



Desorption of SVOCs from Heated Surfaces in the Form of Ultrafine Particles

Wallace, Lance A.; Ott, Wayne R.; Weschler, Charles J.; Lai, Alvin C. K.

Published in:
Environmental Science and Technology

Link to article, DOI:
[10.1021/acs.est.6b03248](https://doi.org/10.1021/acs.est.6b03248)

Publication date:
2017

Document Version
Peer reviewed version

[Link back to DTU Orbit](#)

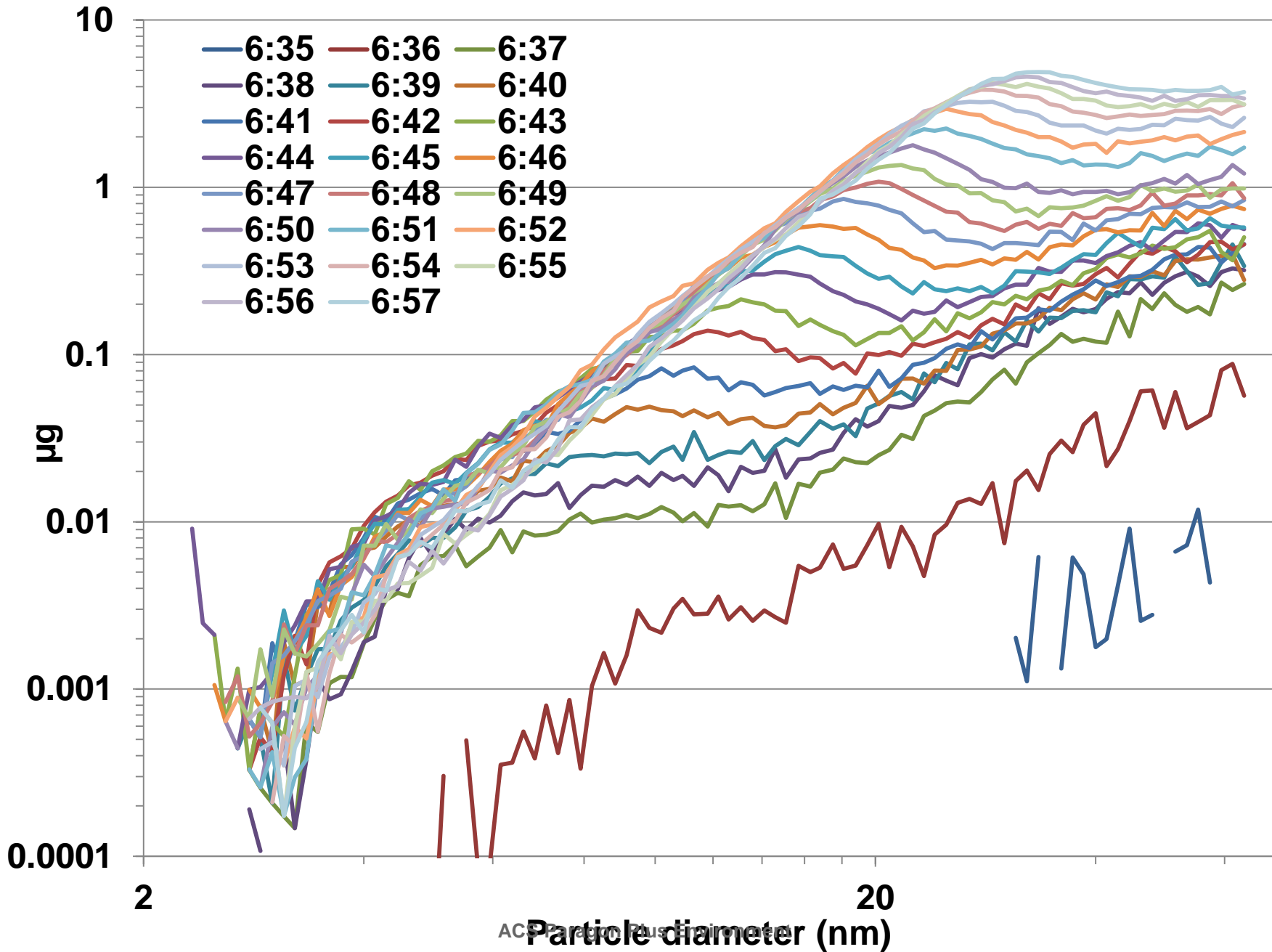
Citation (APA):
Wallace, L. A., Ott, W. R., Weschler, C. J., & Lai, A. C. K. (2017). Desorption of SVOCs from Heated Surfaces in the Form of Ultrafine Particles. *Environmental Science and Technology*, 51(3), 1140-1146.
<https://doi.org/10.1021/acs.est.6b03248>

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.



1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29

Desorption of SVOCs from heated surfaces in the form of ultrafine particles

Lance Wallace
US EPA (retired)
Affiliate, Lawrence Berkeley National Laboratory
Lwallace73@gmail.com

Wayne R. Ott
Stanford University
Dept. of Civil and Environmental Engineering

Charles J. Weschler
Environmental and Occupational Health Sciences Institute, Rutgers University, Piscataway, NJ
International Centre for Indoor Environment and Energy, Department of Civil Engineering, Technical
University of Denmark, Lyngby, Denmark

Alvin C.K. Lai
Department of Architecture and Civil Engineering
City University of Hong Kong

30

31 **Abstract.**

32 Ultrafine particles (UFP) produced by electric heating of stoves and metal cooking pans, absent food,
33 have been hypothesized to be created from a surface film of semivolatile organic compounds (SVOCs)
34 sorbed from the surrounding air. This study tests that hypothesis by size-resolved measurements
35 extending the lower range of the UFP studied from 10 nm to 2.3 nm, and including other surfaces (glass,
36 aluminum, and porcelain). Heating glass Petri dishes or squares of aluminum foil to about 350-400 °C for
37 4-6 minutes removed all sorbed organic substances completely. Subsequent exposure of these “clean”
38 Petri dishes and foil squares to indoor air in two different residences for successively longer periods (1
39 hour to 281 days), followed by heating the materials for 4-6 minutes, indicated a strong relationship of the
40 number, size distribution, and mass of the UFP to the time exposed. Estimates of the accumulation rate of
41 SVOCs on surfaces were similar to those in studies of organic film buildup on indoor windows. Transfer
42 of skin oils by touching the glass or foil surfaces, or after washing the glass surface with detergent and
43 bare hands, was also observed, with measured particle production comparable with that produced by long-
44 term exposure to indoor air.

