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2	Supplementary Information
3	Fates and spatial variations of accumulation mode particles in a multi-zone
4	indoor environment during the HOMEChem campaign
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#### 45 Section S1: Instrument diagnostics and data treatment

#### 46 S1.1: Instrument background intercomparisons

We conducted intercomparisons over four days of the HOMEChem campaign, at the beginning from 1 June to 3 June and then in the middle of the campaign on 23 June, during which the four POPS were co-located with each other and the UHSAS. Both of these periods correspond to background when no activities were going on in the house. The POPS were stacked together with approximately 10 to 24 cm between inlets and were placed on a counter space <1m from the UHSAS inlet. No corrections were done in response to these intercomparisons.



55 Figure S1: Comparison of number concentration sub-range between the overlapping size regions

56 of the four POPS and the UHSAS showing good correlations between the instruments. The 1-57 second data is in grey and the 5-minute average of the data is in black in all the plots.

58 **Table S1:** Fit equations and  $R^2/X^2$  values for the POPS to UHSAS intercomparisons. In the

59 table, the fit to the 1-second data is listed first and the fit to the 5-minute average is listed second

60 in bold with a  $X^2$  instead of an  $R^2$  because of the presence of x and y error from the averaging.

Instruments	Slope	Int.	$R^2 / X^2$
POPS 1 vs UHSAS	$1.035 \pm 0.002$	$8.7 \pm 0.4$	0.901
	$1.11 \pm 0.04$	-6 ± 8	1.54
POPS 2 vs UHSAS	$0.893 \pm 0.002$	$4.5 \pm 0.4$	0.898
	$0.96 \pm 0.04$	<b>-8</b> ± 7	1.97
POPS 3 vs UHSAS	$0.920 \pm 0.002$	$5.4 \pm 0.4$	0.898
	$0.99 \pm 0.04$	-7 ± 7	1.70
POPS 4 vs UHSAS	$0.853 \pm 0.002$	$5.5 \pm 0.3$	0.902
	$0.92 \pm 0.04$	$-6 \pm 6$	1.72

61



63 Figure S2: Comparison of number concentration between the four POPS showing good

64 correlations between the instruments during background periods (average comparison slope of  $1.0694 \pm 0.0006$ ).

1.007

**Table S2:** Fit equations and  $R^2/X^2$  values for the POPS vs POPS intercomparisons. In the table, 67

the fit to the 1-second data is listed first and the fit to the 5-minute average is listed second in 68

69	bold with a X <sup>2</sup>	<sup>2</sup> instead of an R	$R^2$ because	of the presence	of x and	l y error fro	m the averaging.
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Instruments	Slope	Int.	$R^2 / X^2$
POPS 1 vs POPS 2	$1.124 \pm 0.001$	$9.3 \pm 0.3$	0.947
	$1.16\pm0.03$	$4\pm5$	2.30
POPS 1 vs POPS 3	$1.095 \pm 0.001$	$7.6 \pm 0.3$	0.950
	$1.13\pm0.03$	$2\pm5$	0.568
POPS 1 vs POPS 4	$1.181 \pm 0.001$	$6.9 \pm 0.2$	0.951
	$1.21\pm0.03$	$2\pm5$	0.662
POPS 2 vs POPS 3	$0.945 \pm 0.001$	$3.4 \pm 0.2$	0.945
	$\boldsymbol{0.97 \pm 0.03}$	-1 ± 4	1.65
POPS 2 vs POPS 4	$1.021 \pm 0.001$	$2.7 \pm 0.2$	0.948
	$1.05\pm0.03$	-1 ± 4	1.82
POPS 3 vs POPS 4	$1.051 \pm 0.001$	$3.9 \pm 0.2$	0.950
	$1.08\pm0.03$	-1 ± 4	0.432

70



71



instruments. The POPS slightly overestimate particles in their lower range and underestimate

74 75

particles at the upper end of their range when compared to the UHSAS. Comparisons between each POPS and the UHSAS are represented by the blue points; the orange points are the average 76

slope values from all four POPS. 77

*S1.2: Instrument intercomparisons during cooking* 78

79 On 23 June, after the second round of background intercomparisons, the fourth POPS was placed bellow the UHSAS inlet in the kitchen and left there for the remainder of the 80 campaign. Data collected from a breakfast, stir-fry, and chili cooking on 25 June was compared 81 for the overlapping size range  $(0.13 - 1 \,\mu\text{m})$  of the two instruments (Table S6, Figure S7). The 82

83 POPS was shown to be responsive to the sharp changes in aerosol concentration, given that there

84 was no delay time between the UHSAS and POPS response to cooking. Although the maximum

85 concentration listed by the manufacturer for the POPS is  $1,250 \text{ } \# \text{ cm}^{-3}$  the POPS used in the

86 comparison showed good agreement with the UHSAS at higher concentrations. While noise in

87 the POPS measurement increased at these elevated concentrations, averaging the data (1-minute

88 averages in the comparisons) resolved this and resulted in similar results as those obtained from

89 the background intercomparisons.

