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Authors
Tian, Yilin
Arata, Caleb
Boedicker, Erin
et al.

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Indoor Emissions of Total and Fluorescent Supermicron Particles during HOMEChem

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Yilin Tian[1*,*], Caleb Arata[2], Erin Boedicker[3], David M. Lunderberg[2], Sameer Patel[4], Sumit Sankhyan[4], Kasper Kristensen[1†], Pawel K. Misztal[1‡], Delphine K. Farmer[3], Marina Vance[4], Atila Novoselac[5], William W Nazaroff[6], and Allen H. Goldstein[1,6]

[1] Department of Environmental Science, Policy, and Management, University of California, Berkeley, CA

[2] Department of Chemistry, University of California, Berkeley, CA

[3]Department of Chemistry, Colorado State University, CO

[4]Department of Mechanical Engineering, University of Colorado, Boulder, CO

[5]Department of Civil, Architectural and Environmental Engineering, The University of Texas at Austin, Austin, TX

[6]Department of Civil and Environmental Engineering, University of California, Berkeley, CA

[*]***Corresponding email***: *tyl0714@gmail.com*

*****: *Now at Baseline Environmental Consulting*

†: *Now at Department of Engineering, Aarhus University, Denmark*

‡: *Now at Department of Civil, Architectural and Environmental Engineering, The University of Texas at Austin, TX*

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Abstract

Inhalation of particulate matter is associated with adverse health outcomes. The fluorescent portion of supermicron particulate matter has been used as a proxy for bioaerosols. The sources and emission rates of fluorescent particles in residential environments are not well understood. Using an ultraviolet aerodynamic particle sizer (UVAPS), emissions of total and fluorescent supermicron particles from common human activities were investigated during the HOMEChem campaign, a test-house investigation of the chemistry of indoor environments. Human occupancy and activities, including cooking and mopping, were found to be considerable sources of indoor supermicron fluorescent particles, which enhanced the indoor particle concentrations by two orders of magnitude above baseline levels. The estimated total (fluorescent) mass emission rates for the activities tested were in the range of 4–30 (1–11) mg per person-meal for cooking, and 0.1–4.9 (0.05–4.7) mg/h for occupancy and mopping. Model calculations indicate that, once released, the dominant fate of coarse particles (2.5-10 micrometer in diameter) was deposition onto indoor surfaces, allowing for the possibility of subsequent resuspension and consequent exposures over durations much longer than the ventilation time scale. Indoor coarse particle deposition would also contribute to soiling of indoor surfaces.

Keywords

Particulate matter; Fluorescent particle; Human activity; Sources; Cooking; Surface deposition.

Practical Implications

- Indoor sources can be more important than outdoor air for inhalation exposure to supermicron particles in residences.
- Human occupancy and activities, such as cooking and cleaning, increase indoor total and fluorescent coarse particle concentrations.
- Coarse particles emitted from human activities are mainly deposited indoors, and thus contribute to surface contamination, such as in organic films and associated chemistry.

1 Introduction

Epidemiology studies show associations between elevated ambient particulate matter concentrations and adverse respiratory and cardiovascular health outcomes[1,2]. Although most scientific and regulatory attention has focused on fine particulate matter, a systematic review by Brunekreef and Forsberg[3] concluded that coarse particles (2.5-10 μm in diameter) might have independent effects on respiratory morbidity. Consequently, inhalation exposures to coarse particles should not be overlooked. Because, on average, people spend 90% of their time indoors[4] and human activities can cause strong enhancement of indoor particle levels[5], coarse particle concentrations measured at ambient air monitor stations are probably not a good proxy of actual exposure concentrations. Therefore, developing knowledge about indoor sources and emissions of coarse particles could contribute to a better understanding of human exposures, facilitating the investigation of the health effects of coarse particles.

The fluorescent portion of coarse particulate matter has been measured in some studies as a proxy for viable airborne biological particles in ambient air and in the built environment.[6–13] These studies have mainly been undertaken using the ultraviolet aerodynamic particle sizer

(UVAPS) or the wideband integrated bioaerosol sensor (WIBS). Previous studies have observed autofluorescence from living cells (biofluorophores: riboflavin and NAD(P)H)[14–16] and from abiotic materials such as polycyclic aromatic hydrocarbons, humic-like substances, some secondary organic aerosol material, soot, optical brightening agents, fabric fibers, and mineral dust[17–20]. These findings suggest a possibility that fluorescence might be used as a marker of other type of particles besides bioaerosols, if the sources are well-characterized and understood. Previous work on indoor fluorescent particles has mainly focused on human emissions, such as by direct shedding of bacteria-laden skin flakes and resuspension of previously deposited material from clothing and flooring. Other common indoor activities, such as cooking, have received less attention as contributors to indoor fluorescent particles. Moreover, previous research on cooking emissions have mostly focused on $PM_{2.5}$ and ultrafine particles. The influencing factors and source strength of coarse particle cooking emissions indoors are not well-characterized.