45 **Introduction.**

46 UFP in homes are produced by gas and electric stoves¹⁻³, candles⁴, toasters and toaster ovens^{5,6}, steam
47 irons⁵, gas-powered clothes dryers^{7,8}, and power tools⁹. Several sources of the particles have been
48 identified. Combustion processes have been well studied as one source. Electric motors have been
49 identified as sources of copper particles¹⁰. However, for electric stoves, toasters, toaster ovens, and steam
50 irons, the source has not been definitively identified. In 2001, one research group suggested that for
51 electric stoves the source might be the heating elements themselves or else dust or other material
52 deposited on them¹.

53 A recent study found that repeated reheating of an electric burner, three types of pans, and a steam iron
54 with metal surface could eventually drive the particle production to zero, suggesting that the source was
55 not the metal surface itself, but material deposited on the surfaces¹¹. The authors also showed that the
56 particle production was renewed after exposure for some days or weeks to indoor air, indicating that some
57 substances in the air were collecting on the surfaces. They concluded that the source of the particles may
58 have been a sorbed film of organic material derived from SVOCs found indoors¹²⁻¹⁴. It has been
59 demonstrated that the equilibrium concentration of SVOCs in surface films can be estimated from an
60 SVOC's octanol/air partition coefficient (K_{oa}) and its gas phase concentration¹³. A simple calculation was
61 employed to estimate the mass of SVOCs that would be deposited on surfaces over time and was
62 consistent with the particle numbers observed. However, this previous study¹¹ employed a condensation
63 particle counter (CPC) sensitive only to particles > 10 nm and thus provided no indication of the number
64 or mass size distribution or of the relative contribution of particles < 10 nm to the total particle
65 production.

66 Therefore the present study was undertaken to extend these findings to different surfaces and to provide
67 full size distributions including particle diameters from 2.3-160 nm. Also a closer study of the washing
68 process was undertaken, including particularly a test of the possibility of contaminating the surfaces by
69 the use of bare hands during washing.

70 **Materials and Methods**

71 Three sites were included in the present study: private homes in Redwood City and Santa Rosa, CA and a
72 laboratory at Hong Kong City University. In the main room in the Santa Rosa home, (25.8 m³
73 volume)(roughly 3 X 4 X 2 m), the heating source was on the floor ~1 m away from the North and East
74 walls, with the aerosol intakes on a cart at a height ~1 m and placed ~1 m away from the center of the
75 West wall. The straight-line distance from source to intake was ~2 m. Two oscillating fans were placed,
76 one ~1 m high near the SE corner of the room, and one on the floor near the center of the room. The
77 Redwood City home used a 43 m³ room (roughly 4.5 X 3.2 X 3 m) with the aerosol intake of the Model
78 3007 placed 1.34 m from the heat source. A small electric fan was employed. In Hong Kong, selected
79 experiments were performed within a 30 m³ chamber with controlled air exchange rates and including
80 mixing fans.

81 A laboratory hot plate (Cimarec 1309HP15Q Digital Hot Plate, later replaced by Model HP88854100,
82 <http://www.mcqueenlabs.com/stir/cimarec-hp88854100.php> was employed at the Santa Rosa and
83 Redwood City sites to heat the Petri dishes, and at the Santa Rosa site only to heat the aluminum foil. For
84 early experiments at the Redwood City site, a Proctor-Silex 1000-watt electric burner was used to heat a
85 miniature stainless steel pan (DeBury) holding the Petri dishes. The Model 3007 CPC (TSI, Shoreview,
86 MN) used in our previous study¹ was also used at all 3 sites. An SMPS with nano-DMA and a water-
87 based CPC (SMPS Model 3936, CPC model 3786, TSI, Inc. Shoreview, MN) was employed at Hong
88 Kong, and an SMPS with nano-DMA and a water-based CPC (CPC model 3788) was used at Santa Rosa.
89 The Hong Kong SMPS was operated at 15 Lpm (sheath flow) and 1.5 Lpm (aerosol flow) providing a
90 range from 2.3-65 nm; the Santa Rosa SMPS was operated with these settings but also at 30 Lpm and 1.5
91 Lpm, giving a nominal range from 1.5-44 nm; at 6 Lpm and 0.6 Lpm, giving a range from 3.22 to 107
92 nm; and at 3 Lpm and 0.6 Lpm, giving a range from 4.6 to 160 nm. (Since all CPCs used have an
93 estimated 50% cutoff efficiency of about 2.5 nm, the actual range over all configurations was only from
94 2.33-160 nm).

95 The surfaces tested included aluminum foil (www.reynoldsconsumerproducts.com), the porcelain hot
96 plate mentioned above, and borosilicate (Pyrex) glass (55-mm and 100-mm Petri dishes,
97 <https://www.corning.com/>).

98 A K-type thermocouple-based temperature probe (Fluke 51 thermometer equipped with Fluke 80PK-22
99 Immersion Temperature Probe; Fluke Corporation, www.fluke.com) was used to measure the temperature
100 of the burner coils, the hot plate, and the metal and glass surfaces. Later a Fluke 54 II B with logging
101 capability for two data channels and automatic data storage was employed. Partway through the study, it
102 was found that the Fluke 80PK-22 probe was underestimating surface temperatures; it was therefore
103 replaced by two Thermoworks K-type surface probes (Models K-000 and K-020) with temperature ranges
104 to 600 °C and 1000 °C, respectively.

105 Sterile latex examination gloves (Dynarex) were employed for the washing experiments, along with
106 sponges, nylon brushes, brushes with natural bristles, dishwashing detergents (Palmolive), cleansers
107 (Barkeeper's Friend and Comet), and steel wool pads (SOS). The washing experiments focused on the
108 miniature metal cooking pans used in our previous study and were carried out at all three sites. The
109 related experiments involving aluminum foil pressed into these pans and touching the Petri dish surfaces
110 were performed only at the Santa Rosa site.

111 *Data analysis.* Aerosol Instrument Manager (AIM) proprietary software (TSI; AIM 10.0; later 10.2) was
112 used to determine size-resolved number concentrations. Because of the high diffusion coefficients of the
113 smaller UFPs, losses occur in the internal tubing of the classifier and the tubing connecting the classifier
114 to the CPC. Losses range from <10% at the 40-60 nm range up to >90% for particles <2.6 nm, according
115 to the manufacturer. Number concentrations were corrected for diffusion losses using Multi Instrument
116 Manager (v2.0; later 2.1) proprietary software (TSI). Temperature data from both the Fluke 51 and the
117 Thermoworks thermometers were obtained using Fluke proprietary software (Fluke View Forms,
118 v3.8.0003).

