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Indoor particulate matter during HOMEChem: Concentrations, size distributions, and exposures

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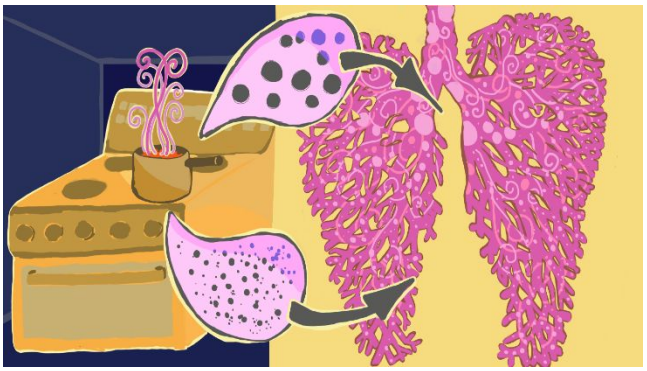
Abstract

It is important to improve our understanding of exposure to particulate matter (PM) in residences because of associated health risks. The HOMEChem campaign was conducted to investigate indoor chemistry in a manufactured test house during prescribed everyday activities, such as cooking, cleaning, and opening doors and windows. This paper focuses on measured size distributions of PM (0.001-20 μm), along with estimated exposures and respiratory-tract deposition. Number concentrations were highest for sub-10 nm particles during cooking using a propane-fuelled stovetop. During some cooking activities, calculated $PM_{2.5}$ mass concentrations (assuming a density of 1 g cm^{-3}) exceeded 250 μg m^{-3} and exposure during the post-cooking decay phase exceeded that of the cooking period itself. The modeled PM respiratory deposition for an adult residing in the test house kitchen for 12 hours varied from 7 μg on a day with no indoor activities, to 68 μg during a simulated day (including breakfast, lunch, and dinner preparation interspersed by cleaning activities), and rose to 149 μg and during a simulated Thanksgiving day.

Abstract Art



1. Introduction

Outdoor air quality in much of the developed world has improved substantially over the past half century, corresponding to expected improvements in pollution-associated health risks.[1,2] On the other hand, indoor air quality (IAQ) is neither well-regulated nor well understood. This knowledge gap is critical because surveys indicate that people spend ~90% of their time indoors[3] and most of that time is spent in one's residence. Pollutants of outdoor origin are an important determinant of IAQ.[4,5] However, with decreasing ambient pollutant levels, the role of pollutants of indoor origin is becoming a relatively more important influence for personal exposure. Beyond intrusion of pollutants from outdoors, indoor emission sources can degrade IAQ. Activities that contribute to indoor air pollution include cooking,[6,7] smoking,[8–10] and cleaning,[11,12] in addition to emissions from indoor constituents like building materials,[13,14] personal care products,[15] consumer electronics,[16] and even human occupants themselves.[17] The physical and chemical phenomena that control pollutant transport and transformations outdoors differ from those which are important indoors, due to factors such as shorter air residence times indoors, altered abundances of oxidative species, and higher surface-to-volume ratios in indoor environments compared to urban and regional atmospheres.[18]

Particulate matter (PM) is a key pollutant from health and environmental perspectives both indoors and outdoors. Exposure to PM is associated with various adverse health outcomes, such as cardiovascular disease, chronic obstructive pulmonary disease, and asthma.[19–21] It is one of the leading global causes of mortality and ill-health.[22] A study quantifying the relationship between global mortality and ambient $PM_{2.5}$ estimated that ~2 million premature deaths can be avoided by reducing $PM_{2.5}$ to 10 μg m^{-3} globally.[23]

Bekö et al. measured ultrafine particle exposure of sixty nonsmoking residents of Copenhagen and found that their homes and other built environments accounted, respectively, for 50% and ~40% of their daily personal exposure.[24] A recently developed framework attributed 42 ± 24% and 28 ± 26% of the total exposure (including different microenvironments and outdoors) to $PM_{2.5}$ of outdoor and indoor origin in residences, respectively.[25] Among all indoor microenvironments considered, residences contributed the most to exposure and associated mortality burden. The considerable uncertainty associated with these estimates highlights our limited understanding of PM in residences.

Many studies have reported on the characteristics of PM in residences. Sources considered in such studies include cooking,[26] other combustion sources such as candles and incense,[27,28] activities involving hot surfaces such as irons and hair dryers,[29,30] and penetration from outdoors.[4] Studies with multiple activities and sources in a house-like controlled environment are sparse.

The House Observations of Microbial and Environmental Chemistry (HOMEChem) campaign was conducted in a test house in June 2018 to investigate how everyday activities impact the emissions, chemical transformations, and removal of trace gases and particles in a residential environment.[31] Performing prescribed activities such as cooking and cleaning in the test house enabled the simulation of residential conditions that were more controlled than observational field campaigns but less controlled than chamber studies. This work focuses on insights from PM size distributions (0.001-20 μm) measured using a range of particle sizing instruments. Overall size-segregated PM number and calculated mass concentrations and their variation throughout different indoor activities are discussed. Sources of PM in different size modes and estimated exposure and lung deposition are also reported.

