to residential NO_2 concentrations that exceed ambient air quality standards. While these model-based results should be checked with measurements, the scope and severity of potential health impacts warrant priority attention by public health agencies.

KEYWORDS
gas appliances, indoor air modelling, air exchange rate, nitrogen dioxide, carbon monoxide

INTRODUCTION
Natural gas burners can emit carbon monoxide (CO), nitrogen dioxide (NO_2), formaldehyde, ultrafine particles and other pollutants. When used without venting – common for cooking burners –pollutants can reach unhealthful levels. Large field studies in the 1980s and 1990s documented strong associations between pollutant levels and gas cooking burner use, and several epidemiological studies established links to respiratory health impacts (see for example, Jarvis et al, 1998 and Wong et al., 2004). The distribution of exposures to cooking related pollutants across the current population is unknown and not readily estimated from older studies owing to changes in appliances, cooking patterns and residential ventilation rates. To facilitate investigation of these exposures, we developed a physics-based modeling framework that utilizes large existing databases of housing, household and other data for California. The overall framework of our exposure modeling approach is depicted in **Figure 1**.

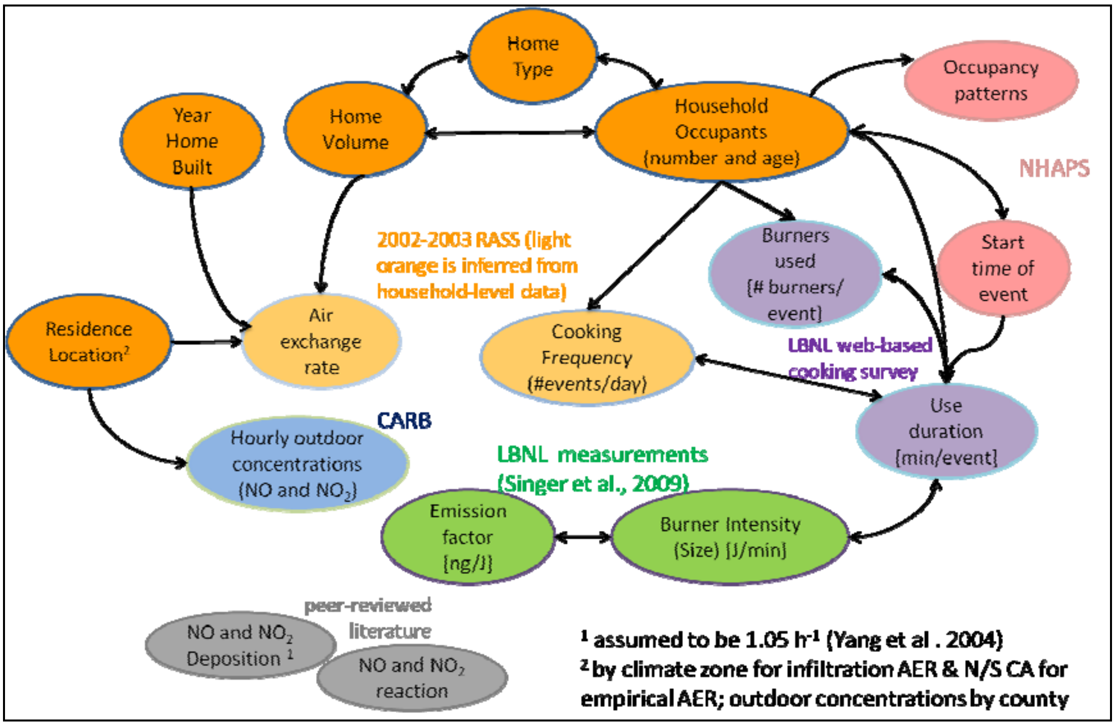

Figure 1. Exposure model influence diagram linking key parameters that determine indoor pollutant concentrations and exposures, resulting from use of gas cooking appliances.