119 **Study design**

120 In the Santa Rosa home, a controlled experiment was conducted to estimate the rate of buildup of organic
121 film on a borosilicate surface. Four dozen 55-mm Petri dishes (with 60-mm covers) were employed. All
122 96 Petri dish covers and bottoms were heated for 4-6 minutes to temperatures exceeding 300 °C. They
123 were then reheated to the same temperatures (repeatedly if necessary) to assure that no particles were
124 produced. The “clean” Petri dishes were then arrayed (open sides up) on a clean surface consisting of
125 freshly-unrolled aluminum foil (Reynolds Wrap). Other samples of aluminum foil from interior
126 (unexposed) sections had been previously tested and found to produce no particles on initial heating. The
127 Petri dishes or small squares of the aluminum foil were then exposed to indoor air for varying lengths of
128 time (a few hours to 281 days). After exposure, the Petri dishes or aluminum foil squares were picked up
129 with tongs and collected dust was blown off. They were then heated on the Cimarec laboratory hot plate
130 for 4-6 minutes in the 25.8 m³ room with the floor fan on continually to promote good mixing. Total size-
131 resolved particle emissions and mass per unit surface area were then determined. Since the peaks were
132 achieved generally within 1-2 minutes after the end of the heating period, a correction for deposition and
133 air exchange during this short period was deemed unnecessary. To ensure that no organic material
134 remained on the Petri dish after heating, it was allowed to cool and then reheated to the same temperature.
135 In all cases, no or very few particles were produced, confirming that all material had been removed. An
136 example of this procedure is provided in Figure S1 in Supporting Information. A total of 177
137 experiments (155 with Petri dishes, 22 with aluminum foil) were performed.

138 In the Redwood City home, the same approach as above was employed except that the particle counter
139 was sensitive only to particles >10 nm. Early experiments used an electric burner (Proctor-Silex) to heat a
140 miniature stainless steel pan repeatedly until both burner and pan were producing no particles. Then Petri
141 dish covers or bottoms were heated in the pan as above following exposure to indoor air. Later
142 experiments employed the Cimarec laboratory hot plate to heat the Petri dishes. 44 experiments spanning
143 1 to 207 days exposure were performed.

144

145 **Results and Discussion**

146 *Quality Assurance.* In the Santa Rosa home, the SMPS and Model 3007 were run side by side during all
147 experiments. A comparison of the SMPS results for particles >10 nm agreed well with the Model 3007
148 results up to about 90,000 cm⁻³ (Figure S2 in Supporting Information). This is near the 100,000 cm⁻³
149 value claimed by the manufacturers and confirmed in an independent study¹⁵. At higher concentrations,

150 the Model 3007 suffers from particle obscuration in the sensing chamber, although correction
151 algorithms¹⁵ can be applied to increase the range of useful measurement to about 300,000 cm⁻³.

152 In the Redwood City home, two Model 3007s were employed. One was new and had been recently
153 calibrated by the manufacturer. The other was older and had not been recently calibrated. The two
154 instruments were run side by side and showed very high correlations, but the older instrument readings
155 averaged about 80% of those for the newer instrument. Therefore the experiments using the older
156 instrument were corrected by a factor of 1.25. Observed maximum concentrations exceeded 90,000 cm⁻³
157 on only two of 44 experiments and a correction using the algorithm¹⁵ would have been only a few percent
158 so was not attempted.

159 *Number and mass concentrations and total particle production.* Background number concentrations
160 indoors at Santa Rosa generally ranged from 100-3000 cm⁻³. Peak concentrations resulting from heating
161 the metal, glass or porcelain surfaces averaged 186000 (SE 6000) cm⁻³ for particles <10 nm, and 124000
162 (7000) cm⁻³ for particles >10 nm. The total particles produced were in the range of 10¹² to 10¹³, typically
163 over a 4-6-minute heating period. The total mass produced, assuming a density of 1 g cm⁻³, ranged from
164 <1 to 60 μg in the 25.8 m³ room. The longer a surface was exposed to indoor air, in general, the higher
165 the particle production when heated. The relationship may depend partly on intermittent activities such as
166 cooking or baking in the home that contribute to the SVOCs that eventually coat the surfaces, or on
167 temperature changes affecting the equilibrium between solid and gaseous phases, so a simple relationship
168 cannot be expected. Also, air exchange with the outdoors continuously brings in different SVOC
169 concentrations. However, assuming a roughly constant indoor gas-phase concentration for SVOCs, then a
170 newly “clean” surface might display a number or mass per unit area accumulation $y(t)$ approaching an
171 equilibrium concentration asymptote a with an exponential time constant τ , where t is the amount of time
172 exposed to indoor air:

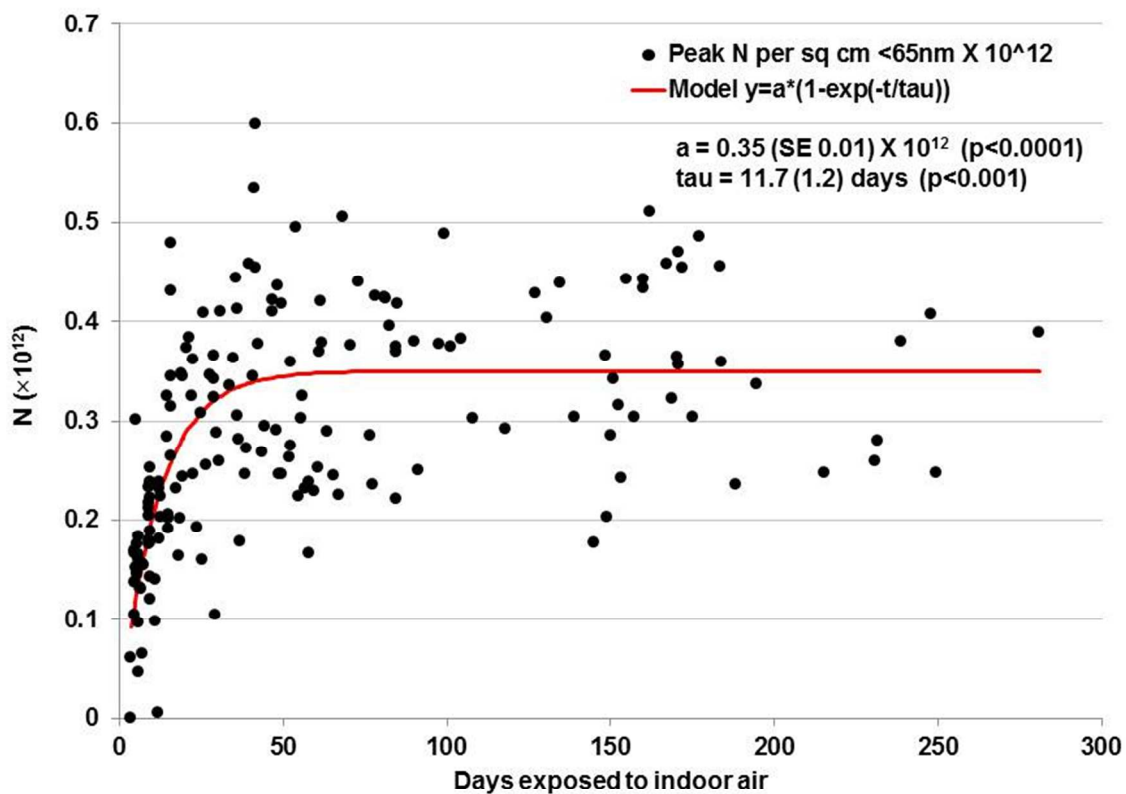
$$173 \quad y(t) = a(1 - e^{-t/\tau}) \quad (1)$$