2. Materials and Methods

2.1 Test House

The HOMEChem study was conducted in a three-bedroom, two-bathroom manufactured test house (111 m^2 floor area and ~250 m^3 volume), located at the University of Texas at Austin. The house layout and instrument locations are presented in Fig. S1 of the supporting information (SI). The test house has been described in detail elsewhere;[31] main characteristics are summarized here. The outdoor air supply system was kept continuously on to maintain a positive pressure while providing an air exchange rate of 0.5 ± 0.1 h^{-1}. The air conditioning system turned on and off intermittently to maintain the target temperature (~25 °C, under thermostatic control). The fan in the air handling unit operated continuously, without a filter, at a flow rate of 2000 m^3 h^{-1} to maintain a high rate of internal mixing (equivalent to 8 house volumes h^{-1}). All external doors and windows were kept shut, except when specific experiments required otherwise. All internal doors, except those of the two bathrooms, were kept open throughout. More details about the test house location and its ventilation system are available in the SI.

2.2 Experimental design

The HOMEChem experimental design is comprehensively detailed in Farmer et al.[31] Two categories of experiments were performed: (1) Sequential, in which the same type of activity was performed repeatedly throughout the day at regular intervals, sometimes interspersed by a venting period, with opened doors and windows; (2) Layered, in which different types of activities were performed throughout the day—without opening the house—to allow emissions to interact. Sequential experiments focused on quantifying the impacts of specific activities; layered experiments simulated a "day in the life" of a residential environment.

Three types of sequential experiments, Sequential Stir-Fry, Sequential Cleaning, and Sequential Ventilation, were performed. In Sequential Stir-Fry experiments, a meal consisting of a vegetable stir-fry and white rice was cooked. This process was repeated four times (three on the propane stove and one on an electric hot plate) on each of four days. Similarly, during each day devoted to Sequential Cleaning, the test house was mopped multiple times using a variety of cleaners. The Sequential Ventilation experiment consisted of repeatedly opening and closing all external doors and windows throughout the day to assess its impact on the time-dependent relationship between indoor and outdoor PM levels.

Two types of Layered experiments were conducted: Layered Day and Thanksgiving Day. In the four Layered Day experiments, volunteers entered the house in the morning, cooked breakfast (pan-fried sausage, fried eggs, fried tomato, toast, and coffee), mopped the floors with a pine-scented cleaner, prepared lunch (same as in Sequential Stir-Fry), prepared coffee and toast, cooked dinner (either lasagna or chili), mopped the house with bleach, started the dishwasher, and left the house. During the two Thanksgiving Day experiments, four volunteers entered the house in the morning, prepared breakfast (same as in Layered Days), then prepared a typical Thanksgiving meal, including oven-roasted turkey, bread stuffing/dressing, brussels sprouts, and sweet potato casserole, in addition to pies, cranberry sauce, and gravy. Then, 12-15 guests entered the house, dined, performed cleaning activities, started the dishwasher, and exited the house. Example activity schedules followed during these experiments are presented in Tables S1-S5.

Two more experiments (naked stove and naked hot plate) were performed to characterize PM generated during the operation of the stove and hot plate, without a pan or pot, at the highest power level for ~20 minutes. These two experiments were not part of either Layered Day or Sequential Day experiments. Test house PM levels were at background levels before the start of experiments.

The Institutional Review Board's (IRB) human research review approval requirement was waived for the HOMEChem study because no identifying information was collected from house occupants.

2.3 Instrumentation, sampling scheme, and data analysis

An A11 Nano Condensation Nucleus Counter system (A11-nCNC, Airmodus Oy, Helsinki, Finland) was deployed to sample particles in the 1-4 nm activation size range.[32–34] Two Scanning Mobility Particle Sizers (SMPS, TSI Inc., Shoreview, MN) measured number distributions in the 4-532 nm electrical mobility size range. Aerodynamic size distributions in the diameter range 0.5-20 μm were measured using Aerodynamic Particle Sizers (APS, TSI 3321). Optical measurements made by an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS, Droplet Measurement Technology, Longmont, CO) were used to compensate for data loss in the 105-533 nm size range from one of the SMPS units on certain days. The A11-nCNC and both SMPSs were set to capture one size distribution every five minutes, whereas the APS and UHSAS recorded a size distribution every minute and every second, respectively. To minimize particle losses and to ensure that the smaller particles were captured, all particle sizers except the UHSAS were placed inside the test house (Figs. S1b and S1c). More details about the instruments are available in the SI.

Depending on PM composition, airborne particle density typically varies within the range 0.8-2.5 g cm^{-3}.[35–38] For this study, mass concentrations were calculated from the number size distribution data assuming spherical particles with a density of 1 g cm^{-3} similar to previous studies on indoor PM.[39,40] The SI presents a discussion on time-resolved PM_1 density estimated using composition data obtained from an aerosol mass spectrometer, density assumptions, and associated uncertainties (Figs. S2 and S3, Tables S6 and S7). Briefly, using a constant density of 1 g cm^{-3}

underestimated PM_1 mass concentrations during cooking and post-cooking decay phase by less than 25% compared to PM_1 mass concentrations calculated using composition-dependent density.

For quantitative analyses, calculated PM mass concentrations were segregated into six size groupings: $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$, PM_{10}, and $PM_{20,}$ whereas the number concentrations were divided into two size bins: 4-100 nm (commonly referred to as ultrafine particles, or UFP) and 0.1-20 μm. For Layered Days and Thanksgiving, average concentrations over 12 hours (8 AM - 8 PM local time) are reported. The average mass and number concentrations corresponding to different meals were calculated over the period between the moments in which the heat source is first turned on and finally turned off.