METHODS

We calculate time-dependent pollutant concentrations in each homes using a single-zone mass balance model that accounts for pollutant emissions from cooking burner use, dilution based on home volume, entry of pollutants from time-resolved outdoor concentrations and removal by deposition and outdoor air exchange, as described in Equation 1 below:

$$V\frac{dC_i}{dt} = E_i - k_i V C_i - Q(C_i + p_i C_{i,out}) \qquad (1)$$

In this equation, written for pollutant species *i*, V is volume of the residence (m^3), C_i is the indoor concentration ($\mu g\ m^{-3}$), E_i is the emission rate ($\mu g\ h^{-1}$), $C_{i,out}$ is the outdoor concentration ($\mu g\ m^{-3}$), Q is the air flow indoors and is equal to the flow outdoors ($m^3\ h^{-1}$), p_i is penetration efficiency (unitless) for pollutants coming indoors from outdoors, and k_i is the indoor pollutant deposition rate (h^{-1}). E_i is estimated based on the emission factors (ng/J) measured from a recent lab study at LBNL (Singer et al., 2009), and assuming an average cooktop burner firing rate of 123 kJ/min (7 kBtu/h).

Eq 1 is solved recursively for C_i, so that any of its parameters can be varied across time (i.e., they are constant within a given time step but can change from one time step to another). In addition, the equation can be used to separately track pollutant mass originating from indoor emissions and from outdoor sources, with total concentrations calculated as the sum of the contributions from the two sources. This superposition approach was used by Klepeis (1999) to combine discrete source emissions in a residence.

The total indoor concentration at time (t) is calculated as the sum of contributions from indoor and outdoor sources. Outdoor concentrations of NO_2 and CO were obtained from the

US EPA’s State and Local Air Monitoring Network. All California households in a county were assigned the same hourly-averaged outdoor air pollutant concentration profile.

The recursive model is implemented and solved in R-programming code. Output includes indoor concentrations of NO_2 and CO over a course of a week, at 1-min resolution. The modeled concentrations [Eq 1] are linked with age-associated archetypal time-activity patterns for weekdays and weekends to assess individual exposures. These patterns are derived from analysis of the National Human Activity Patterns Study (NHAPS) (Klepeis et al., 2001). Key features of our exposure model - including linking household (HH) level data with cooking activity and building characteristics, characterizing air exchange, and near-source (proximity) effects - are described below.

Linking Household level data with cooking activity and building characteristics

As shown in **Figure 1**, core data for the model is drawn from the publicly available 2002-2003 Residential Appliance Saturation Survey (RASS), containing anonymous data for over 10,000 California households (HH) that specifically report cooking with gas appliances. The RASS database is statistically representative of the population of California households

The exposure model links residential-level data from the RASS on frequency of cooking, building type, home size, number and ages of occupants; and supplemental cooking activity data from a web-based cooking survey conducted by LBNL. The web-based survey provides data on the duration of meal-specific (Breakfast, Lunch, and Dinner) oven and cooktop use, and number of cooktop burners used, related to household characteristics. In the model, meal occurrences are assigned based on RASS responses related to cooking frequency; specific activity factors (use of oven, number of cooktop burners, cooking duration) are assigned from distributions derived from the survey.

Air Exchange Rate (AER)

In the exposure model, air-exchange can be modeled assuming infiltration only, or sampled from lognormal distributions of winter or non-winter empirical AERs, the latter including window use and mechanical ventilation. To estimate annual-average AER from infiltration (assuming no open windows), we apply the following equation from the American Society of Heating, Refrigerating and Air-conditioning Engineers Standard (ASHRAE) 136-1993:

$$\mathrm{AER} = NL \times W \qquad (2)$$

where NL is normalized leakage [-], and W is a factor that takes into account local weather effects (ranging from 0.57 to 0.92 across California). We use results of Chan et al (2005) to assign NL based on building age and floor area for each household in the RASS cohort.

To characterize seasonal variations in AER, we analyzed raw data of AER measurements from winter and non-winter seasons across California (Wilson et al., 1993, 2003; Offermann 2009; AER, 2010). We found that empirical AERs are lognormally distributed across California with distinct distributions for northern and southern regions. The AERs varied by construction year; older homes having higher AERs, presumably due to higher infiltration rates associated with less airtight building shells. For the southern California region, we obtained a sufficient quantity of data to develop distinct lognormal AER distributions by season (winter, non-winter), and by year of construction (pre-1980, 1980-1995, post-1995).