174

175

176 At the Santa Rosa site, the characteristic time τ for particles <65 nm per cm² of heated surface was 11.7
177 (SE 1.2) days, with an estimated steady-state value of $0.35 (0.01) \times 10^{12}$ particles cm⁻² (Figure 1). Fitting
178 the two surface types separately gave estimates for the characteristic time of 11.0 (1.2) days for the Petri
179 dishes (N=155) and 13.5 (4.3) days for the aluminum foil (N=22). The estimated steady state values were
180 also similar at $0.36 (0.01) \times 10^{12}$ and $0.32 (0.03) \times 10^{12}$ particles cm⁻² for the Petri dishes and aluminum
181 foil, respectively.

182



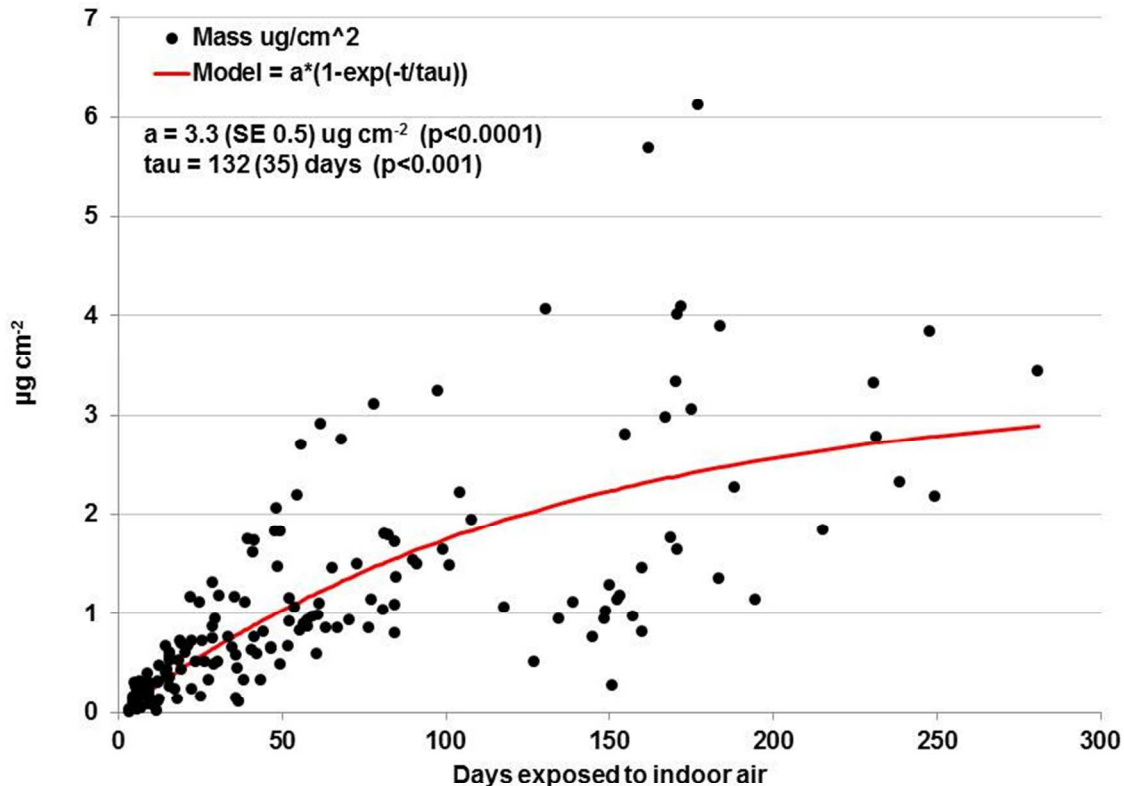
183

184

185 **Figure 1.** Number per cm² of particles released during 4-6-minute heating periods of Petri dishes
186 or aluminum foil exposed to room air for up to 281 days. Data from Santa Rosa site.