Integrated $PM_{2.5}$ exposures for an occupant residing continuously in the test house were calculated over 12 hours for Layered Days and for Thanksgiving. For each cooking activity, exposures during the cooking and post-cooking decay period, are reported. PM mass deposited in the respiratory tract (0.004-20 μm) was estimated using a lung deposition model (Eq. S1-S4) of the International Commission on Radiological Protection[41,42] for a particle density of 1 g cm^{-3}, utilizing an age-weighted average volumetric inhalation rate for 20-60 year old adults of 11 L min^{-1}.[43] Because measurements were performed at a single location near the kitchen, the reported exposures and respiratory deposition estimates are specific to that location.

3. Results and Discussion

3.1 Overview of particulate matter concentrations and trends

Fig. 1 presents the temporal variation of size-segregated PM mass concentrations during four experimental days. Data corresponding to the other experimental replicates are presented in Fig. S4-S6. Cooking activities were the single largest source of indoor PM on a mass basis per activity.

During cooking, $PM_{2.5}$ concentrations as high as 250 μg m^{-3} were recorded (Fig. 1a). The highest PM concentrations observed were similar to those in the world's most polluted cities;[44,45] however, peak levels were short-lived. Average PM levels and geometric mean diameters (both number- and volume-based) are shown in Table 1.

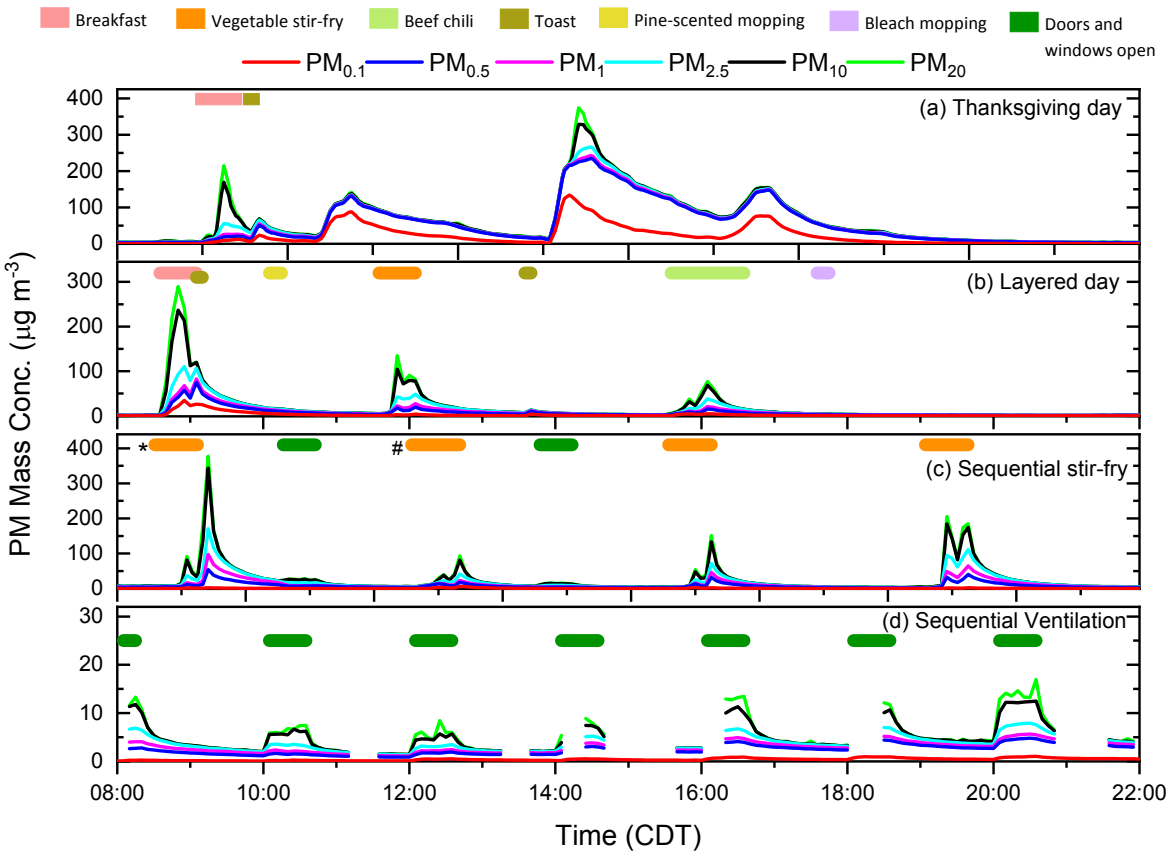


Figure 1. Temporal variation of size segregated mass concentrations of particulate matter ($PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$, PM_{10}, and PM_{20}) during (a) Thanksgiving Day (TG-2), (b) Layered Day (LD-2), (c) Sequential Stir-fry experiment (SF-2), and (d) Sequential Ventilation experiments. Relevant activities performed during the experiments are marked for all experiment types except the Thanksgiving Day experiment. A detailed activity log for the Thanksgiving Day experiment is presented in Fig. S11. The symbol * indicates vegetable stir-fry cooked on the hotplate; the symbol # denotes vegetable stir-fry cooked in a cast-iron pan. The gaps in the $PM_{0.5}$, PM_1, $PM_{2.5}$, PM_{10}, and PM_{20} time-profiles for the Sequential Ventilation experiment correspond to periods when the UHSAS instrument was sampling outdoor air.