90 The two optical instruments were also compared against an SMPS present during the

91 campaign. Using the same data from 23 June data from each cooking type was compared from

92 the overlapping size ranges  $(0.06 - 0.535 \,\mu\text{m}$  for the UHSAS and 0.134 - 0.455 for the  $\mu\text{m}$ 

93 POPS) (Table S3, Figure S4). The comparisons indicate that average number distributions
94 measured by the instruments are comparable, which validates the use of these optical instruments

95 for the size dependent analysis presented. There was disagreement in the smallest sizes of the

96 UHSAS distribution, however, this data was not used in the spatial analysis presented.

97 Additionally, deposition of particles < 100 nm is not presented because of saturation (discussed</li>

98 in the next section) therefore excluding the size region that had the largest difference from the

99 SMPS's number distribution. The comparisons do indicate that the optical instruments

100 underestimate total particle number concentrations during the cooking events. Underestimation is

101 likely a result of differences in the composition and therefore refractive index of cooking

102 particles and the PSL's used to calibrate the instruments as well as saturation effects during

103 cooking. This does not impact this work because the focus here is not on characterizing the total

104 cooking emissions.

105 **Table S3:** Fit equations and  $R^2/X^2$  values for the POPS vs UHSAS vs SMPS intercomparisons 106 during cooking. In the table, the fit to the 1-second data is listed first and the fit to the 1-minute

average is listed second in bold with a  $X^2$  instead of an  $R^2$  because of the presence of x and y

error from the averaging. Background comparison from POPS 4 vs UHSAS is included in the

109 first column for context.

Instruments		Background	Breakfast	Stir-Fry	Chili
POPS vs	Slope	$0.853 \pm 0.002$	$0.766 \pm 0.007$	$0.803\pm0.009$	$0.573\pm0.006$
UHSAS		$\boldsymbol{0.92 \pm 0.04}$	$\boldsymbol{0.92 \pm 0.04}$	$\boldsymbol{0.87 \pm 0.02}$	$\textbf{0.83} \pm \textbf{0.08}$
	Int.	$5.5 \pm 0.3$	$29 \pm 2$	$-10 \pm 20$	$132 \pm 3$
		-6 ± 6	$-10 \pm 10$	$-2 \pm 8$	$20 \pm 30$
	$R^2 / \mathbf{X^2}$	0.902	0.928	0.854	0.740
		1.72	2.65	83.8	9.58
POPS vs	Slope		$0.55\pm0.08$	$0.62 \pm 0.01$	$0.5 \pm 0.1$
SMPS	Int		$-140 \pm 50$	$-22 \pm 9$	$20 \pm 60$
	X <sup>2</sup>		10.9	2.07	5.23
UHSAS vs	Slope		$0.43\pm0.04$	$0.427\pm0.005$	$0.51 \pm 0.06$
SMPS	Int.		$-10 \pm 50$	$260 \pm 40$	$100 \pm 200$
	X <sup>2</sup>		1.49	34.8	22.3



112 Figure S4: A sub-plot of each cooking event that had enough data to be used for

- 113 intercomparisons (breakfast, stir-fry, and chili) are presented on the **top row** followed by
- 114 distribution comparisons in the **second row**, intercomparison plots between the POPS and
- 115 UHSAS data in the third row, and intercomparison plots between the optical particle counters
- 116 (OPC) and the SMPS in the **fourth row**. The events are in order of the time they were done
- 117 during the experiment day from left to right in the plots. In the POPS vs UHSAS
- 118 intercomparison plots raw 1-second data is presented as grey, with the dashed line being the raw

119 intercomparison line, and 1-minute averaged data is presented in black, with the solid line being

the intercomparison line. In the OPC vs SMPS the black traces are for the POPS and the bluetraces are for the UHSAS.