187

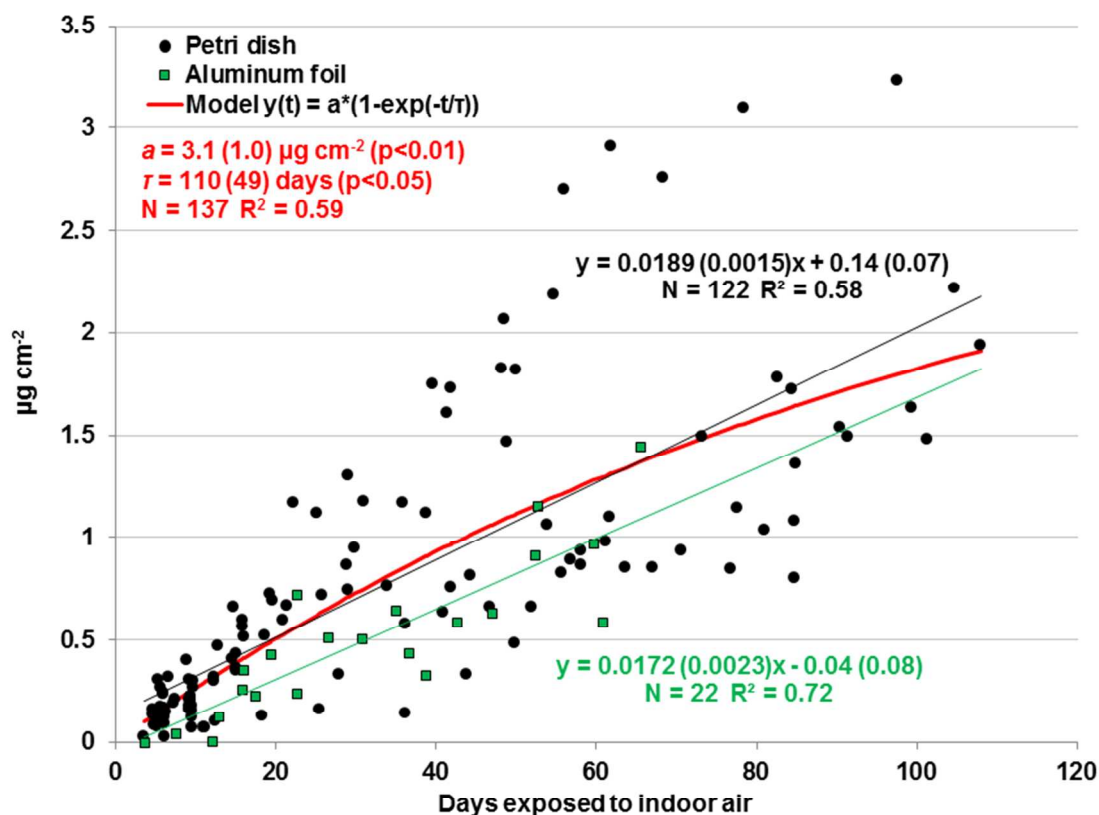
188 The model fit for the mass per unit surface area gave a much larger characteristic time of 132 (SE 35)
189 days (Figure 2).



190

191 **Figure 2. Mass per cm² surface of particles released during 4-6-minute heating periods of Petri**
192 **dishes or aluminum foil exposed to room air for up to 281 days. Data from Santa Rosa site.**

193 To estimate the rate of accumulation of mass, we can look at the period of the first 108 days when the
194 increase was nearly linear (Figure 3). The data for the Petri dishes and aluminum foil were analyzed
195 separately (black and green points and lines). The slopes of the lines, a measure of the rate of mass
196 buildup, were similar for the aluminum foil and Petri dishes, at about 0.017 and 0.019 µg cm⁻² per day,
197 respectively.



198

199

200 **Figure 3.** Mass per cm^2 surface of particles released during 4-6-minute heating periods of Petri
 201 dishes or aluminum foil exposed to room air for up to 108 days. A 2-parameter fit (red curve) is
 202 performed on the full data set and is only barely significant. An ordinary least squares regression
 203 is performed on each of the two types of surfaces and is highly significant, with quite similar slopes
 204 (black and green lines). Data from Santa Rosa site.

205

206 The particle production and associated mass released for four size categories in each of these 177
 207 experiments are provided in Table S1 (Excel file in Supporting Information). A summary of the
 208 experiments is provided in Tables S2 (Santa Rosa) and S3 (Redwood City).

209 With increasing time of exposure, the mass distribution shifted toward larger particles (Figures S3 and
 210 S4), whereas the number distribution held steady with a mode between 3 and 10 nm (Figure S5).
 211 Although the number mode does not change over time, due to the preponderance of particles < 10 nm in
 212 diameter, the fraction of particles in this size category does decrease sharply over time, dropping from 60-
 213 90% of all particles in the first few weeks to 50-65% from about 60-280 days exposed to indoor air
 214 (Figure S6).

215 An obvious question is why the particle number concentration (Figure 1) reaches steady-state so much
216 faster than the particle mass concentration (Figure 2). At present, we cannot answer this question
217 definitively, but suggest possible explanations that might be probed in future studies. When the exposure
218 time has been only a few days, the number of particles is small, and there may be insufficient surface area
219 on pre-existing particles to accommodate condensation of all the supersaturated vapors; nucleation is
220 required to produce new particles for supersaturated vapors to condense into. As the exposure time
221 increases, and we know from Figure S5 that more large particles are produced, there is more surface area
222 available for condensation, and nucleation of new particles becomes decreasingly necessary to
223 accommodate condensation of the supersaturated vapors. Another potential limiter to particle count
224 concentration is coagulation. For low exposure duration, most particles are small and coagulation is
225 limited. As the exposure time increases, larger particles become available for scavenging the smaller
226 particles, and they do so with far greater efficiency according to the Fuchs correction¹⁶. At high
227 coagulation rates, the lifetime of freshly formed particles is quite short, limiting their contribution to the
228 total particle count. The number counts (usually < 100,000 particles/cm³) are lower than levels at which
229 coagulation would be expected to occur¹⁶. However, these concentrations are measured in the room after
230 thorough mixing. The actual count concentration in the plume where nucleation occurs is likely much
231 higher.

232 The absorption of thermally desorbed mass to existing and freshly nucleated particles is an ongoing
233 process. Regarding the mass mode shift to larger particles with additional exposure time for the Petri
234 dishes and foils, we suggest that once the critical point has been reached where there is sufficient
235 particulate surface area to accommodate condensation of all the supersaturated vapors, additional material
236 that has sorbed to the Petri dish or foil surfaces, when thermally desorbed, condenses on the now
237 sufficient particulate surface area. This continues particle growth and the shift of the mass mode to larger
238 and larger particles.

239 The observation that the particle mass concentration does not attain steady-state over a period of 108 days
240 (Figure 3) relates to the rate at which organics accumulate on the Petri dish or foil surfaces. We anticipate
241 that absorption of organics in the surface film will continue to occur as long as there are SVOCs in the
242 room air that are not in equilibrium with their levels in the film. Organics that have larger K_{oa} values
243 require more time to equilibrate than organics with smaller K_{oa} ^{12,17-19}. The time required to reach an
244 overall steady-state will be determined by the time required for less volatile SVOCs to equilibrate. We
245 can roughly estimate such a time using equation 4.1 in Weschler and Nazaroff¹², which states that the
246 time scale for an individual organic to achieve equilibrium absorption, τ , is given by $K_{oa} * X / v_d$, where K_{oa}
247 is its octanol-air partition coefficient, X is the equivalent thickness of the organic portion of the film, and
248 v_d is the mass transfer coefficient from room air to the film. Table S4 provides a range of values for these
249 three parameters. Both the residence time τ and $3 \times \tau$ (the time to reach 99% of the final steady state
250 concentration) are calculated. Thicknesses for the organic portion of films have been estimated on
251 different indoor surfaces and are in the range of 5 to 30 nm^{13,18,20}. A reasonable range for v_d , for indoor
252 SVOCs, indoor surfaces and typical near surface-airflows is from 2.5 to 4 m/h¹². The biggest uncertainty
253 is the value to use for K_{oa} . Above a certain K_{oa} , the gas-phase concentration of an SVOC is so low that its
254 contribution to film growth is negligible²¹. Based on reported correlations between gas-phase
255 concentrations and surface film concentrations for certain high K_{oa} SVOCs^{13,19}, we infer that the “critical
256 log K_{oa} ” likely lies in the range of 11 to 13. To be more specific, it would take between 400 and 600 days
257 for a gas-phase SVOC with a log K_{oa} of 12 to reach steady-state partitioning with a 10 to 15 nm thick

258 organic film assuming a v_d of 3 m/h. This estimation is consistent with the observation that it takes much
259 longer for the particle mass concentration (Figure 2) to reach steady-state than the particle number
260 concentration (Figure 1).