To contribute toward filling these knowledge gaps, the primary goal of this work was to characterize the concentrations and emissions of supermicron particles from select human activities in a residential environment. Using an UVAPS, concentrations of total and fluorescent particles ranging from 0.6 to 10 μm in diameter were monitored in real-time during the 4-week campaign known as House Observations of Microbial and Environmental Chemistry (HOMEChem).[21] Two categories of experiments, sequential and layered, were tested with replication. Applying a mass-balance approach, total and fluorescent particle emission rates were estimated for these activities. In addition, the fate of indoor particles at HOMEChem was studied by means of model calculations, and the average particle mass accumulation rates were estimated for each type of experiment. The concentrations and emissions reported in this work are

restricted to particles in the aerodynamic diameter range 0.6-10 μm. A broad overview of particle concentrations and emissions during HOMEChem was reported by Patel et al.[22]

2 Methods

2.1 Site description

The experiments took place at the Building Energy and Environments test house (UTest House) at the J.J. Pickle Research Campus of the University of Texas at Austin during June 2018. The UTest House is a 111-m^2 manufactured house with a volume of 250 m^3, including a kitchen-living area, two bathrooms, and three bedrooms, as shown in Figure S1 (Supporting Information). Interior doors to bedrooms were left open during the campaign to facilitate mixing, while the bathroom doors were kept closed. The kitchen is equipped with a propane-fueled gas stove and oven, plus a dishwasher and a refrigerator. An electric hot plate was also used for some cooking experiments. The exhaust hood above the gas stove was not operated during this study. The UTest House is normally unoccupied and operated only for research purposes. There is vinyl flooring throughout. More details about the UTest House are reported in Novoselac and Siegel.[23] During the HOMEChem experiments, the UTest House was unfurnished except for three tables and some chairs in the kitchen-living area. To maintain consistent environmental conditions, the heating, ventilation, and air conditioning (HVAC) system was set to deliver outdoor air at a constant air-change rate of approximately 0.5 h^{-1}. Internal recirculation through the HVAC system was operated continuously at a rate equivalent to 8 house volumes per hour. No filter was used in the recirculation system to eliminate the influence of time-varying filter conditions. In addition to the forced-convection induced by the air handling system, a ceiling fan in the living area operated continuously to further promote mixing inside the UTest House. The thermostat of the HVAC system was set to maintain the temperature in the kitchen and living space at 25 °C.

The average temperature during the campaign period was measured to be 25 ± 2 °C and the corresponding indoor RH was 57 ± 6%.[21]

2.2 Experimental design

Experiments were conducted in the UTest House during June 1-28, 2018. Indoor total particle and fluorescent particle concentrations, as well as outdoor total particle concentrations, were monitored continuously throughout the campaign. Two categories of experiments, sequential and layered, were undertaken in this study. On sequential days, a single type of activity was undertaken multiple times in succession, with either enough house vacant time or a window and door open period between each experimental trial to minimize the influence of one run on the next. On layered experimental days, a series of scripted activities occurred over a period of approximately 10 h during the day. The layered days were designed to mimic real-life scenarios and were undertaken without any periods of vacancy or enhanced ventilation. The goal of sequential experiments is to study emissions and dynamic behavior of pollutants from an isolated event. The layered experiments provide the opportunity to probe potential influences from the interactions of common household activities such as cooking and cleaning.