#### 122 S1.3: Saturation of optical instruments at high concentrations

123 At high aerosol concentrations, optical instruments will saturate causing the instruments 124 to undercount particles. We present a brief analysis of those saturation levels through comparison of the aerosol number concentration of the two instruments to the SMPS that was present during 125 the HOMEChem study. The analysis uses the end of the day decay period from the two 126 Thanksgiving experiments conducted during the campaign (2018/06/18 and 2018/06/27) because 127 it was a period during which the aerosol concentration decayed naturally (no intentional venting) 128 129 from an extremely elevated level back to background while the house was unoccupied. For the 130 POPS analysis, data from POPS 1 in the kitchen were used. The comparison shows that the POPS consistently overestimates compared to the SMPS and a saturation level for the instrument 131 132 cannot be identified. The UHSAS agrees well with the SMPS for concentrations below ~3000 133 particles cm<sup>-3</sup> and saturation is a significant problem at concentrations above  $\sim 10,000$  particles 134 cm<sup>-3</sup>. This saturation is only affecting the second detector of the USHAS, which measures particles < 240 nm in diameter. For this analysis saturation could lead to slight suppression in the 135 136 number size distribution below 240 nm, however, saturation does not affect the volume estimations since particles from the first detector ( > 240 nm) are what dominate this 137 measurement. Additionally, since saturation leads to underestimation of concentration for 138 particles < 240 nm the measured loss rate of these aerosols is also suppressed. Periods where 139 saturation was affecting the data are identified accordingly in this analysis. 140



142 Figure S5: Total concentration from both optical particle counters (OPC) – the UHSAS and

143 POPS – plotted against the total concentration of the SMPS for the size regions that overlapped

144 between the instruments (60 - 530 nm for the UHSAS and SMPS, and 130 - 530 nm for the

145 POPS and SMPS).

### 146 S1.4: Data treatment and density assumption

147 Both instruments assume spherical particles in all number to area, volume, and mass conversions. All the mass measurements reported in the supplementary text used an assumed 148 density of 1.2 g cm<sup>-3</sup>, which was selected because of the high organic content of the observed 149 cooking-derived particles.1 Total mass is not the focus of this work and mass measurement 150 151 approximations are reported only to provide context. We recognize that for cooking aerosols this density is likely an overestimation and for background periods this density is a slight 152 underestimation. Patel et al. (2020) provides a detailed description of aerosol density during the 153 HOMEChem campaign using Aerosol Mass Spectrometer (AMS) data that was available. Patel 154 et al. (2020) report that aerosol density fluctuated between  $\sim 1.0$  g cm<sup>-3</sup> during cooking and  $\sim 1.5$  g 155 cm<sup>-3</sup> during non-cooking periods. Given this range, we take 1.2 g cm<sup>-3</sup> as a density for all the 156 data to provide mass estimations of measured aerosols, unless otherwise stated. Unit density (1.0 157 158  $g \text{ cm}^{-3}$ ) is used in the model simulations since it involved only cooking aerosol.

## 159 Section S2: Summary of the Lai and Nazaroff (2000) Indoor Deposition Model

160 Tables S3 – S5 summarize the variables and equations used by the Lai and Nazaroff (2000)

161 to calculate deposition velocity and loss rate of indoor aerosols. The information presented is a

162 recreation of the equation summary tables presented in the original paper.

Variable	Definition	Set Value
dp	Particle diameter	
u	Friction velocity	1 m/s
ρ <sub>p</sub>	Particle density	1000 kg m <sup>-3</sup>
$v_s$	Gravitational settling velocity	
v	Kinematic viscosity of air	
D	Brownian diffusivity of the particle	
$\mathbf{A_v}^\dagger$	Area of vertical surfaces	250 m <sup>2</sup>
$\mathbf{A_u}^\dagger$	Area of upward-facing surfaces	111 m <sup>2</sup>
$\mathbf{A_d}^\dagger$	Area of downward-facing surfaces	111 m <sup>2</sup>
$\mathbf{V}$ †	Volume of the room	250 m <sup>3</sup>

163 Table S4: Variables used in the Lai and Nazaroff (2000) model.

164 *†* These values are from the house measurements listed in the HOMEChem overview paper <sup>4</sup>.