In terms of number concentration, UFP dominated indoor PM number concentration during cooking (Table 1 and Fig. S7-S9) and throughout the Sequential Ventilation experiment (Fig. S10).

Indoor $PM_{2.5}$ levels were comparable to prior literature reports, although we note that observed concentrations are the product of not only emission rates but also ventilation conditions and house volume. For example, Wan et al. [46] reported an average $PM_{2.5}$ concentration of ~160 μg m^{-3} for 30 cooking episodes in the kitchens of 12 Hong Kong homes. Long et al. [47] reported an average $PM_{2.5}$ concentration of 37 ± 31 μg m^{-3} for stir-frying compared to 30 ± 10 μg m^{-3} in this study (Table 1).

Comparable PM levels (Table 1) and trends (Fig. 1a and S5) were observed during both Thanksgiving days. These represented the highest UFP levels during HOMEChem, in terms of both mass (as high as 100 μg m^{-3}) and number (as high as 2.7×10^6 cm^{-3}, Fig S8). During this experiment, the $PM_{2.5}$ level remained above 50 μg m^{-3} for more than five hours. Multiple cooking activities, using both the oven and stove, were performed in parallel throughout the day (Fig. S5 and S11). These PM levels reflect the cumulative influence of emissions from different sources, but most prominently from cooking.

Average $PM_{2.5}$ mass concentrations (over 12-h periods) were ~60 μg m^{-3} (Table 1) for both Thanksgiving days, about four times higher than during Layered Days (~15 μg m^{-3}). While both types of day-long experiments showed a preponderance of UFP on a number basis (97% for Thanksgiving and 99% for Layered Days), the mass fraction of UFP among total PM was nearly three times higher for Thanksgiving (34%) compared to Layered Days (11%). A larger geometric mean particle size was observed for the Thanksgiving (~18 nm) compared to Layered Days (10 ± 2 nm, average ± standard deviation) (Fig. S12a). In terms of particle volume (Fig. S12b), the geometric mean size for Thanksgiving (~180 nm) was much smaller than that for Layered Days (800 ± 200 nm) consistent with the observation that $PM_{0.5}$ constituted a higher fraction of total PM mass during Thanksgiving (86%) compared to the Layered Days (50 ± 10%).

229

230 **Table 1.** Average size-segregated PM number[#] (<100 nm and >100 nm) and mass ($PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$, PM_{10}, and PM_{20}) concentrations

		Number Conc. (cm^{-3}) (average ± SD)		Mass Conc. ($\mu g\ m^{-3}$) (average ± SD)						Geometric mean diameter (nm) (average ± SD)	
	Cooking duration (min)	$N_{<100\ nm}$	$N_{>100\ nm}$	$PM_{0.1}$	$PM_{0.5}$	PM_1	$PM_{2.5}$	PM_{10}	PM_{20}	By number	By volume
12-hour averages											
Layered Day	NA	$(2.2 \pm 0.9) \times 10^5$	$(2 \pm 1) \times 10^3$	2.0 ± 0.5	9 ± 2	11 ± 2	14 ± 2	18 ± 2	19 ± 2	10 ± 2	$(8 \pm 2) \times 10^2$
Thanksgiving Day	NA	7.7×10^5	2.4×10^4	23.4	59.0	61.1	63.1	66.3	67.7	18	1.7×10^2
		8.0×10^5	2.3×10^4	22.0	56.4	57.9	60.9	64.8	66.5	17	1.9×10^2
Meal-wise (averaged over the cooking duration)											
Breakfast	34 ± 10	$(1.2 \pm 0.5) \times 10^6$	$(10 \pm 8) \times 10^3$	11 ± 6	30 ± 20	35 ± 20	48 ± 30	80 ± 50	100 ± 50	11 ± 5	$(1.8 \pm 0.7) \times 10^3$
Stir-fry (gas stove)	37 ± 10	$(9 \pm 5) \times 10^5$	$(3 \pm 1) \times 10^3$	3 ± 1	13 ± 5	17 ± 7	30 ± 10	44 ± 20	50 ± 20	8 ± 3	$(1.4 \pm 0.7) \times 10^3$
Stir-fry (hot plate)	52 ± 6	$(1.5 \pm 0.6) \times 10^5$	$(1.9 \pm 0.4) \times 10^3$	1.8 ± 0.4	10 ± 3	15 ± 7	25 ± 10	40 ± 30	46.3 ± 30	16 ± 5	$(1.9 \pm 0.5) \times 10^3$
Chili	68 ± 3	$(8 \pm 2) \times 10^5$	$(2 \pm 1) \times 10^3$	3 ± 1	11 ± 6	15 ± 8	20 ± 10	30 ± 20	40 ± 20	7 ± 1	$(1.2 \pm 0.1) \times 10^3$
Lasagna	75	2.7×10^5	3.3×10^3	3.0	11.5	12.0	12.9	13.3	13.4	13	200
Toast	*	$(10 \pm 2) \times 10^4$	$(8 \pm 7) \times 10^3$	2.1 ± 0.4	9 ± 2	11 ± 2	12 ± 2	12 ± 2	12 ± 2	16 ± 3	$(1.9 \pm 0.6) \times 10^2$
Sequential Ventilation experiment											
All external doors and windows open	^	$(1.5 \pm 0.2) \times 10^4$	$(9 \pm 3) \times 10^2$	0.5 ± 0.3	3 ± 1	4 ± 1	6 ± 1	9 ± 2	10 ± 3	21 ± 8	$(1.4 \pm 0.3) \times 10^3$
Unoccupied background	NA	$(2 \pm 1) \times 10^3$	$(2.3 \pm 0.4) \times 10^2$	0.13 ± 0.04	1.2 ± 0.3	1.5 ± 0.3	2.3 ± 0.3	2.5 ± 0.4	2.5 ± 0.4	35 ± 10	$(6.1 \pm 0.9) \times 10^2$