Three activities were tested in sequential experiments: vegetable stir-fry, wet-mopping, and staggered occupancy. Two types of layered experiments were undertaken: baseline layered days and simulated Thanksgiving days. Experimental procedures for these five types of experiments are presented in S1, Supporting Information. Moreover, a detailed experimental schedule including an overall diary of the 4-week campaign has been reported in Farmer et al.[21] In summary, three broad categories of common indoor particle sources were studied in this work: cooking, quiet occupancy, and cleaning. Cooking activities include cooking stir-fry, cooking breakfast (baseline layered day), cooking chili (baseline layered day), and cooking Thanksgiving

dinner. Quiet occupancy includes the staggered occupancy experiment (staggered occupancy) and the time between activities on the baseline layered day (seated occupancy) when volunteers sat at the kitchen table doing computer work. Cleaning activity includes wet-mopping. For the particle size range of primary interest here, 1-10 μm, cooking emissions were mainly from the ingredients cooked (including cooking oils), while emissions from quiet occupancy and cleaning were attributable to shedding from occupants' skin and clothing, as well as resuspension from flooring and other indoor surfaces contacted by the occupants.

In total, 38 volunteers participated in HOMEChem. Each participant was assigned a volunteer ID (i.e. V1). The requirement for a human subject protocol was waived for the HOMEChem campaign. As a condition for this waiver, no personal information was recorded. Volunteer ID was recorded in the HOMEChem activity logs only to facilitate the analysis of person-to-person variability.

In addition to the two broad types of experimental days, seven other days of two types of activities were programmed into the HOMEChem campaign: unoccupied background days (n = 2) and instrument maintenance days (n = 5).

2.3 Instrumentation

An ultraviolet aerodynamic particle sizer (UVAPS; model 3314; TSI Inc, Shoreview, MN, USA) was placed in the middle of the kitchen-living area (Fig. S1), with its sampling inlet at about 1.5 m in height, which corresponds to the breathing zone of a standing person. The UVAPS measures aerodynamic diameter, number concentration, and fluorescence intensity of particles. For particle fluorescence intensity measurements, the UVAPS uses a fixed excitation wavelength of 355 nm, and detects an emission region of 420-575 nm.

In addition to the UVAPS, two aerodynamic particle size spectrometers (APS; model 3321; TSI Inc, Shoreview, MN, USA) were deployed to monitor the aerodynamic diameter and concentrations of particles over the same size range indoors and outdoors, respectively. The indoor APS (APS2) was located next to the UVAPS at a lower height (~0.5 m) to explore the vertical gradient of supermicron particles. As shown in Figure S1, the outdoor APS (APS1) was placed in Bedroom 2 and sampled outdoor air through electrically conductive tubing and a diffusion dryer. Outdoor sampling tubing length, including the diffusion dryer, was 0.9 m. APS1 data were post-processed to correct for tubing losses, based on the theoretical estimation presented in Figure S2. Outdoor fluorescent particle concentrations were measured using the UVAPS on June 14-16, 2018, and then on June 23-24, 2018, when no experiments were scheduled.

The UVAPS and APS have similar characteristics regarding particle aerodynamic measurement. The instruments have 52 size channels, and sample at a 1 L/min flow rate with an additional 4 L/min of sheath air. With 1-min sampling interval including a 10-s wait time, the detection limit of both two instruments was 1.2 particles/L. The data reported in this study range from 0.6 to 10 μm in aerodynamic diameter. Measurements of particles larger than 10 μm are not reported here due to their rapid deposition. For some analyses, UVAPS and APS particle size channels ranging from 0.6 μm to 10 μm diameter were clustered into 13 bins as shown in Table S1, or into 3 bins: 1-2.5 μm, 2.5-5 μm, and 5-10 μm. Besides particle size channels, the UVAPS has 64 fluorescence intensity channels (FI, reported in arbitrary units). Based on fluorescence intensity, the UVAPS data were sorted into two categories: total particles, N_T (FI $\geq$ 0); and fluorescent particles, N_F (FI $\geq$ 2). The fluorescent intensity channel 2 (FI = 1) was excluded from

the fluorescent particle count to eliminate interference from non-fluorescent particles.[6,24] All APS data are for total particles, without regard for their fluorescence.