Parameters	Equation
Integral †	$I = [3.64 Sc^{2/3} (a - b) + 39]$
	$a = \frac{1}{2} \ln \left[ \frac{\left(10.92  Sc^{-1/3} + 4.3\right)^3}{Sc^{-1} + 0.0609} \right] + \sqrt{3} \tan^{-1} \left[ \frac{8.6 - 10.92  Sc^{-1/3}}{\sqrt{3}  10.92  Sc^{-1/3}} \right]$
	$b = \frac{1}{2} \ln \left[ \frac{(10.92  Sc^{-1/3} + r^{+})^3}{Sc^{-1} + 7.669 \times 10^{-4}  (r^{+})^3} \right] + \sqrt{3} \tan^{-1} \left[ \frac{2  r^{+} - 10.92  Sc^{-1/3}}{\sqrt{3}  10.92  Sc^{-1/3}} \right]$
Schmidt number ( <i>Sc</i> )	$Sc = v D^{-1}$
$r^+$	$r^+ = d_p u^* (2v)^{-1}$
Deposition velocity, vertical surface	$v_{dv} = \frac{u^*}{I}$
Deposition velocity, upward horizontal surface	$v_{du} = \frac{v_s}{1 - exp\left(-\frac{u_s I}{u^*}\right)}$
Deposition velocity, downward horizontal surface	$v_{dd} = \frac{v_s}{exp\left(\frac{u_s I}{u^*}\right) - 1}$
First-order loss coefficient for deposition, rectangular cavity	$\beta = \frac{v_{dv}A_v + v_{du}A_u + v_{dd}A_d}{V}$

165 **Table S5:** Equations used in the Lai and Nazaroff (2000) model.

- 166 † Evaluated using the approximation that Brownian diffusivity is negligible compared to the
- 167 eddy diffusivity. This approximation is valid for diameters larger than 0.01  $\mu$ m. For smaller
- 168 particles, a numeric integration is required (see Table S9).

Particle Diameter, $d_p$ (µm)	Integral, I (–)
0.001	29.1
0.0015	49.1
0.002	71.0
0.003	120.3
0.004	174.9
0.005	234.2
0.006	297.4
0.007	364.0
0.008	432.7
0.009	504.5
0.01	579.3

169 **Table S6:** Numeric integration of the integral (*I*), done by Lai and Nazaroff (2000).

170

# 171 Section S3: Summary of the Emerson Outdoor Deposition Model

Tables S6 and S7 summarize the variables and equations used by Emerson et al. (2020) to

173 calculate deposition velocity and loss rate of aerosols, as well as the variables we selected to

174 represent the indoor environment. These calculations are a modification of the Zhang et al.

175 (2001) model.

# 176 Table S7: Variables used by Emerson et al. (2020). The constants in this table are bolded and the

177 value they were set to for the indoor environment are in the last column of the table.

Variable	Definition	Set Value
d <sub>p</sub>	Particle diameter	
u	Friction velocity	0.02 m s <sup>-1</sup>
v	Kinematic viscosity of air	
D	Brownian diffusivity of the particle	
C	Cunningham slip correction factor	
<i>u<sub>H</sub></i>	Windspeed	0.1 m s <sup>-1</sup>
ρ <sub>p</sub>	Particle density	1000 kg m <sup>-3</sup>
Zr	Hight of deposition measurement	2 m
LUC**	Land use category	1
SSC**	Seasonal select category	1
z <sub>0</sub> †	Roughness length	0.8 m
A <sup>†</sup>	Characteristic radius of collectors	2.0 mm
£0	Empirical constant for surface resistance	3
γ <sup>†,‡</sup>	Brownian constant	0.56
C <sub>B</sub> <sup>‡</sup>	Empirical constant for Brownian	0.3

α †	Impaction constant	1.0	
β‡	Empirical constant for impaction	0.6	
C <sub>Im</sub> <sup>‡</sup>	Empirical constant for impaction	0.1	
υ‡	Empirical constant for interception	1	
C <sub>In</sub>	Empirical constant for interception	2.5	

178 \*\*The LUC and SSC follow the numbering system used in Zhang et al. (2001), which

- 179 parameterized 15 land use categories and 5 seasonal categories.
- 180 *†*These parameters are dependent on the LUC and the SSC. The value tables for these parameters
- 181 are listed in Zhang et al. (2001), the values listed here are the ones associated with the LUC and
- 182 SSC that were used in this analysis.

183 *‡* These parameters were altered from the Emerson et al. (2020) model to better represent the

- 184 indoor data.
- 185 **Table S8:** Summary of main equations used in the Emerson et al. (2020) model.