#Size-segregated number concentrations of sub-100 number particles are presented in Table S8.
*Since toast preparation took only 2-3 minutes, the scan including the toast preparation and the following scan has been used to calculate average values.
^External doors and windows were opened for 28 ± 6 minutes

Comparing average PM levels over the cooking duration by individual meal type, the highest PM mass concentrations—at all PM size ranges—were observed during breakfast, which might be attributed to the cumulative effect of cooking multiple items (sausage, tomato, and egg) in an open pan with oil. Average $PM_{2.5}$ levels for breakfast were about twice those for stir-fry and chili and nearly 4× those for lasagna and toast. However, PM levels for many stir-fry events were comparable or exceeded that for some breakfasts. Lasagna and toast exhibited the highest $PM_{2.5}$ mass fraction (~97% and ~98%, respectively), compared to <65% for other meals. Stir-fry, breakfast, and chili preparation required ingredients to be cooked in hot oil and stirred during the process, which generates coarse particles[47] and, therefore, lowers $PM_{2.5}/PM_{20}$ ratios. The formation of UFP observed during cooking is discussed in §3.2.

Temporal PM mass concentration (Fig. 1d) and size distribution (Fig. S10) during the Sequential Ventilation experiment showed that indoor PM levels peaked within five minutes after opening the house and remained nearly constant at concentrations of 6-17 μg m^{-3}, only ~6% lower than outdoor levels (Fig. S13). As compared with cooking events, UFP constituted smaller fractions of the total PM mass (5 ± 2%) and number (94 ± 2%). After doors and windows were closed, it took 44 ± 5 minutes for PM concentrations to return to stable background levels.

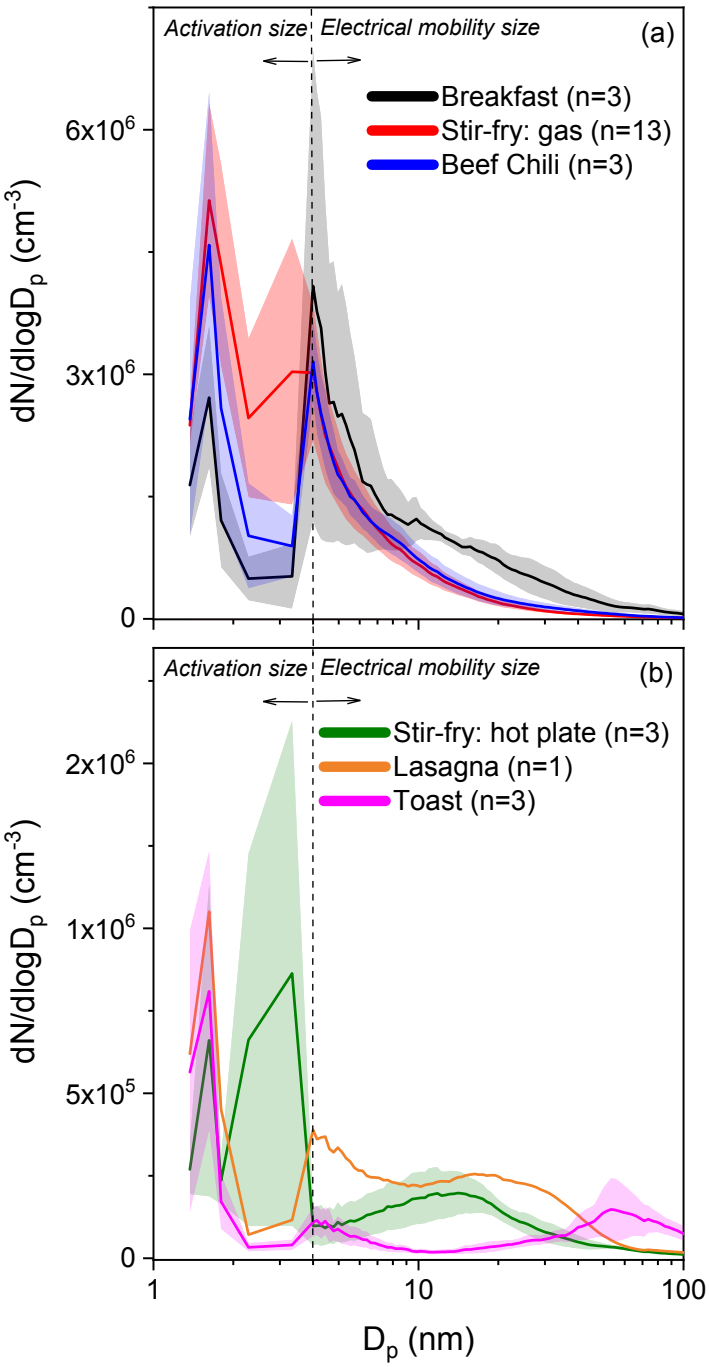
Mopping did not produce distinct changes in mass concentrations of PM_1 and smaller mass fractions during Layered Days (Fig 1b and S4) and Sequential Cleaning experiments (Fig. S14 and S15). Observed increases in concentrations of larger size fractions during mopping (Fig. S14) are probably attributable to particle release through human movements, such as dust resuspension, shedding of skin cells, and emission of clothing fibers.[48,49] The formation and growth of sub-30 nm particles observed after some mopping events are discussed in §3.3.