2.4 Quality assurance

Instrument maintenance and performance checks were conducted every week throughout the HOMEChem campaign. The flow rates were confirmed using a primary standard flow meter (model: Defender 510; Mesa Laboratories, Butler, NJ, USA). Particle sizing calibration of the UVAPS and the two APS units plus the fluorescence response of the UVAPS were examined using monodispersed polystyrene latex (PSL) particles and fluorescent particles in the size range 0.6-1.5 μm (Duke Scientific Corp., Fremont, CA, USA; Thermo Scientific, Fremont, CA, USA). A sizing offset of less than 0.1 μm was detected for the UVAPS. The UVAPS response was adjusted to correct the offset, using the calibration curve provided in Figure S3a. After adjustment, the lower bound of the UVAPS shifted to 0.6 μm. APS sizing performance agreed well with the manufacturer's set values. Using data obtained from a side-by-side collocation test, the adjusted UVAPS response was evaluated against APS2 and showed good agreement (Figure S3b). Assuming the number-weighted size distribution $\mathrm{d}N/\mathrm{d}(\log d_a)$ is constant across each size channel, the corrected UVAPS responses were processed to match the upper and lower range of the 13 size bins presented in Table S2. Collocation tests were carried out at the beginning and the end of the campaign; the resulting adjustment factors (*AF*) are presented in Table S2. The UVAPS was designated as the reference unit. During the collocation tests, A1 ultrafine Arizona Test Dust (ATD; ISO-12103-1, Powder Technology Inc, Arden Hills, MN, USA) was released in the test house multiple times to elevate the particle concentrations. During these tests, the UTest House ventilation system was operated in the same way as on regular experimental days.

2.5 Data analysis

2.5.1 Assessing emissions

Particle emissions were assessed for six activities, including the three used in sequential experiments (vegetable stir-fry, wet-mopping, and staggered occupancy), and three activities isolated during the layered day experiment (breakfast preparation, chili cooking, and seated occupancy). Analysis is based on a single-compartment material-balance model as shown in Equation 1, which assumes well-mixed conditions throughout the house volume.

$$\frac{dN_{in}(t)}{dt} = \frac{E(t)}{V} - (a+k)N_{in}(t) + apN_{out}(t) \quad (1)$$

In Equation 1, N_{in} and N_{out} are the indoor and outdoor particle concentrations (number/m^3) at time t, E is the particle emission rate (number/h), V is the indoor mixing volume (m^3), a is air-change rate (h^{-1}), p is penetration factor (-), and k is deposition loss-rate coefficient (h^{-1}) representing all particle loss mechanisms except air change. Detailed calculation procedures are discussed in S2, Supporting Information.

2.5.2 Converting number concentration to mass concentration

All particle mass concentrations and mass emission rates (mg h^{-1}) reported in this study were converted from measurements of particle number concentration. To obtain mass concentrations, the number concentrations were first converted to volume concentrations assuming all particles were spherical and that the volume-weighted size distribution (dV/d (log d_a) is constant within each size bin, using the method described in Zhou et al[25]. The conversion factors used in this analysis step are listed in Table S3. Then, mass concentrations were estimated using the volume concentrations and assuming that all particles have a density of 1 g/cm^3. The density of atmospheric particulate matter can vary with composition from 1 g/cm^3 to

2.5 g/cm^3.[5,26] Cooking oil has a slightly lower density than this range, about 0.9 g/cm3. Mass concentrations reported here should be considered as near-lower-bound estimates. In addition, the APS determines particle aerodynamic diameter based on the particle velocity in an accelerating airflow through a nozzle, and the motion of particles can be outside of the Stoke regime (Re > 0.5). Particle density affects the sizing of particles whose density is different from the spherical particles used to calibrate the instrument (ρ = 1.05 g/cm^3). Estimated using equations obtained from Wang and John, particles with a density of 2.5 g/cm^3 at 1 µm, 3 µm, and 10 µm would be oversized by 2%, 7%, and 12%, respectively.[27] In this study, particulate matter (PM) mass concentration PM_1, $PM_{2.5}$, PM_{10} refer to the mass of particles in the 0.6-1 µm, 0.6-2.5 µm, and 0.6-10 µm aerodynamic diameter range, respectively.

3 Results and Discussion

3.1 Total and fluorescent particle concentrations

Figure 1 presents the total particle and fluorescent particle mass concentration time series (upper two panels) and particle number size distributions (lower two panels) for each type of experiment. These data illustrate the influence of common human activities on coarse particle levels. Figures 1a-1c focus on cooking emissions; Figure 1d depicts the influence of a cleaning activity (mopping), and Figure 1e shows the effect of quiescent human occupancy. Detailed activity logs for each experimental day are provided in Tables S5-S9.