Parameters	Equation
Deposition velocity $(V_d)$	$V_d = V_g + \frac{1}{(R_a + R_s)}$
Gravitational settling velocity $(V_g)$	$V_{g} = \frac{\rho_{p} d_{p}^{2} g C}{18 \eta}$
Aerodynamic resistance above the canopy $(R_a)^{**}$	$R_a = \frac{\ln\left(\frac{z_r}{z_0}\right) - \psi_H}{\kappa u^*}$
Surface resistance $(R_S)$	$R_s = \frac{1}{\varepsilon_0 u^* (E_b + E_{im} + E_{in})R_1}$
Collection efficiency from Brownian diffusion $(E_b)$	$E_b = C_B S c^{-\gamma}$
Collection efficiency from impaction $(E_{im})$	$E_{im} = C_{Im} \left(\frac{St}{\alpha + St}\right)^{\beta}$
Collection efficiency	

from interception  $(E_{in})$ 

$$E_{in} = C_{In} \left(\frac{d_p}{A}\right)^{\nu}$$

Bounce correction factor  $R_1 = exp(-St^{0.5})$ 

Schmidt number (Sc)

Stokes number (St)

 $St = \frac{V_g u^*}{g A}$ 

 $Sc = v D^{-1}$ 

186 \*\*For the indoor environment this term is used to represent the conditions of the air above the

187 sample inlet. The stability function ( $\psi$ ) was set to zero for the indoor environment.

# 188 Section S4: Experimental days and background conditions during HOMEChem

189 When no activities are performed indoors and all external doors and windows are closed,

190 penetration of outdoor particles into the building is the most significant source of indoor

191 particles. Out of the three main indoor events simulated during the HOMEChem campaign (i.e.

192 cooking, cleaning, and human occupancy), cooking produced the largest concentration of

193 submicron particles (Figure S14).

194 During background periods, the UHSAS (60 – 1000 nm) measured an indoor concentration of

195  $175.2 \pm 0.5$  particles cm<sup>-3</sup> (~1.4 ± 0.3 µg m<sup>-3</sup>) during the day and  $100.2 \pm 0.4$  particles cm<sup>-3</sup> (~ 0.8

196  $\pm 0.2 \ \mu g \ m^{-3}$ ) at night. Particle concentrations and size distributions were homogeneous

197 (concentration within 18%) throughout the house, and no inherent gradient was observed

198 between rooms (Figure S15). Trends in indoor particle concentration mirrored those observed

199 outdoors (Figure S15). Both the indoor and outdoor size distributions were bimodal with similar

200 modes but differences in overall magnitude (Figure S15). This similarly indicates that the indoor

201 particle concentrations during background periods are dominated by infiltration of outdoor

202 particles, consistent with previous findings.<sup>7-12</sup>

203 Particle infiltration can be characterized by the ratio between indoor and outdoor (I/O ratio)

204 particle concentrations when the house is closed. This I/O ratio is  $0.5 \pm 0.1$  for number

205 concentration ( $0.6 \pm 0.9$  for mass concentration) of submicron particles (Figure S15). The I/O

206 ratio varies according to particle size, with ultrafine (< 100 nm) and coarse (> 1  $\mu$ m) mode

207 particles penetrating least effectively and accumulation mode particles (100 nm – 1  $\mu$ m) entering

208 most freely with the peak in I/O around 150 nm (Figure S15). This trend is the result of

209 diffusion losses due to Brownian motion dominating small particles and gravitational settling

210 forces, impaction, and interception dominating larger particles, causing both modes to deposit in

211 the ventilation system and be removed when moving through the building envelope. For

212 submicron particles, the average I/O ratio is  $0.74 \pm 0.02$ . Size-dependent I/O ratios published by

- 213 Zhao et al. (2019) and Hussein et al. (2005) both showed the same trend with peaks in I/O at 100
- 214 200 nm. In the context of this work, the I/O ratio represents a general relationship that can be
- 215 used to investigate the dominating sources of particles indoors.



217 Figure S6: Time-series of three full sequential days when cooking (top), cleaning (middle), and

218 human occupancy (bottom) experiments were done in the house. Cooking had the largest number

219 of particles reported, while cleaning and human occupancy did not show a substantial increase

220 above the background concentration for the particle range sampled (60 - 1000 nm).