Near-source (proximity) effects

Reflecting observed trends from exposure monitoring studies, the exposure model recognizes that individuals close to the activity- cooking with a gas appliance- are exposed to higher pollutant concentrations than the home average. This proximity effect is applied to the household member assigned as the "cooker", and to young child(ren) (0-5 years) assumed to be near the cooker. To calculate exposure concentrations for these individuals, we apply a proximity factor (F_{prox}) to the time-dependent concentration of indoor-origin pollutants, i.e.,

$$C_{i.\exp}(t) = F_{prox} C_{i.indoor-origin}(t) + C_{i.outdoor-origin}(t) \tag{3}$$

Based on a literature review on the proximity effect as it relates to cooking exposures, we assume F_{prox} of 2.0 for the adult or senior cooker. This value incorporates both the near source and room level increments. Only one adult or senior in the household is assumed to be the cook, and a senior is assumed, if present. For young children, we assume an F_{prox} of 1.5.

RESULTS

To explore the influence of air exchange on modeled residential exposure concentrations, we present results from two winter scenarios applied to Southern California (SoCal) RASS households[1]: (1) infiltration-only AER, and (2) AER sampled from a lognormal distribution of empirical AER. Across all SoCal household (HHs), empirical AER ranged from 0.08 to 2.48 h^{-1} with GM (GSD) of 0.40 (1.6). Infiltration AER ranged from 0.05 to 0.42 h^{-1}, with GM (GSD) of 0.18 (1.3).

Tables 1 and 2 present the GM (GSD) of modeled 1-week average and maximum (max) 1-hr NO_2 and CO concentrations assuming only indoor sources (C_{in}), and from the indoor and outdoor contributions (C_{in+out}). The tables also show the number of households (HHs) that exceed an acute health-based standard (1-h for NO_2; 1-h or 8-h for CO).

Table 1. NO_2 results: GM (GSD) [$\mu g/m^3$] of 1-wk average and max 1-hr C_{in} and C_{in+out}; percent of homes with concentrations exceeding USEPA's 1-h NO_2 ambient air quality standard (190 $\mu g/m^3$).

	1-wk		max 1-hr		% of HHs with C_{in} exceeding 1-h NAAQS[a]
	C_{in+out}	C_{in}	C_{in+out}	C_{in}	
Infiltration only	15.5 (2.0)	7.3 (2.6)	172 (2.3)	161 (2.3)	44%
Empirical AER	20.2 (1.7)	6.0 (2.7)	166 (2.3	145 (2.4)	40%

[a] C_{in+out} exceeded the 1-h standard in 136 and 269 (infiltration, empirical AER) additional homes (of 6634)

Table 2. CO results: GM (GSD) [ppm] of 1-wk average and max 1-hr C_{in} and C_{in+out}; percent of homes with concentrations exceeding the USEPA's 1-h and 8-h standard (35 and 9 ppm, respectively).

	1-wk		max 1-hr		% of HHs exceeding	
	C_{in+out}	C_{in}	C_{in+out}	C_{in}	1-h NAAQS (C_{in})[a]	8-h NAAQS (C_{in+out})
Infiltration only	1.4 (2.4)	0.4 (3.4)	3.9 (2.9)	2.4 (3.2)	2.1%	10.7%
Empirical AER	1.1 (2.5)	0.2 (4.3)	3.5 (3.0)	2.0 (3.4)	1.4%	5.4%

[a] Including outdoor sources C_{in+out} exceeded 1-h standard in 8 and 3 more homes (infiltration, empirical AER)

DISCUSSION

The average 1-wk C_{in+out} summarized in **Tables 1 and 2** display the key role that outdoor air concentrations have in explaining indoor concentration levels when increased air exchange is assumed. Assuming empirical AERs, the average 1-wk C_{in+out} for NO_2 and CO are roughly a

[1] Includes 6634 HHs from six Southern California counties: Los Angeles, San Bernardino, Riverside, San Diego, Orange, and Ventura County

factor of 3 and 5 greater, respectively, than C_{in}. When infiltration is considered, the average 1-wk C_{in+out} is only a factor of roughly 2 and 3 greater than C_{in} for NO_2 and CO, respectively.

