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

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

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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 ^{o}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 stoves[1-3], candles[4], toasters and toaster ovens[5,6], steam
47   irons[5], gas-powered clothes dryers[7,8], and power tools[9]. 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 particles[10]. 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 them[1].

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 surfaces[11]. 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 indoors[12-14]. 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 concentration[13]. 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 study[11] 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 m^3
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 m^3 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 m^3 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 study[1] 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 ^{o}C and 1000 ^{o}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.

3

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 $^{\circ}$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 m^3 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 study[15]. At higher concentrations,

150 the Model 3007 suffers from particle obscuration in the sensing chamber, although correction

151 algorithms[15] 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 algorithm[15] 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 10^{12} to 10^{13}, 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 m^3 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 $$y(t) = a(1-e^{(-t/\tau)}) \tag{1}$$

174

175

176 At the Santa Rosa site, the characteristic time τ for particles <65 nm per cm^2 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
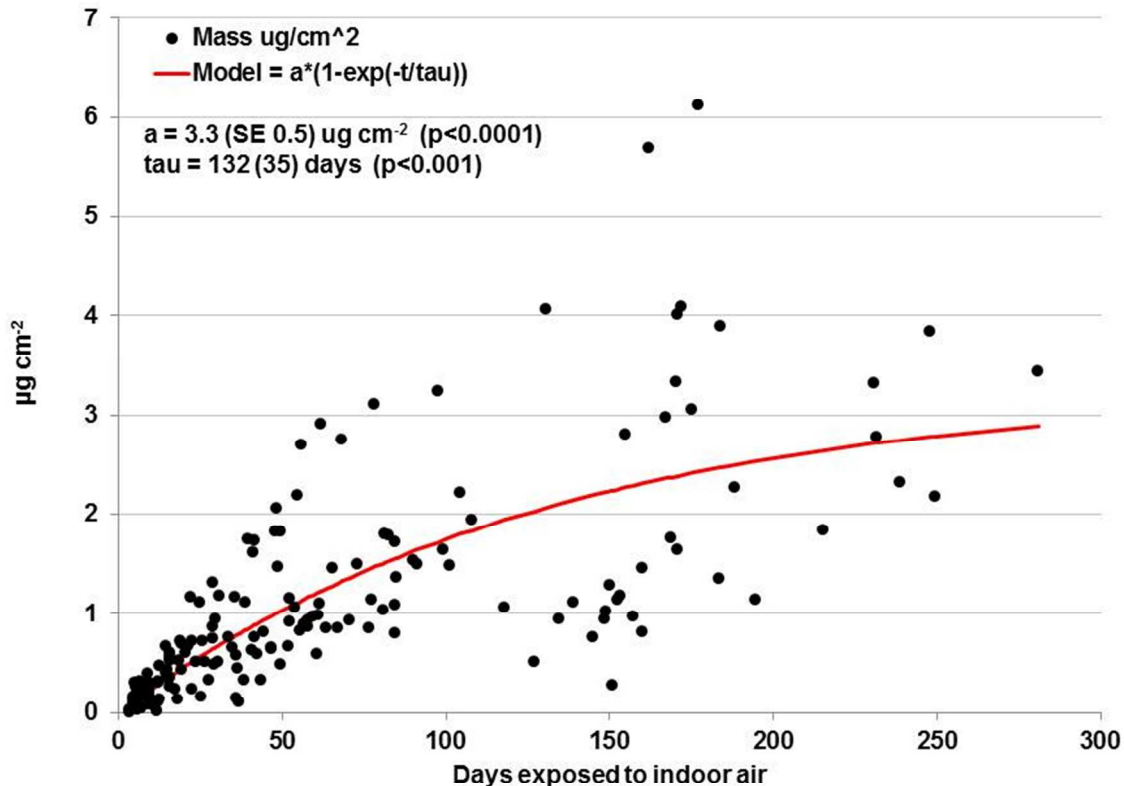
181 foil, respectively.