Figure S7: (a) Indoor and outdoor aerosol concentrations during a background period. (b) Size 224 225 distribution of aerosols indoors and outdoors during background. (c) During background periods 226 there were no significant gradients present throughout the house, even during the daytime periods (highlighted regions) when aerosol concentration spiked. (d) The distribution was also 227 consistent across the house during these periods. (e) Ratio of indoor to outdoor aerosol 228 concentration at all four sampling points. (f) Size-dependent indoor to outdoor aerosol 229 concentration ratio. 230

### 232 Section S5: Aerosol concentrations and distributions during cooking

- 233 During all cooking events, particle number concentration indoors increased to at least ten times
- 234 the indoor background concentration, and at least six times higher than the measured outdoor
- 235 concentration. Stir-fry events resulted in peak number concentrations, measured by the UHSAS,
- 236 of approximately  $5200 \pm 300$  particles cm<sup>-3</sup> ( $25 \pm 2 \mu g m^{-3}$ ), breakfast events had concentrations
- 237 of  $10800 \pm 600$  particles cm<sup>-3</sup> (19.9 ± 0.9 µg m<sup>-3</sup>), and chili cooking events had an average peak
- 238 concentration of  $5000 \pm 300$  particles cm<sup>-3</sup> (14.4 ± 0.6 µg m<sup>-3</sup>). Toasting bread produced the
- 239 largest peak number concentration of particles at  $12400 \pm 500$  particles cm<sup>-3</sup> ( $16.8 \pm 0.8 \ \mu g \ m^{-3}$ ).
- 240 All the cooking events produced high concentrations of fine and ultra-fine particles, with an
- 241 average count median diameter of  $110 \pm 10$  nm for all cooking. Median diameters were relatively
- 242 consistent through emission periods except for some stir-fry events, which exhibited a change in
- 243 their distribution during emission periods (Table S9, Figure S9).
- 244 S5.1: Distribution characteristics for the different cooking events
- 245 **Table S9**: Median diameters of aerosol distributions observed by the UHSAS during
- 246 HOMEChem cooking events

Cooking Event	CMD (nm)	SMD (nm)	VMD (nm)
Breakfast	$98 \pm 7$	$126 \pm 9$	$140 \pm 10$
Toast	99 ± 6	$123 \pm 8$	$136 \pm 7$
Stir-fry**	$110 \pm 10$	$180 \pm 20$	$230\pm20$
Chili	$101 \pm 9$	$140 \pm 10$	$170 \pm 20$
Total Cooking	$110 \pm 10$	$150 \pm 20$	$180 \pm 20$

- 247 \*\*This represents an overall average distribution during the stir-fry cooking events, however,
- some of the stir-fry events had a shift in the distribution during the event that resulted in different CMD values (see section S 3.2).



Figure S8: Average distributions for the different cooking events: (a) breakfast, (b) toast, (c) stir-fry, and (d) chili. Each plot has number distribution (dN) on the left, represented by the solid trace, and volume distribution (dV) on the right, represented by the dashed trace. The shaded region on the plots represents the standard deviation of the distributions. The seam at 240 nm is an artifact of the two-detector system on the UHSAS, and this point represents the switching point between the two instruments. In these distributions, both the breakfast and the toast sub 240 nm number concentrations are likely underestimated as a result of saturation.

258 S5.2: Distribution shifts during select stir-fry events For all the stir-fry experiments, two main

259 peaks in concentration were observed (Figure S9). Out of the stir-fry experiments done, six of

260 the events showed a shift in the distribution during the second peak in concentration. This is

261 likely due to a change in the emission during these events.



263 Figure S9: Time series of total number concentration (dN) from the UHSAS showing the typical

trend observed during stir-fry events. In six of the observed stir-fry experiments, the aerosol size

265 distributions between these two peaks changed substantially. This stir-fry event from 12<sup>th</sup> June

266 was one of the stir-fry experiments that exhibited this change in emission.

Table S10: Median diameters of aerosol distributions observed for the different periods during astir-fry event and the overall median diameter of the event



270 Figure S10: Average distributions for the two peaks during a stir-fry event: (a) first peak and (b)

271 second peak. Both plots have number distribution on the left, the solid trace, and volume

272 distribution on the right, the dashed trace. The shaded region on the plots represents the standard

- 273 deviation of the distributions. The seam at 240 nm is an artifact of the two-detector system on the
- 274 UHSAS, and this point represents the switching point between the two instruments.

# 275 Section S6: Loss of cooking aerosol

276 S7.1: Measured loss rate for cooking aerosols



277



279 cooking events (right). The size-dependent loss rates for the three types of stir-fries and the

280 different cooking events were not substantially different from each other. The highlighted region

- 281 of both plots represents the data below 100 nm that was heavily affected by saturation of one of
- 282 the UHSAS detectors, leading to suppressed concentration peaks and loss rates.