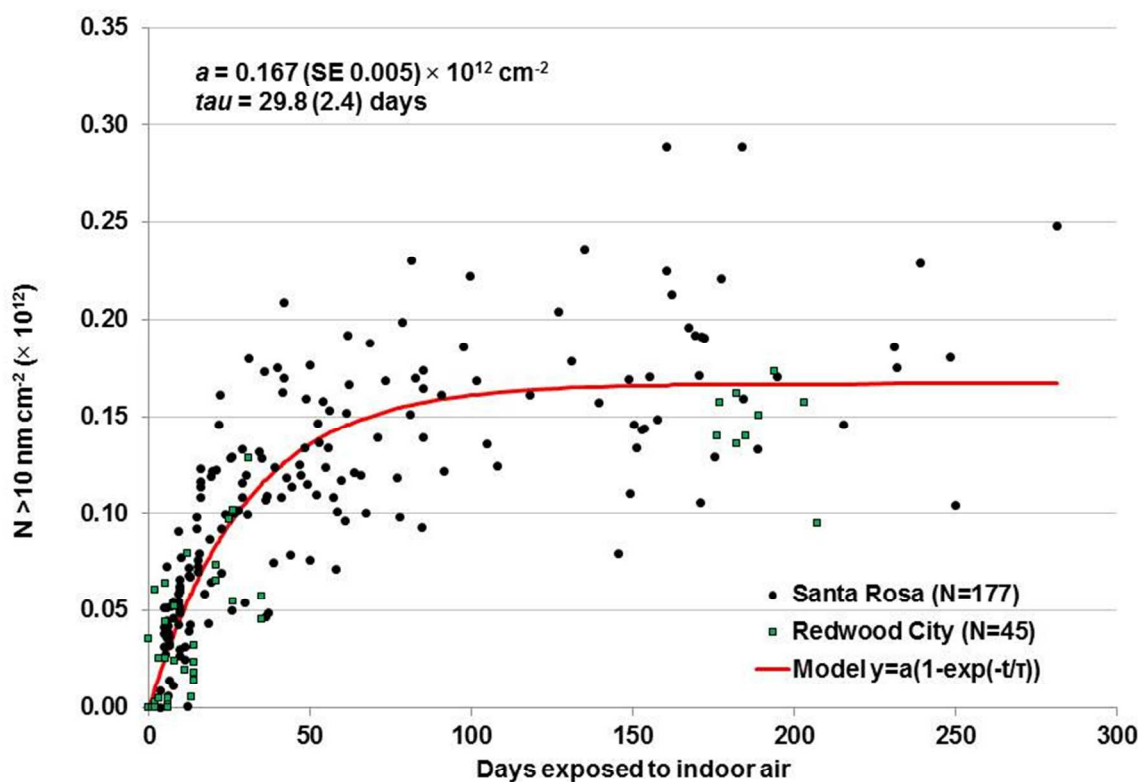
261 In some experiments, the temperature was ramped up slowly to determine at what temperature particles
262 first appeared. In general, no particles were observed at temperatures below 150 °C. Most particles were
263 generated between 200 and 250 °C, although in some experiments particle generation was observed at
264 temperatures above 300 °C (Figure S7).

265 There is remarkably good agreement between the rate at which organic mass accumulates on Petri dishes
266 or foils, based on the measurements in Figure 3, and the rate at which organic mass accumulates on
267 indoor windows based on measurements reported in several studies. For example, Figure 1 of Huo et al.¹⁸
268 shows total accumulation of film mass on freshly cleaned windows over a 49-day period in winter and a
269 77-day period in summer for two urban buildings in northeastern China. The total (organic and inorganic)
270 accumulation rate is roughly 0.034 to 0.036 $\mu\text{g cm}^{-2}$ per day. If we use the author's assumption that 50%
271 of the film is organic material, this corresponds to growth rates for the organic portion of the film of 0.017
272 to 0.018 $\mu\text{g cm}^{-2}$ per day compared to our measured growth of about 0.017 and 0.019 $\mu\text{g cm}^{-2}$ per day for
273 the aluminum foil and Petri dishes, respectively. Considering the many differences in SVOC sources and
274 environmental parameters between the two studies, we acknowledge that this agreement exceeds the
275 accuracy of the measurements. Li et al.²² report growth rates for the organic portion of window films of
276 0.008 to 0.063 $\mu\text{g cm}^{-2}$ per day for north facing windows and 0.005 to 0.076 $\mu\text{g cm}^{-2}$ per day for north
277 facing windows for a building in Guangzhou, China. A final example comes from one of the first studies
278 to measure growth of indoor window films²⁰. The films listed in Table 1 of that paper were reported to
279 have been measured 3 to 5 months after cleaning the windows from targeted buildings in greater Toronto;
280 calculated growth rates for the organic portion of the indoor films are 0.003 to 0.017 $\mu\text{g cm}^{-2}$ per day.

281 We judge that the major source of organics in both the window films and the surface of the Petri dishes
282 and foils is sorption from the gas-phase. The rate at which organic films grow on indoor windows, Petri
283 dishes and foils precludes a dominant contribution from the deposition of airborne particles. Particles
284 smaller than 2.5 μm diameter are predicted to deposit at rates two to three orders of magnitude slower
285 than the measured rates (see calculations in Supporting Information). Particles larger than 2.5 μm are not
286 expected to soil vertical window surfaces and are anticipated to have very low concentrations in the room
287 used for the measurements reported here (due to the filters used in the home's air distribution system). A
288 possible contributor to the organic material collecting on the Petri dish surface is skin flakes or clothing
289 particles from the experimenters. We have confirmed by direct experiment that skin flakes can contribute
290 to the particle number and mass, presumably due to thermal desorption of skin oils contained in the skin
291 cells (Figures S8 and S9). However, we calculate in the SI that the mass from this source is a small
292 fraction of that from room air. Since the dust was blown off the Petri dishes before heating, we expect that
293 most of the skin flakes or clothing fibers were removed. However, for situations in which dust is not
294 removed, as in the use of cooking pans, it is possible that the organic material contained in the dust could
295 contribute substantially to the total airborne particle number or mass. We plan to investigate this
296 possibility in future work.