A representative sequential vegetable stir-fry cooking day is illustrated in Figure 1a, with cooking activities highlighted in lilac. Rice cooking, which was conducted in the first half of vegetable stir-fry experiments, did not strongly influence indoor total particle levels. The small increase in coarse fluorescent particles during this period is likely attributable to the motion of

human occupants rather than to cooking per se. In contrast, stir-frying vegetables enhanced the total and fluorescent particle concentrations by two to three orders of magnitude. Two peaks were observed during the stir-frying experiments for both types of particles. The first peak was associated with adding about 1.5 L of frozen vegetable to hot oil (average pan temperature at the time of addition: 100 ± 20 °C); the second peak, which occurred close to the end of stir-fry cooking, was associated with adding sauce to the browning vegetables. Averaged over event duration, submicron particles within the range monitored (0.6-1 μm) contributed approximately 70% and 15% of the total particle number concentration and mass concentration, respectively, considering the range 0.6-10 μm diameter. Whereas the number concentrations were dominated by submicron particles, the majority of particle mass emitted from stir-fry cooking was in the supermicron range (1-10 μm). Regarding submicron fluorescent particles, no influence of stir-fry cooking was observed. Among supermicron fluorescent particles, about 91% of the mass concentration was in the coarse mode (2.5-10 μm).

Similar trends were observed for the other two types of cooking experiments, the baseline layered day (Figure 1b) and Thanksgiving day (Figure 1c). As is clearly evident in Figure 1b, activities such as cooking breakfast, stir-frying vegetables, and cooking chili were prominent indoor sources of total and fluorescent particles, resulting in approximately two to three orders of magnitude higher particle concentrations compared to the background levels. During breakfast cooking, the spikes in total and fluorescent particle concentrations at the end of the event were associated with adding tomatoes to a non-stick pan with hot oil and sausage grease in it, which caused evident splattering. For chili cooking on the layered day (Figure 1b), the initial particle concentration spike occurred with the addition of ground beef to hot oil, and the other two spikes were associated with increasing pan temperature and adding more ingredients such as jalapeño

pepper. During chili cooking, no evident emissions of supermicron particles were observed after adding sliced tomatoes and beef stock to the wok, which effectively changed the cooking method from pan-frying to stewing. Apart from these three cooking activities, which are mostly oil-based, the preparation of other dishes, such as roasting turkey in the oven (dry cooking) and cooking cranberry sauce (water-based cooking), were done consecutively on the simulated Thanksgiving day. As shown in Figure 1c, most of the Thanksgiving cooking (highlighted in light green) did not cause high emissions of supermircron total and fluorescent particles that would be comparable to cooking breakfast, except for browning meat in a pan to make gravy. Also, cooking toast using an electric toaster, which was known to be a source of ultrafine particles[28], was conducted on the layered day and no emission of supermicron particles was observed. In summary, oil-based cooking with high cooking temperature, such as stir-frying and browning/charring, were associated with strong emissions of supermicron total and fluorescent particles, whereas certain types of dry cooking (oven baking and toasting) and water-based cooking (boiling and stewing) tested in this work did not materially influence supermicron particle levels. These results are qualitatively similar with previous studies on ultrafine particles and $PM_{2.5}$, in which it was found that pan-frying produced higher indoor particle concentrations compared to boiling, stewing, and oven cooking[29–31].

As illustrated in Figure 1d, wet-mopping enhanced supermicron total particles and fluorescent particle concentrations, with stronger influence observed for the fluorescent portion. One person mopping vigorously led to a sharp increase in coarse particle concentrations over a short period of time (~10 minutes). No increment in submicron particle concentrations was observed for both types of particles, likely due to the combination of moderate background levels in the house and negligible emission of particles in this size range.

Figure 1e shows that indoor fluorescent particle concentrations were positively correlated with the number of occupants in the UTest House (light green line) during the staggered occupancy experiments. This observation is consistent with previous studies investigating the effect of human occupancy on fluorescent particles and airborne bacteria concentrations[11,13,32]. For supermicron fluorescent particles, human occupancy elevated indoor concentrations to about two orders of magnitude above the background level, probably because of a combination of direct shedding from the human envelope, particle release from clothing, and resuspension from floors and other contacted surfaces[33,34].

We observed that opening windows and doors (highlighted in light blue) acted as a net source on the sequential experiment days (Figures 1a, 1d, 1e). This finding applied for both total particles and for fluorescent particles. Except for the drop in PM_1 concentrations on the vegetable stir-fry day, opening the windows and doors of the house resulted in higher particle concentrations indoors due to enhanced introduction of particles from outdoor air. As shown in Figures 1d and 1e, opening windows and doors of the UTest House led to higher supermicron total particles than either the cleaning activity (wet mopping) or dense seated occupancy. After the window and door open period, indoor total and fluorescent supermicron particle concentrations declined to baseline levels within an hour.