3.2 Particle formation during cooking

For all meals, UFP dominated number concentrations with number geometric mean diameters <20 nm, while the volume geometric mean diameter varied from ~200 nm for toast and lasagna to ~2 μm for stir-fry (Table 1). Multi-modal size distributions were observed for all meals (Fig. 2) with 1-2 modes in size range measured by diethylene glycol (DEG) activation (<4 nm). Peaks at 1.6 nm may be the result of clusters of gaseous species activating and growing with DEG as observed in a study of atmospheric aerosol nucleation.[50] A study of traffic-related emissions also reported multi-modal size distributions in the DEG activation size range.[51]

In terms of PM > 4 nm, the number mode from toast emissions was 50 ± 10 nm, which is comparable to the 30-50 nm range reported for toasters in previous studies.[52–54] High concentrations of sub-10 nm particles were observed for meals cooked on the gas stove (breakfast, stir-fry, and chili, Fig. 2a) with number modes in the range 4-11 nm. Even though the oven was also propane-fuelled, sub-10 nm particle concentrations were 5-7× lower for the oven-baked lasagna (Fig. 2b) compared to the meals cooked on the stove. PM emissions taking place in the oven might be subjected to enhanced condensational growth, coagulation, and wall losses, especially for sub-10 nm particles.

During cooking, particles can originate from both the heat source and the food, leading to some distinguishing source-specific characteristics. The combustion of gaseous fuels for cooking is known to generate high numbers of sub-10 nm particles[6] and electric appliances, such as a toaster, a sandwich maker, and a hot plate (with no food) have also been shown to release large numbers of <10-nm particles.[30,55]



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277 **Figure 2.** Ultrafine particle number size distributions, averaged over the cooking duration, for different
278 types of meals, plotted in two panels with different *y*-axis scaling to facilitate visualization: (a) breakfast,
279 vegetable stir-fry cooked on gas, and beef chili and (b) vegetable stir-fry cooked on the hot plate, lasagna,
280 and toast. The shaded region represents standard error. n = number of replicates.
281

Naked hot plate and naked stove experiments were performed to characterize PM generated during the operation of just the heat source. Indoor particle number concentrations were >1000× higher than background levels during the operation of the stove and hot plate (with no food or pans). While most particles were <10 nm for both the stove (99.2%) and hot plate (96.2%), the concentration of sub-10 nm particles from operating the stove (7.6×10^6 cm^{-3}) was almost 7× higher than from operating the hot plate (1.1×10^6 cm^{-3}, Fig. S16a). Wallace et al.[6] also reported higher emissions of sub-10 nm particles from a gas burner compared to an electric stovetop. Additionally, we observed a second aerosol mode at ~21 nm for the hot plate (Fig. S16a). A similar observation made by Wallace et al.[6] was attributed to a thin film formation or deposition of dust on the stovetop surface between uses, and this explanation was further strengthened in another study[55] where the continuous use of a hot plate eventually led to zero UFP emissions, but the same hot plate could again generate UFP when heated after a few days of no use. Residues of detergents, skin oils, and organics accumulated on a cooking pot surface can also generate UFP at rates varying with factors such as pan type, temperature, and duration of disuse.[55,56]

Sub-10 nm particle concentrations for stir-fry cooked on the stove ($5.5 \pm 0.4 \times 10^7$ cm^{-3}) were >8× higher than for those cooked on the hot plate ($6.3 \pm 0.6 \times 10^6$ cm^{-3}, Fig. S16b). Because particles originating from the heat source were too small to affect PM mass, most of the mass-based PM emissions observed during cooking can be attributed to food ingredients, especially oils.[31] A previous study[7] reported a much lower sub-100 nm particle concentration (2.7×10^4 cm^{-3}) for vegetable stir-fry because their lower size cut-off was 10 nm, and therefore, a big fraction of particles emitted from the gas stove was not measured. The same study also reported a much lower particle diameter mode (118 nm) because their upper size cut-off was 950 nm. In this study, we observed a mode beyond 1 μm (Fig. S16).