297 *Comparison with Redwood City site.* The 44 experiments using Petri dishes at Redwood City employed
298 only the Model 3007 particle counter sensitive to particles >10 nm. To compare with these observations,

299 only particles >10 nm as measured by the Santa Rosa SMPS were selected. The model parameters were
300 very similar in the two homes, despite the many likely differences in SVOC sources and concentrations,
301 temperature, relative humidity, etc. The asymptotic particle production levels per square cm of surface
302 area were $0.168 (0.005) \times 10^{12}$ and $0.148 (0.009) \times 10^{12}$ in Santa Rosa and Redwood City, respectively.
303 The characteristic times were 27.8 (2.6) days and 42.2 (7.3) days, respectively. Neither the asymptotic
304 values nor the characteristic times were significantly different. The combined data (N=221) and overall
305 model fit are provided in Figure 4.



306

307 **Figure 4. Particles (>10 nm) produced per square cm of surface area at Santa Rosa and Redwood**
308 **City sites.**

309

310 *Contamination of surfaces by transfer of skin oil.* As reported in our previous paper¹¹, “clean” pans (zero
311 particle production) washed with detergent produced large numbers of particles on being heated. Those
312 experiments and many of those described below employed the Model 3007 instrument with a lower size
313 cutoff of 10 nm. After May of 2015, the skin oil experiments employed the SMPS with a lower size
314 cutoff of 2.33 nm. Although at first we suspected the detergent might be the source of the particles, it was
315 also possible that using our bare hands in the washing process may have contaminated the pans with skin
316 oils even though the pans had been rinsed thoroughly. Therefore, we tried washing pans using sterile

317 gloves. In 9 experiments washing the cast iron pans with detergent or other cleansers while wearing the
 318 gloves, 4 cases produced no particles at all, and the mean (SD) was only $4.5 (5.2) \times 10^{10}$. By contrast, 29
 319 experiments washing the cast iron pans with bare hands resulted in a mean (SD) particle production of 3.5
 320 $(3.0) \times 10^{12}$, about 80 times as many. We conclude that transfer of skin oils while washing is the likely
 321 cause of the increased particle production following washing.

322 Aluminum foil is often used to line baking or roasting pans. Fish or poultry skin that would normally
 323 stick to the pans sticks instead to the foil, which can easily be disposed of, leaving a pan that is easy to
 324 clean. In 16 experiments, we used bare hands to tamp the foil down inside the miniature cast iron pans
 325 and then heated the pans. In another 8 experiments, we also used sterile gloves to tear off the foil and
 326 tamp it down in the pans before heating. In three more experiments, the experimenter washed his hands
 327 and sprayed them with isopropanol before tamping down the foil. The mean (SD) particle production
 328 after using bare hands to press down the foil and heating to temperatures ranging from 207-226 °C was
 329 $1.1 (1.2) \times 10^{12}$, compared to a mean (SD) of $3.8 (9.0) \times 10^{11}$ after heating to temperatures of 207-223 °C
 330 for the 11 experiments with either the sterile gloves or the clean hands. One of the experiments using the
 331 sterile gloves resulted in a particle production of 3.0×10^{12} , and was probably a case of contamination.
 332 The median value for these 11 experiments was not different from background levels, and the mean (SD)
 333 dropping the outlier was $1.1 (1.3) \times 10^{11}$, a factor of 10 smaller than the mean of the experiments using
 334 bare hands.

335 We experimented with touching the cast iron pan directly. In four experiments, the experimenter touched
 336 the pan in a single spot on the bottom interior surface just after taking a shower. In all four experiments,
 337 no particles were observed after heating the pan to temperatures ranging from 193-202 °C. In 7
 338 experiments when no shower was taken, the pan was again touched by a thumb or finger; the mean
 339 particle production after heating to 145-202 °C was $2.0 (0.9) \times 10^{12}$.

340 The results of 76 experiments addressing these three modes of possible contamination are summarized
 341 below (Table 1). The range of the mean number of particles produced for the three modes of skin oil
 342 transfer (1.1 - 3.5×10^{12}) is comparable to the mean number of particles produced by 177 experiments with
 343 the Petri dishes and aluminum foil: $8.0 (0.3) \times 10^{12}$ (Table S2).

| Table 1. Contamination of surfaces by touching with bare hands compared to use of sterile gloves: Particles produced ($\times 10^{12}$) | | | | | | |
|---|--------------------|----------------|----------------------------|-------------------------------|------------------------------|-------------------------|
| | Washing pan | | Placing foil on pan | | Touching pan directly | |
| | Bare hands | Sterile gloves | Bare hands | Sterile gloves or clean hands | Bare hands | Bare hands after shower |
| N | 29 | 9 | 16 | 11 | 7 | 4 |
| Mean | 3.50 | 0.04 | 1.14 | 0.38 | 1.98 | 0.00 |
| SE | 0.57 | 0.02 | 0.30 | 0.27 | 0.35 | 0.00 |

344
 345 The concentrations observed after 4-6 minutes of heating in a room of volume 25.8 m^3 are in the range of
 346 10^5 to 10^6 cm^{-3} , far higher than most values observed in ambient air even near high-traffic-density
 347 roadways. Although the smallest particles disappear within minutes, the remainder of the particles can
 348 remain at elevated concentrations for several hours. Therefore, we conclude that cooking and other
 349 operations involving heating surfaces to temperatures $> 150 \text{ }^\circ\text{C}$ may have a substantial impact on total

350 exposure to ultrafine particles. If the hypothesis is confirmed that most of these particles derive from
351 SVOCs deposited on the surfaces, the findings suggest a newly appreciated pathway of exposure to
352 SVOCs—inhale of SVOCs liberated from heated surfaces. It appears that desorption of the material
353 on surfaces that have previously been exposed to indoor air releases material from these surfaces, causing
354 large quantities of airborne ultrafine particles to be produced.