182
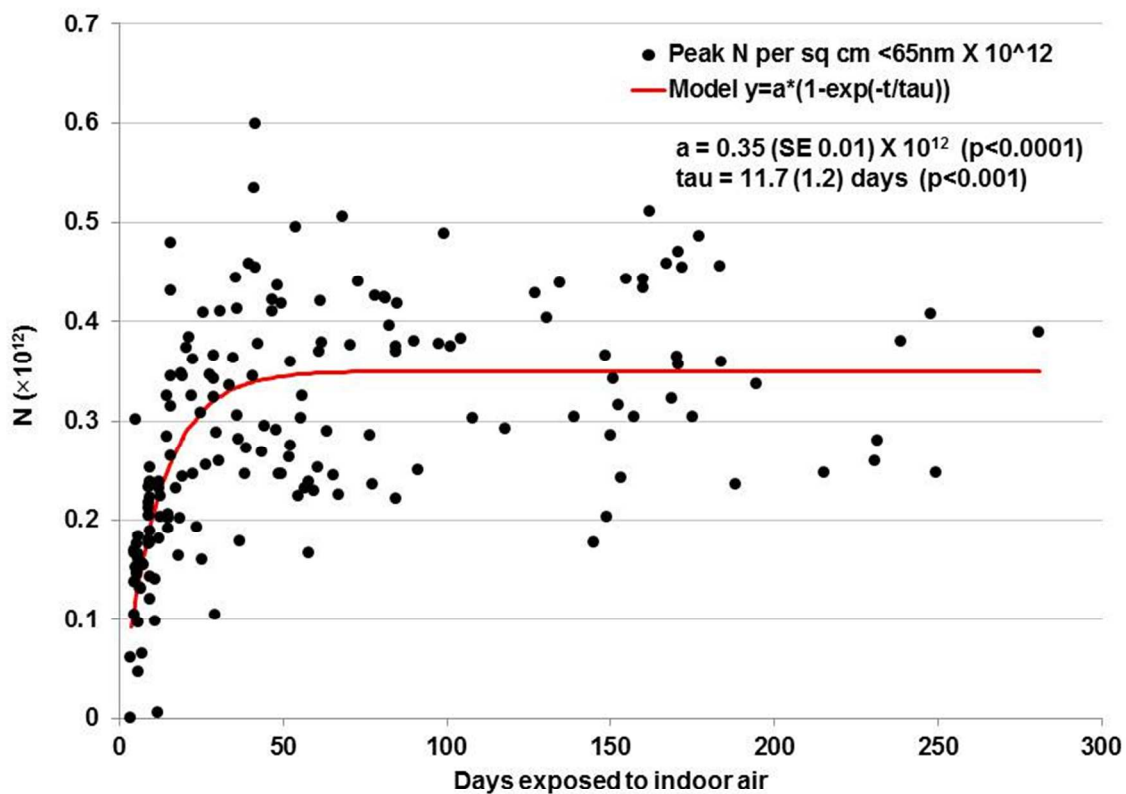
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184

185   **Figure 1. Number per cm^2 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

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

To estimate the rate of accumulation of mass, we can look at the period of the first 108 days when the increase was nearly linear (Figure 3). The data for the Petri dishes and aluminum foil were analyzed separately (black and green points and lines). The slopes of the lines, a measure of the rate of mass buildup, were similar for the aluminum foil and Petri dishes, at about 0.017 and 0.019 µg cm^{-2} per day, respectively.
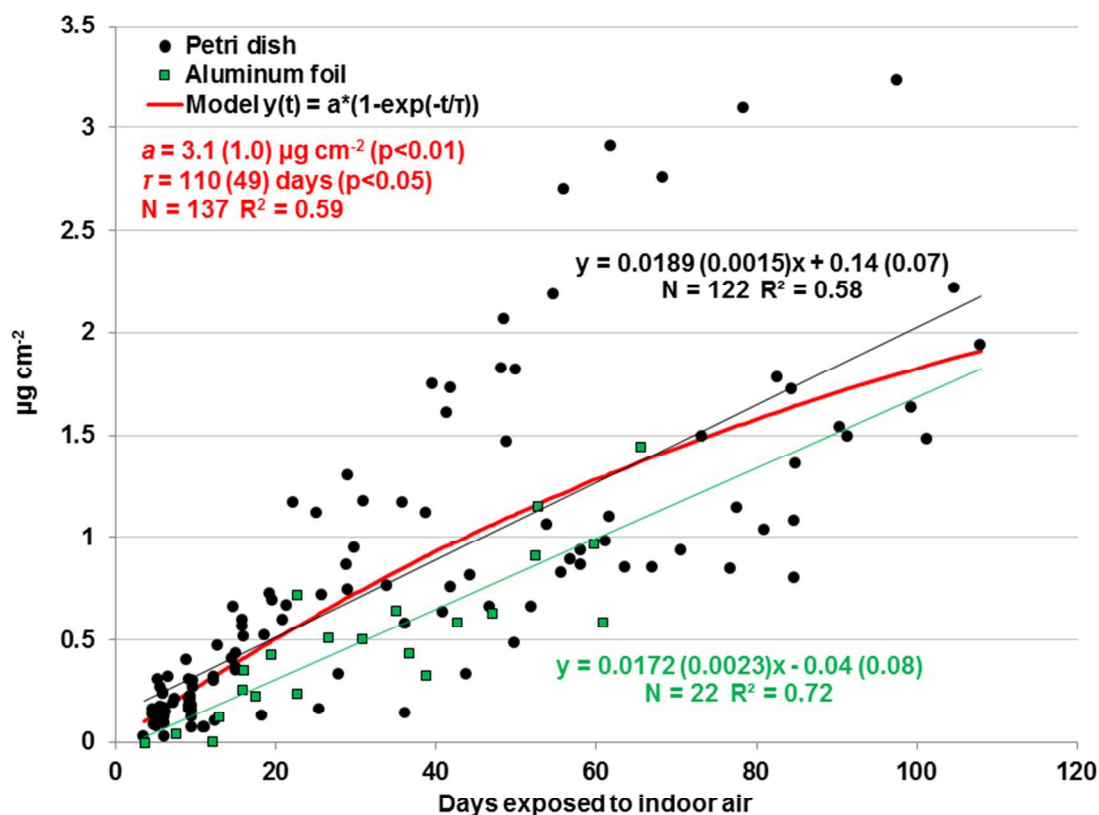
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198

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).