3.3 Ultrafine particle formation during mopping

Few studies have investigated the effects of chlorine-based cleaners on particle-phase chemistry indoors. Wong et al.[12] recorded increases in particle-phase chlorine after each bleach cleaning as measured by an aerosol mass spectrometer but could not attribute the increase in particle-phase chlorine to a chemical or physical process. Various studies have investigated chemical pathways taken by chlorinated species originating from bleach,[12,57,58] but these investigations focused on gas-phase chemistry or on uptake of gas species on PM surfaces.[59] Wang et al.[60] reported particle formation from terpenes and bleach emissions upon illumination via radical chemistry initiated by the photolysis of HOCl and Cl_2.

During HOMEChem, the living room and kitchen floors were mopped with a bleach solution for 10 minutes on the evenings of Layered Days. The living room had large west-facing windows and the kitchen had small east-facing windows. Particle formation and growth (up to ~30 nm) were observed within five minutes of the start of mopping (Fig. 3). Peak number concentrations were comparable to those associated with cooking; however, the resulting increase in PM mass concentration was <10 ng m^{-3}. The time-series concentration of the smallest particles, a proxy indicator for nucleation, peaked within ten minutes of the start of mopping (Fig S17). One size distribution was recorded every five minutes and, therefore, the transition from particle formation via nucleation to particle growth via condensation might not have been fully captured.

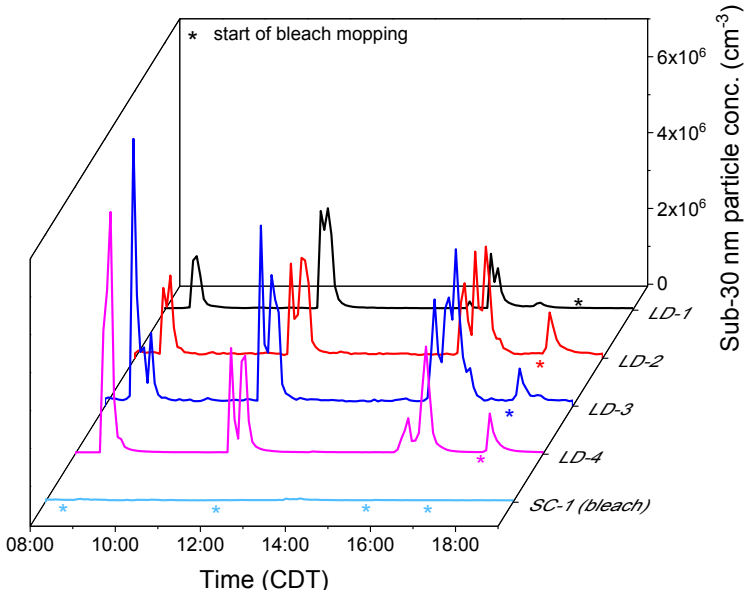

Figure 3. Sub-30 nm particle concentration in the test house during the four Layered Day experiments (coded as LD-1, LD-2, LD-3, and LD-4) and one Sequential Cleaning experiment with bleach (coded as SC-1). During LD-1, a lasagna was cooked for dinner, whereas chili was prepared for all other Layered Day experiments.

Interestingly, no new particle formation was observed in association with any of the bleach mopping events during the two Sequential Cleaning experiments (Fig. 3 and Fig. S15a-b) or during the Layered Day in which bleach mopping was preceded by lasagna instead of chili cooking. We hypothesize that residual gas-phase emissions associated with chili cooking and bleach cleaning participated in the physicochemical processes leading to new particle formation.

3.4 PM exposure and uptake in the human respiratory system

The United States Environmental Protection Agency (USEPA) ambient air quality standard for $PM_{2.5}$ is 35 μg m^{-3} for 24 hours, corresponding to a cumulative daily exposure of 840 μg m^{-3} h. $PM_{2.5}$ exposure levels over 12-h periods during the Thanksgiving events (~740 μg m^{-3} h) were close to this health-based exposure limit. Exposures for 12-h periods on Layered Days (170 ± 20 μg m^{-3} h) were about a quarter of those during the Thanksgiving experiments, well below the

USEPA threshold. Table 1 focussed on PM concentrations averaged over the cooking duration. To quantify the importance of the post-cooking decay period, Table 2 presents the exposures during cooking and post-cooking decay periods. These post-cooking periods contributed 36-77% of total exposure for each type of cooking activity. Exposures during breakfast, stir-fry, and chili cooking were comparable, but average exposure during the post-cooking decay was >2× higher for breakfast compared to chili.

Table 2. Average exposures for $PM_{2.5}$ ($\mu g\ m^{-3}\ h$) over the duration of cooking and post-cooking decay period for different meals cooked during the Layered Day (breakfast, stir-fry, toast, beef chili, and lasagna) and Sequential Day experiments (stir-fry).

Meal	During cooking	During post-cooking decay period
Breakfast	25 ± 20	40 ± 20
Stir-fry	20 ± 10	30 ± 20
Beef chili	30 ± 10	15 ± 10
Lasagna	16	10
Toast	3 ± 1	10 ± 4

Fig. 4 presents the PM respiratory deposition (0.004-20 μm) calculated for an adult residing in the test house from 8 AM to 8 PM on a Layered Day and on Thanksgiving Day. Two comparison cases, Indoor-no activity and Outdoor, are also included. The Indoor-no activity case represents the lower bound of indoor PM respiratory deposition during HOMEChem; all windows and doors were closed, and no activities were performed. The Outdoor bar represents the respiratory deposition of an adult spending the same duration outside the test house. Lacking direct outdoor measurements, indoor data recorded when doors and windows were open (Sequential Ventilation, Table 1) were used as a proxy for outdoor data (Fig. S13).