We find that slightly more than 5% of the SoCal HHs in our gas-appliance cooking cohort exceed the 8-hr CO standard (9 ppm) at least once, over the course of the week, assuming empirical AER. Roughly double as many HHs (10.7%) exceed this standard at least once assuming infiltration-only AER. In comparison, Wilson et al (1993) found that among 161 homes located in the SoCal region (i.e., within the SoCal Gas and SDG&E utility districts), roughly 7% of the residences had measured winter indoor CO concentrations that exceeded the 8-hr CO standard (9 ppm). However, the measurements of Wilson et al. (1993) may include other indoor CO sources, such as cigarette smoking and presence of a gas pilot.

We acknowledge that the max 1-hour C_{in} for NO_2 seem quite high, resulting in at least one exceedance of the 1-h NAAQS in about 40% of SoCal HHs that reported at least some cooking with their gas appliances when responding to the RASS (**Table 1**). The number of exceedances is relatively insensitive to the air exchange scenario. Max 1-h concentrations are primarily related to emission factors, cooking activity (duration and number of burners), and home volume. Because RASS HHs are considered representative of all California HHs, and hence inferred to be representative of specific regions, roughly 1.3 million gas fueled SoCal HHs that use cooking appliances (cooktop and/or oven) may have at least one exceedance each week of the USEPA's 1-hr NO_2 standard. This assumes that roughly 50% of 6.3 million SoCal HHs (EIA, 2001 and Census 2000 data) have gas appliances, and that 92% of them use gas cooking appliances (based on RASS). These results highlight the need for monitoring efforts to better characterize indoor 1-hr NO_2 levels arising from the use of natural gas cooking appliances.

Owing to the lack of measured 1-hr average C_{in} with which to compare our modeled results, we estimate a simple steady-state solution of 1-hr average NO_2 C_{in} for each RASS household, assuming winter empirical AER; maximum emission rate (2 cooktop burners and oven use) associated with dinner cooking[2]; building volume reported in RASS (GM=311 m^3 GSD = 1.6); and an NO_2 deposition rate of 0.95 hr^{-1}. The distribution of estimated steady-state "max"1-hr C_{in} of NO_2 (GM= 129 μg/m3; GSD=2.3) closely agree with our modeled results, indicating that the number of HHs exceeding the 1-hr NO_2 standard (**Table 1),** is reasonable.

Although the web-based cooking survey may not be entirely representative of the cooking activity of SoCal HHs, it is based on a sufficiently large sample of HH to be considered reasonable. Further, because we associate cooking activity patterns, such as cooktop burner use and oven-use duration, with specific meals and household characteristics, such as number of occupants and presence/absence of child(ren), we are able to link key exposure factors that have not been considered in previous gas cooking-appliance exposure modeling efforts.

The results presented in **Tables 1 and 2** highlight the primary intent of the model, which is to characterize household-level residential concentrations from gas cooking appliances. As we discuss above, the model allows us to explore the variation in household-level impacts depending on the particular input scenario. In a related presentation (***Abstract 188***), we

[2] The GM (GSD) of inputs used to estimate a maximum emission rate include cooktop and oven NO_2 emission rates of 2.1 (1.4) mg/min, and 2.0 (1.3) mg/min, respectively, and 28 (1.8) min cooktop duration, and an oven-burner time of 14 (1.6) minutes. Note that oven burner time reflects the oven-specific on/off cycling of the oven burner, and is translated from reported oven duration in the web-based cooking survey.

present NO_2 and CO impacts and policy implications of gas cooking appliance emissions resulting from a transition from baseline NG to liquefied natural gas in San Diego county.

Lastly, while individual exposure concentrations are not presented, as part of the continued diagnostic evaluations, we are assessing the sensitivity of the modeled individual exposure concentrations to F_{prox} (which may be underestimated currently).

CONCLUSIONS

We demonstrate how the exposure model can assess average residential concentrations, and household-level impacts of pollutants emitted from gas cooking appliances. Foundational to our modeling approach is the ability to link key data inputs on residential-level and environmental characteristics, thereby reducing the uncertainty in our model inputs. Our results demonstrate a critical need for more information and monitoring efforts to assess acute (1-hr average) NO_2 levels in homes that use gas cooking appliances.

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