The experimental activities were observed to change the ratios of fluorescent to total particle number concentrations (N_F/N_T). (See Figure S7.) Compared to the baseline level during house unoccupied periods, human occupancy and activities were associated with higher N_F/N_T ratios. Although opening windows and doors produced higher particle levels than occupancy-associated emissions, the enhanced ventilation intervals had a much smaller influence on N_F/N_T ratios than did emissions from occupancy and from occupant activities.

In comparing fluorescent particles to total particles during these experiments, we observed substantial differences in number size distributions, especially for the three oil-based cooking activities (breakfast preparation, vegetable stir-fry, and chili cooking). Total particle concentrations decreased with increasing particle size, whereas fluorescent particle concentrations peaked in the 1.6-3 μm and 5-7 μm range for vegetable stir-frying and breakfast cooking, respectively. To further explore the possible source of fluorescent particles, the following supplemental activities were tested: heating the non-stick pan, cooking tomato in non-stick pan without oil, heating the non-stick pan with oil until the oil is smoking, and splashing water into smoking oil to produce oil splatter. Only the last of these activities produced high concentrations of supermicron fluorescent particles. As displayed in Figure S8, the number size distributions of fluorescent particles produced by oil splattering and breakfast cooking are comparable, as they both peaked in the 5-7 μm range. This result demonstrates that oil splattering is a source of indoor supermicron fluorescent particles, consistent with the report of Kanaani et al.[35] who found that aerosolized canola oil produced strong UVAPS fluorescent signals.

3.2 Cooking, occupant, and cleaning emissions

Six types of activities that caused discernible increases in total and fluorescent particle concentrations were analyzed: cooking vegetable stir-fry, cooking breakfast, cooking chili, staggered occupancy, seated occupancy, and wet-mopping. Arithmetic mean mass emissions of size-integrated supermicron total and fluorescent particles from these activities are reported in Figure 2. Approximately 80% and more than 95%, respectively, of total and fluorescent particle mass emitted were in the coarse size range (2.5-10 μm). Size-resolved particle number emissions are presented in Figures S9 and S10.

As shown in Figure 2a, cooking related sources emitted an average of 8-15 mg of particles, including 2.5-6.4 mg of fluorescent particles, per person-meal. (Here, one person-meal represents the amount of food appropriate to feed one person one meal.) Cooking a vegetable stir-fry meal, which took 17 ± 4 min, produced a higher number of total particles than did cooking breakfast (cooking time also 17 ± 4 min). Cooking vegetable stir-fry and breakfast were associated with similar emissions of fluorescent particles. As shown in Figure S9, cooking activities emitted different total and fluorescent particle size distributions. Total particle emissions decreased with increasing particle size, while fluorescent particle emissions peaked at ~3 to 4 μm.

Per person emission rates during quiet occupancy experiments, such as staggered occupancy and seated occupancy, are presented in Figure 2b. Staggered occupancy (average occupancy level = 7.5) and seated occupancy (occupancy level = 3) produced emission rates of 0.19 mg h^{-1} and 0.15 mg h^{-1} of total supermicron particles per person, and 0.17 mg h^{-1} and 0.09 mg h^{-1} of fluorescent supermicron particles per person, respectively. Staggered occupancy produced more particles per person than did seated occupancy, mainly because of differences in activity level. Staggered occupancy included volunteers entering and leaving the UTest House (walking with shedding and resuspension) eight times and sitting at a table doing light work, whereas seated occupancy mostly involved stationary activities with more limited movement. The overall average total particle emission rate for quiet occupancy, 0.18 ± 0.06 mg h^{-1} per person, is in good agreement with the 0.25 ± 0.04 mg h^{-1} per person mass emission rate for seated occupants reported by Licina et al.[36] As shown in Figure S10b and S10d, the fluorescent particles associated with human emissions peaked in the 2-4 μm diameter range, which also is

341 consistent with previous studies investigating fluorescent biological aerosol particles using
342 UVAPS.[6,11,12,13]

343 As shown in Figure 2b, wet-mopping was associated with about an order of magnitude
344 higher total and fluorescent particle emission rates than was staggered occupancy, likely
345 attributable to the difference in physical activity level. McDonagh and Byrne[37] found that high
346 physical activity produced about 10 times more particle mass than did low physical activity via
347 shedding of previously deposited materials from clothing. A clear effect of physical activity level
348 was also reported by Bhangar et al.[11], who evaluated fluorescent particle emissions from seated
349 and walking occupants using UVAPS. As illustrated in Figure S10c and S10d, total and
350 fluorescent particle number emission rates for wet-mopping had different modes. The former had
351 a mode at around 1 μm, while the latter peaked at around 3 μm.