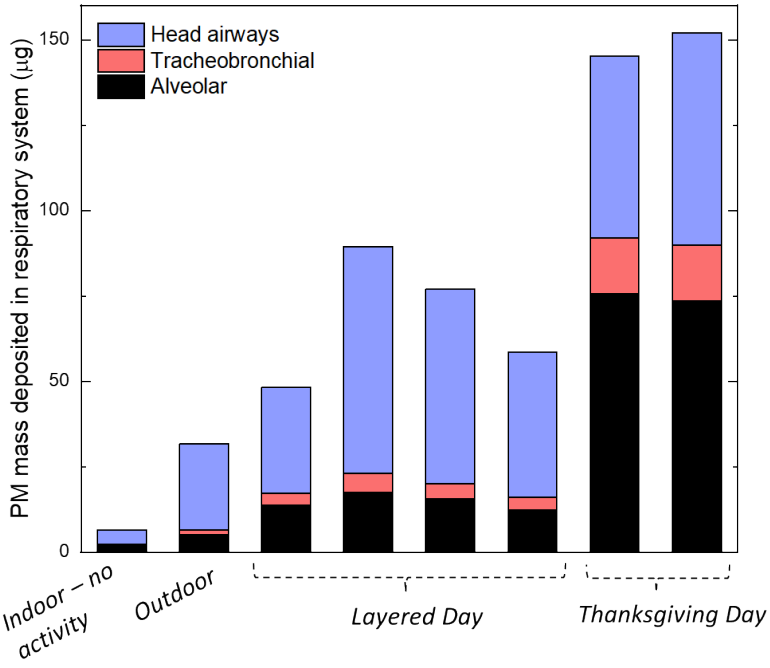

Figure 4. Estimated PM mass deposited in different parts of the respiratory system (head airways tracheobronchial, and alveolar) of an adult residing in the test house kitchen during four Layered Day experiments and two Thanksgivings. Indoor-no activity represents the potential exposure of an adult residing in the test house kitchen when no activities were performed and Outdoor represents spending the same amount of time outside the test house. For each bar, the duration of exposure is 12 h, spanning 8 AM to 8 PM.

The respiratory deposition during Indoor-no activity (7 μg) was lower than that corresponding to Outdoors (32 μg), as expected. In the absence of indoor sources, the house envelope provides partial protection from outdoor PM. Respiratory deposition masses were highest during Thanksgiving (145 μg and 152 μg) followed by Layered Day (70 ± 20 μg), 5× and ~2× higher, respectively, than that for Outdoors. These results suggest that in the US and in other countries that generally meet outdoor air quality standards, indoor exposure levels may dominate the overall exposure for groups of people who spend most of their time in residences, especially if cooking activities are frequent.

The PM fraction deposited in the head airways dominated (~75%) total mass deposited for all experiments except for the two Thanksgiving days, when the fraction deposited in the alveolar

region (~50%) was higher than in the head airways (~39%). During Thanksgiving, UFP were prominent contributors to PM mass, and particle deposition efficiency in the alveolar region is particularly high for this size range.

Respiratory depositions corresponding to the combined cooking and post-cooking decay period for different meals (Table S9) follow the same trends as exposures (Table 2). For all meal types, <10% of the total respiratory deposition occurred in the tracheobronchial region. The head airways fraction represented ~70% for stir-fry, chili, and breakfast and ~50% for lasagna and toast. The highest fraction of alveolar deposition (~50% of total) was obtained for toast and lasagna.

3.5 Implications

The test house was operated continuously at fixed ventilation and recirculation rates (without filter) to characterize indoor air pollutants in a simplified setting relative to a real house. Therefore, the test house is not representative of a typical residence with regards to ventilation. However, the measured PM concentrations and calculated exposures and resulting respiratory deposition during simulated daily activities highlight the importance of seeking a deeper understanding of indoor air quality in residences. In the absence of indoor sources, PM exposure and respiratory deposition in indoor environments are expected to be lower than outdoors, but cooking (and potentially other) activities can lead to high indoor concentrations of PM and, therefore, potentially higher indoor exposure levels compared to spending the same amount of time outdoors. PM exposures during the post-cooking decay phase were higher than those during the cooking phase for some meals. Exposure to cooking emissions is likely also important for house occupants away from the kitchen area. High concentrations of UFP were observed during all cooking activities and propane combustion emitted mostly sub-10 nm particles. We lack an understanding of the health effects of exposure to high number concentrations of UFP, especially sub-10 nm particles from stove and

hot plate, which rarely accumulate to a high enough mass concentration to be of concern based on current air quality standards and guidelines. Apart from the apparent source of PM, i.e. cooking, new particle formation from the interplay between the emissions from cooking and bleach demonstrates that there might be other, non-obvious sources of UFP in residential environments.

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Supporting Information (SI)

- Descriptions of instrumentation and settings with figures; discussion, figures, and tables on particulate matter density assumption and associated uncertainty; equations used for lung deposition modeling; additional figures showing measurements during cooking, cleaning, and ventilation experiments.

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