355 Finally, we raise the possibility that a substantial portion of the UFP observed during cooking may arise
356 from the release of SVOCs on the cooking surface, rather than arising from the cooking oil or food itself.
357 Further study is indicated to determine whether this is indeed an important contributor to UFP observed
358 during cooking.

359 References

- 360
361 1. Dennekamp, M.; Howarth, S.; Dick, C. A. J.; Cherrie, J. W.; Donaldson, K.; Seaton, A. Ultrafine
362 particles and nitrogen oxides generated by gas and electric cooking. *Occup. Environ. Med.* **2001**, *58*, 511–
363 516.
- 364
365 2. Wallace, L.A.; Emmerich, S.J.; Howard-Reed, C. Source strengths of ultrafine and fine particles due to
366 cooking with a gas stove. *Environ. Sci. Tech.* **2004**, *38*, 2304-2311.
- 367
368 3. Wallace, L.A.; Wang, F.; Howard-Reed, C.; Persily, A. Contribution of gas and electric stoves to
369 residential ultrafine particle concentrations between 2 nm and 64 nm: size distributions and emission and
370 coagulation rates. *Environ. Sci. Technol.* **2008**, *42*, 8641-8647.
- 371
372 4. Bekö, G.; Weschler, C.J.; Wierzbicka, A.; Karotki, D.G.; Toftum, J.; Loft, S.; Clausen, G. (2013)
373 Ultrafine particles exposure and source apportionment in 56 Danish homes. *Environ. Sci. Technol.* **2013**,
374 *47*, 10240-10248.
- 375
376 5. Schripp, T.; Kirsch, I.; Salthammer, T. Characterization of particle emission from household electrical
377 appliances. *Sci. Total Environ.* **2011**, *409*, 2534-2540.
- 378
379 6. Wallace, L.A.; Ott, W.R. Personal exposure to ultrafine particles. *J. Expo. Sci. Env. Epid.* **2010**, *21*,
380 20-30.
- 381
382 7. Wallace, L.A. Ultrafine particles from a vented gas clothes dryer. *Atmos. Environ.* **2005**, *39*, 5777-
383 5786.
- 384
385 8. Bhangar, S.; Mullen, N.A.; Hering, S.V.; Kreisberg, N.M.; Nazaroff, W.W. Ultrafine particle
386 concentrations and exposures in seven residences in northern California. *Indoor Air* **2011**, *21*, 132-144.
- 387
388 9. Rim, D.; Green, M.; Wallace, L.A.; Persily, A.K.; Choi, J. Evolution of ultrafine particle size
389 distributions following indoor episodic releases: relative importance of coagulation, deposition and
390 ventilation. *Aerosol Sci. Tech.* **2012**, *46* (5), 494-503.
- 391
392 10. Szymczak, W.; Menzel, N.; Keck, L. Emission of ultrafine copper particles by universal motors
393 controlled by phase angle. *J. Aerosol Sci.* **2007**, *38*(5), 520–531.

- 390 11. Wallace, L.A.; Ott, W.R.; Weschler, C.J. Ultrafine particles from electric appliances and cooking
391 pans: experiments suggesting desorption/nucleation of sorbed organics as the primary source. *Indoor Air*
392 **2015**, *25*, 536-546.
- 393
- 394 12. Weschler, C.J.; Nazaroff, W.W. Semivolatile organic compounds in indoor environments. *Atmos.*
395 *Environ.* **2008**, *42*, 9018-9040.
- 396
- 397 13. Weschler, C.J.; Nazaroff, W.W. SVOC exposure indoors: fresh look at dermal pathways. *Indoor Air*
398 **2012**, *22*, 356-377.
- 399
- 400 14. Weschler, C.J. Indoor/outdoor connections exemplified by processes that depend on an organic
401 compound's saturation vapor pressure, *Atmos. Environ.* **2003**, *37*, 5455-5465.
- 402
- 403 15. Hämeri, K.; Koponen, I.K.; Aalto, P.P.; Kulmala, M. The particle detection efficiency of the TSI-3007
condensation particle counter, *J. Aerosol Sci.* **2002**, *33*, 1463-1469.
- 404
- 405 16. Hinds, W. C., *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles*, 2nd
Edition, Wiley, 1999.
- 406
- 407 17. Pan, S.-H.; Li, J.; Lin, T.; Zhang, G.; Li, X.-D.; Yin, H., Polycyclic aromatic hydrocarbons on
408 indoor/outdoor glass window surfaces in Guangzhou and Hong Kong, south China. *Environmental*
Pollution **2012**, *169*, 190-195.
- 409
- 410 18. Huo, C.-Y.; Liu, L.-Y.; Zhang, Z.-F.; Ma, W.-L.; Song, W.-W.; Li, H.-L.; Li, W.-L.; Kannan, K.; Wu,
411 Y.-K.; Han, Y.-M.; Peng, Z.-X.; Li, Y.-F., Phthalate Esters in Indoor Window Films in a Northeastern
412 Chinese Urban Center: Film Growth and Implications for Human Exposure. *Environ. Sci. Tech.* **2016**, *50*,
(14), 7743-7751.
- 413
- 414 19. Venier, M.; Audy, O.; Vojta, Š.; Bečanová, J.; Romanak, K.; Melymuk, L.; Krátká, M.; Kukučka, P.;
415 Okeme, J.; Saini, A.; Diamond, M. L.; Klánová, J., Brominated flame retardants in the indoor
416 environment — Comparative study of indoor contamination from three countries. *Environment*
International **2016**, *94*, 150-160.
- 417
- 418 20. Liu, Q. T.; Chen, R.; McCarry, B. E.; Diamond, M. L.; Bahavar, B., Characterization of polar organic
419 compounds in the organic film on indoor and outdoor glass windows. *Environ. Sci. Technol.* **2003**, *37*,
(11), 2340-2349.
- 420
- 421 21. Pankow, J. F., An absorption model of gas/particle partitioning of organic compounds in the
atmosphere. *Atmos. Environ.* **1994**, *28*, (2), 185-188.
- 422
- 423 22. Li, J.; Lin, T.; Pan, S.-H.; Xu, Y.; Liu, X.; Zhang, C.; Li, X.-D., Carbonaceous matter and PBDEs on
424 indoor/outdoor glass window surfaces in Guangzhou and Hong Kong, South China. *Atmos. Environ.*
2010, *44*, (27), 3254-3260.
- 425
- 426 **Acknowledgements.** Peter DeCarlo provided helpful comments regarding the number of particles
427 produced by heating surfaces exposed for different time periods. Bill Nazaroff raised the possibility of
428 skin flakes and particles from clothing adding to the SVOC loading on the Petri dishes.