352 **3.3 Fate of indoor particles at HOMEChem**

353 Relative contributions of outdoor air and indoor sources to the indoor total and
354 fluorescent particle concentrations at HOMEChem were evaluated for the layered experiments,
355 including four baseline layered days and two simulated Thanksgiving days. The indoor
356 concentration attributable to outdoor particles was estimated using outdoor total particle
357 measurements, size-resolved fluorescent to total particle ratio, and size-resolved infiltration
358 factors as described in S1, Supporting Information. Figure 3 shows the normalized indoor
359 particle apportionment for two categories: introduction via ventilation from outdoor air, and
360 emissions from indoor sources. For both layered experimental days, introduction via ventilation
361 from outdoor air contributed less than 25% and 10% of the indoor total particle and fluorescent
362 particle concentrations, respectively. Emissions from indoor sources contributed to a higher

363 portion on the Thanksgiving days than on the baseline layered days, as a result of enhanced
364 emissions from cooking activities.

365 In addition to particle sources, particle sinks must be considered to understand the
366 influence of human activities on indoor environmental quality. For HOMEChem, major sinks of
367 indoor airborne particles are removal via ventilation and deposition onto interior surfaces
368 including walls, horizontal surfaces, furniture, cabinets, and internal surfaces of the ventilation
369 system. Removal by filtration is not considered here because there was no filter in the
370 recirculating airflow for the UTest House HVAC system during the HOMEChem campaign. For
371 the particle size range of interest in this study, coagulation is negligible and so it is excluded
372 from consideration. Figure 4 shows the normalized removal rates by means of ventilation and
373 deposition, estimated using the experimentally determined average air-change rates and size-
374 dependent deposition loss rate coefficients, *k*. Two sets of size-resolved *k* values were used:
375 those estimated using vegetable stir-fry data (cooking) and those obtained from releases of
376 Arizona test dust (dust). The former represents the house conditions when cooking-related
377 activity was conducted, whereas the latter is used to represent the house condition during seated
378 occupancy experiments. The latter house condition provides a lower bound estimate of particle
379 deposition, because there was no cooking heat source or vigorous human activity to increase air
380 movement. For both conditions, the dominant fate of supermicron particles is deposition onto
381 interior surfaces, mainly attributable to gravitational settling. Even for the smaller particles in the
382 range studied, deposition could be important: about 30-70% of particles in the size range 0.6-1
383 μm are removed by deposition. These results indicate that considerable amounts of particles
384 emitted into the house (particularly from cooking) were deposited onto interior surfaces, which
385 might influence the formation of surface films that could affect indoor air composition through

386 interfacial chemistry. The results are representative for summer conditions where central air
387 conditioning systems are operated frequently. For moderate climate conditions, the relative
388 contributions of ventilation and deposition are expected to vary because of different window
389 opening behavior, for example, which can strongly influence air change rates of occupied
390 residences.

391 To estimate the contribution of indoor sources to the rate of coating of upward indoor
392 surfaces, average masses of total and fluorescent particles deposited per experimental day or per
393 event were estimated, assuming a well-mixed condition. Particle deposition onto the ceiling and
394 vertical walls was not considered in making these estimates. As shown in Figure 5a, similar total
395 particle accumulation rates of about 1.2 mg m^{-2} d^{-1} (about 0.5 mg m^{-2} d^{-1} for fluorescent
396 particles) were estimated for Thanksgiving Day and for the baseline layered day. These results
397 agree in scale with dustfall studies of Edwards et al.,[38] who reported average mass deposition
398 rates of 3.3 mg m^{-2} d^{-1} and 2.2 mg m^{-2} d^{-1} for residences in summer and winter, respectively. For
399 context, we note that Weschler and Nazaroff[39] predicted a smaller accumulation rate of 0.03-0.3
400 mg m^{-2} d^{-1} for the growth of organic films from gas-phase mass transfer onto impervious indoor
401 surfaces, independent of surface orientation. For sequential experiments (Figure 5b), stir fry was
402 associated with the highest total particle accumulation rate of ~ 0.4 mg m^{-2} $event^{-1}$ (about 0.15
403 mg m^{-2} $event^{-1}$ for fluorescent particles). The total particle accumulation rates for mopping and
404 occupancy were much smaller, about 0.015 mg m^{-2} $event^{-1}$ (about 0.005 mg m^{-2} $event^{-1}$ for
405 fluorescent particles) and 0.06 mg m^{-2} $event^{-1}$ (about 0.03 mg m^{-2} $event^{-1}$ for fluorescent
406 particles), respectively.