## 283

Figure S12: Measured loss rates at the four measurement points throughout the house. The highlighted region in the figure represents data that was heavily affected by UHSAS saturation, leading to suppressed concentration peaks and loss rates. No significant variation was observed between rooms for the aerosol loss rate.

288

289 S7.2: Deposition velocity and rate



- 291 Figure S13: Total average deposition rate and deposition velocity for all HOMEChem cooking
- 292 events from the UHSAS and the POPS. Error bars represent the standard deviation of the
- 293 measured deposition across the different experiments. The highlighted region in the figure
- 294 represents data that was heavily affected by UHSAS saturation, leading to suppressed deposition
- 295 rates. Data in this region is omitted from the modeled deposition analysis.



Figure S14: Percent difference between the modeled and measured deposition velocities and rates for the (a) Lai and Nazaroff (2000) indoor model and (b) Emerson et al. (2020) outdoor model.<sup>3,5</sup> These residuals were calculated for the UHSAS data and the deposition rate values from Tian et al. (2020).<sup>15</sup>



302 Figure S15: Deposition loss rates produced by different friction velocities in the Lai and

303 Nazaroff (2000) model.<sup>3</sup> Green points represent the UHSAS data (open circles) and the

304 deposition rate data presented in Tian et al. (2020) (closed circles) from the HOMEChem

305 study.<sup>15</sup>



308 Figure S16: Constraint of the surface-area-to-volume ratio (SA/V) in the Lai and Nazaroff (2000) model.<sup>3</sup> Surface area was increased by 20% (light grey line), 50% (grey line), and 80% 309 (black line) for the (a) vertical surfaces, (b) upward-facing surfaces, (c) downward-facing 310 surfaces, and (d) all surfaces. In all cases volume was decreased by 10% from the reported 311 volume for the test house. This manipulation was based on the work of Manuja et al. (2019), 312 where it was reported that on average the contents of a room result in a 50% increase in the total 313 surface area and a 10% decrease in the total volume.<sup>16</sup> None of these variations in the surface-314 area-to-volume ratio had a significant impact on the agreement between the observed deposition 315 rates and those produced by the model. Green points represent the UHSAS data (open circles) 316 and the deposition rate data presented in Tian et al. (2020) (closed circles) from the HOMEChem 317 study.<sup>15</sup> 318



**Figure S17**: Comparison between the values and model parameters found in Thatcher et al.

- 321 (2002) (open markers are bare room and closed markers are for a furnished room)and the values
- 322 obtained during HOEMChem. The HOMEChem deposition measurements lie within the range of
- 323 variability for indoor measurements that Thatcher et al. (2002) explored and found similar
- 324 underestimation of deposition from the Lai and Nazaroff (2000) model<sup>3,17</sup>. All model runs
- 325 assume unit density  $(1.0 \text{ g cm}^{-3})$  in this figure.



326 S7.3: Coagulation during cooking

- 328 Figure S18: Concentration map for a typical stir-fry event (left) and one of the Thanksgiving
- 329 Day experiments (right). Coagulation was not a major loss mechanism for particles produced
- 330 during pulsed events, however, when concentration was high for an extended period, like it was
- 331 during Thanksgiving, coagulation was observed.
- 332 Table S11: Major equations used in coagulation calculations, following Fuchs form of the
- 333 Brownian coagulation coefficient.<sup>18</sup>



$$\begin{array}{ll} \begin{array}{l} \text{Diffusivity}\\ (D_{i})^{*} & D = \frac{k_{B}T \ C_{C}}{3 \ \pi \ \mu \ d_{p,i}} \\ \end{array}$$

$$\begin{array}{l} \text{Mean thermal}\\ \text{speed of the}\\ \text{particle}\\ (\overline{c}_{i})^{**} & \overline{c}_{i} = \left(\frac{8 \ k_{B} \ T}{\pi \ m_{i}}\right)^{1/2} \\ \end{array}$$

$$\begin{array}{l} \text{Mean free}\\ \text{path} \ (l_{i}) & l_{i} = \frac{8 \ D_{i}}{\pi \ \overline{c}_{i}} \\ \end{array}$$

$$\begin{array}{l} \text{Mean free}\\ \text{path} \ (l_{i}) & l_{i} = \frac{8 \ D_{i}}{\pi \ \overline{c}_{i}} \\ \end{array}$$