8

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 correction[16]. 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/cm^3) are lower than levels at which
229   coagulation would be expected to occur[16]. 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 Nazaroff[12], 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/h[12]. The biggest uncertainty
253   is the value to use for K_{oa}. Above a certain Koa, the gas-phase concentration of an SVOC is so low that its
254   contribution to film growth is negligible[21]. 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 µg 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 µg cm^{-2} per day compared to our measured growth of about 0.017 and 0.019 µg 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 µg cm^{-2} per day for north facing windows and 0.005 to 0.076 µg cm^{-2} per day for north
277   facing windows for a building in Guangahou, China. A final example comes from one of the first studies
278   to measure growth of indoor window films[20]. 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 µg 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.

a = 0.167 (SE 0.005) $\times 10^{12}$ cm^{-2}
tau = 29.8 (2.4) days
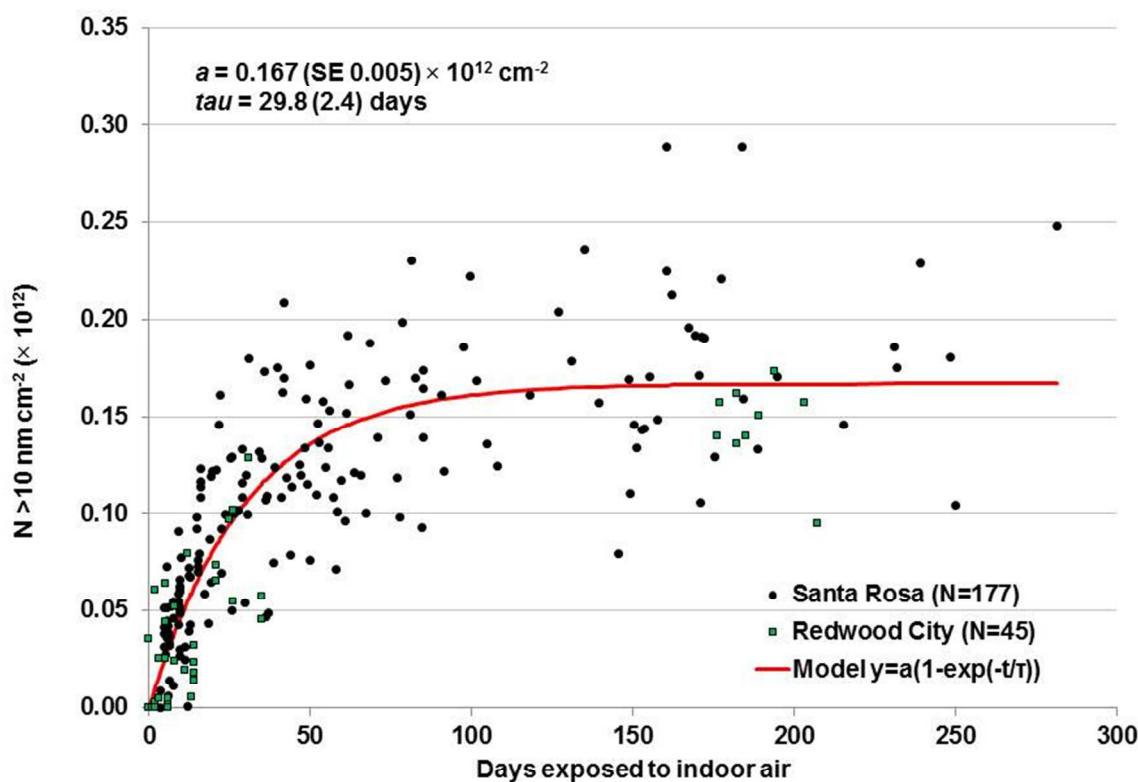
306

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 paper[11], "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

11

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) × 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) × 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 ^{o}C was
329 1.1 (1.2) × 10^{12}, compared to a mean (SD) of 3.8 (9.0) × 10^{11} after heating to temperatures of 207-223 ^{o}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) × 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 ^{o}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 ^{o}C was 2.0 (0.9) × 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) × 10^{12} (Table S2).

Table 1. Contamination of surfaces by touching with bare hands compared to use of sterile gloves: Particles produced (× 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

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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 ^{o}C may have a substantial impact on total

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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—inhalation 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.

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