4 Conclusion

Indoor concentrations of supermicron total and fluorescent particles were strongly influenced by human occupancy and activities. Cooking-related activities tested at HOMEChem enhanced indoor supermicron total and fluorescent particle concentrations by two orders of magnitude above the background level measured during unoccupied periods. Seated human occupants caused a marginal increase in coarse total particle levels; vigorous movement, such as during wet-mopping, led to a sharp increase in supermicron fluorescent particle concentrations.

Among the human activities tested at HOMEChem, the dominant source of indoor supermicron total and fluorescent particles was oil-based cooking. Detailed investigation suggests that the fluorescent particles emitted from oil-based cooking likely originated from oil splattering. The water that caused the splattering may come from frozen vegetables or moist ingredients being added to hot oil. In contrast, water-based cooking, such as stewing and boiling, did not emit measurable quantities of supermicron particles. These results indicate that reduction in supermicron emissions can be achieved by altering cooking methods. On average, cooking activities tested at HOMEChem emitted 8–15 mg of total particles per person-meal, including 2.5–6.4 mg of fluorescent particles.

Most of the coarse particles emitted from human activities are predicted to deposit on the interior surfaces of the house. This finding suggests that the contributions of coarse particles from cooking to the organic and aqueous films on indoor surfaces should be considered as potentially important contributions to surface composition and therefore, potentially, to indoor surface chemistry.

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Figure 1. Representative indoor total and fluorescent particle mass concentration time series (upper two panels) and particle number size distributions (lower two panels) for five experimental days: (a) cooking vegetable stir-fry (stir-fry), (b) baseline layered day (layered day), (c) simulated Thanksgiving day, (d) wet-mopping (mopping), and (e) staggered occupancy. For the top two panels in each frame, event durations of each activity are highlighted with designated colors. The lilac color (stir-fry) is also used to indicate cooking breakfast, chili, and browning meat for the layered day and on the simulated Thanksgiving day. In frame (e), occupancy level (light green line) indicates the number of people inside the house during the staggered occupancy day. Note the PM_1 (blue lines), $PM_{2.5}$ (red lines), and PM_{10} (black lines) levels presented here have a lower size cut at 0.6 µm. The *y*-axis scales are different between cooking emissions (a-c) and human emissions (d-e).

Figure 2. Arithmetic mean size-segregated emissions of total and fluorescent particles (overall diameter range: 1-10 µm) associated with (a) cooking-related activities (particle mass emitted per person per meal), and (b) occupancy and cleaning (mass emitted per person per h). The respective number of experimental runs included in the analyses is shown above each bar. Note that the *y*-axis scales are different between cooking and occupancy/cleaning-associated emissions.

Figure 3. Arithmetic mean normalized aggregate indoor supermicron total particle (left) and fluorescent particle (right) source apportionment. The data represent the fraction of the indoor concentration that is produced via its source category. The outdoor category represents introduction via ventilation from outdoor air, while the indoor category represents emissions from indoor sources.

Figure 4. Predicted fate of particles emitted in the UTest House. Data represent the 3-h average fraction of the emitted particles that are removed by each process after the emission event. The upper panel depicts normalized removal rates estimated using deposition loss rate coefficients, *k*, determined from cooking experiments, whereas the bottom panel represents those estimated using *k* values determined by the Arizona test dust release experiments.

Figure 5. Estimated average accumulation rate of size-integrated total (L_T) and fluorescent (L_F) particle mass on indoor horizontal upward-facing surfaces (a) per layered experimental day and (b) per sequential experiment event. For sequential experiments, "occupancy" refers to staggered occupancy.