$$\begin{array}{l} \text{Gravitational}\\ \text{constant} \ (g_{i}) & g_{i} = \frac{1}{3 \ d_{p,i} \ l_{i}} \left[ (d_{p,i} + \ l_{i})^{3} - (d_{p,i}^{2} + \ l_{i}^{2})^{3/2} \right] - d_{p,i} \\ \end{array}$$

$$\begin{array}{l} \text{Coagulation}\\ \text{coefficient}\\ (K_{i,j}) & K_{1,2} = 2\pi (D_{1} + D_{2}) (d_{p,1} + \ d_{p,2}) \left( \frac{d_{p,1} + \ d_{p,2}}{d_{p,1} + \ d_{p,2} + 2 (g_{1}^{2} + g_{2}^{2})^{1/2}} + \frac{8 \ (D_{1} + (\overline{c}_{1}^{2} + \overline{c}_{2}^{2})^{1/2}}{(\overline{c}_{1}^{2} + \overline{c}_{2}^{2})^{1/2}} \right) \\ \end{array}$$

334 \* The variable  $C_c$  is the Cunningham slip correction factor and  $\mu$  is the viscosity of air.

<sup>335 \*\*</sup> The variable  $m_i$  is the mass of particle *i*.



337 Figure S19: Results of theoretical calculations for the impact of coagulation on (a) stir-fry (total

338 starting concentration of 5197 # cm<sup>-3</sup>) and (b) Thanksgiving Day experiments (total starting

- 339 concentration of  $11813 \# \text{ cm}^{-3}$ ). In both panels, the average distribution for the experiments is
- 340 represented as the solid black line (t = 0), and the predicted distribution after having coagulation
- 341 as the only loss process for 1 hour is the dashed black line. Percent difference between the two
- 342 distributions, on the right axis, is displayed in green.

### 343 Section S7: Observed gradients in aerosol concentration during cooking

344 Table S12: Statistical data (maximum, 75<sup>th</sup> percentile, median, 25<sup>th</sup> percentile, and minimum) for

345 the percent differences between the kitchen and the living room during cooking. Absent numbers

in the larger bins are a result of excluding data where concentration was less than  $1 \text{ # cm}^{-3}$ .

Mean Diameter (µm)	Maximum	75th	Median	25th	Minimum
139.32	89.25	65.78	44.16	31.16	16.64
152.41	88.29	68.49	54.92	38.98	33.61
167.49	84.87	75.10	52.01	42.40	36.79
184.50	84.29	73.85	46.59	39.65	32.05
202.54	83.38	66.15	48.23	39.21	34.83
222.08	80.54	44.90	37.50	27.54	18.27
251.77	72.47	30.31	18.35	12.43	3.92
298.54	109.89	73.37	59.06	22.75	8.29
387.26	74.71	35.42	27.70	6.32	1.08
522.99	47.29	38.59	26.91	8.45	0.38
764.72	70.03	44.90	24.21	21.05	1.19
1132.40	121.47	75.811	42.03	17.33	5.41
1399.59	77.92	58.14	30.75	20.52	17.91
1627.42	81.90	81.90	43.70	5.50	5.50
2157.74	-	-	-	-	-
3002.62	-	-	-	-	-

347

348 **Table S13**: Statistical data (maximum, 75<sup>th</sup> percentile, median, 25<sup>th</sup> percentile, and minimum) for

349 the percent differences between the kitchen and the bedroom during cooking. Absent numbers in

350 the larger bins are a result of excluding data where concentration was less than  $1 \# \text{ cm}^{-3}$ .

Mean Diameter (µm)	Maximum	75th	Median	25th	Minimum
139.32	93.56	82.43	71.16	56.83	39.92
152.41	96.46	86.45	70.95	62.87	43.45
167.49	97.61	82.03	72.49	63.75	42.02
184.50	97.34	74.38	69.03	61.17	34.42
202.54	96.43	72.59	63.72	57.26	31.12
222.08	95.42	72.25	59.95	53.28	32.10
251.77	91.87	60.92	36.41	20.66	4.50
298.54	86.64	54.14	39.32	22.94	1.29
387.26	90.26	66.60	56.00	35.06	7.01
522.99	85.65	67.93	60.28	47.95	1.29
764.72	72.58	60.73	49.74	34.01	10.99
1132.40	76.85	58.64	50.77	34.36	8.66
1399.59	77.19	56.53	29.61	14.55	5.74
1627.42	74.08	74.08	38.84	3.61	3.61
2157.74	-	-	-	-	-
3002.62	-	-	-	-	-

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