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# 1 Surface emissions modulate indoor SVOC concentrations through 2 volatility-dependent partitioning

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## 19 20 **0 ABSTRACT**

21 Measurements by semivolatile thermal desorption aerosol gas chromatography (SV-TAG) were  
22 used to investigate how semivolatile organic compounds (SVOCs) partition among indoor  
23 reservoirs in (1) a manufactured test house under controlled conditions (HOMEChem campaign)  
24 and (2) a single-family residence when vacant (H2 campaign). Data for phthalate diesters and  
25 siloxanes suggest that volatility-dependent partitioning processes modulate airborne SVOC  
26 concentrations through interactions with surface-laden condensed-phase reservoirs. Airborne  
27 concentrations of SVOCs with vapor pressures in the range of C13 to C23 alkanes correlated  
28 with indoor air temperature. Observed temperature dependencies were quantitatively similar to  
29 theoretical predictions that assumed a surface-air boundary layer with equilibrium partitioning  
30 maintained at the air-surface interface. Airborne concentrations of SVOCs with vapor pressures  
31 corresponding to C25 to C31 alkanes correlated with airborne particle mass concentration. For  
32 SVOCs with higher vapor pressures, which are expected to be predominantly gaseous,  
33 correlations with particle mass concentration were weak or nonexistent. During primary particle  
34 emission events, enhanced gas-phase emissions from condensed-phase reservoirs partition to  
35 airborne particles, contributing substantially to organic particulate matter. An emission event  
36 related to oven-usage was inferred to deposit siloxanes in condensed-phase reservoirs throughout  
37 the house, leading to the possibility of reemission during subsequent periods with high particle  
38 loading.

## 39 40 **1 INTRODUCTION**

41 In indoor environments, semivolatile organic compounds (SVOC) dynamically partition between  
42 the gas phase and various condensed-phase reservoirs, such as airborne particles, surface films,  
43 settled dust, and building materials.<sup>1</sup> Many specific indoor SVOCs are of concern for human  
44 health, such as endocrine-disrupting phthalate diesters and halogenated flame retardants.<sup>2</sup>  
45 Airborne particles are often partly composed of SVOCs. Exposure to particulate matter is among  
46 the leading global mortality risk factors,<sup>3</sup> although the specific roles of SVOCs and the relative  
47 importance of indoor particle exposure contributing to this risk are unknown. Understanding the

48 dynamics and physical behavior of indoor airborne SVOC concentrations is important for risk  
49 assessment and exposure mitigation.

50  
51 Emerging evidence indicates that interactions between indoor air and condensed-phase reservoirs  
52 influence airborne SVOC concentrations. Surface reservoirs are defined as “condensed-phase  
53 materials containing chemical constituents that undergo exchange with the gas phase.”<sup>4</sup> Recent  
54 field measurements of volatile organic compounds (VOCs) and SVOCs in real indoor settings  
55 suggest that organics readily desorb from condensed-phase reservoirs into bulk air. The high rate  
56 of desorption suggests that, for many compounds, the condensed-phase reservoirs are either  
57 interior surface films or thin layers of building materials closely in contact with indoor air.<sup>4-8</sup>  
58 Weschler and Nazaroff described a model for the growth of SVOC-laden organic surface films  
59 indoors and suggested that surface films may provide functional and chemical homogeneity  
60 among the diverse surfaces interacting with bulk indoor air.<sup>9</sup>

61  
62 Models describing indoor surface emissions have assumed the existence of a boundary layer  
63 immediately adjacent to surfaces, with equilibrium partitioning maintained at the surface-air  
64 interface.<sup>10-14</sup> Emissions from surfaces are then regulated by the concentration difference across  
65 the boundary layer combined with a convective mass transfer coefficient that limits mass transfer  
66 between the boundary layer and bulk air. In a chamber study of vinyl flooring, Clausen et al.  
67 noted that temperature was a primary factor controlling diethyl hexyl phthalate (DEHP)  
68 emissions.<sup>14</sup> Temperature changes affect equilibrium partitioning between the gas-phase and  
69 condensed-phase surface reservoirs, including surface films. Such processes have been  
70 characterized using thermodynamic models.<sup>15-18</sup>

71  
72 Airborne particles may significantly influence SVOC emission rates from surfaces by  
73 transporting SVOC mass out of the boundary layer and/or by providing an airborne condensed-  
74 phase sink.<sup>19</sup> Chamber studies of organophosphate flame retardants and plasticizers have  
75 demonstrated similar partitioning phenomena whereby the presence of airborne particles  
76 enhances the rate of SVOC emissions from source materials.<sup>20-22</sup> Moreover, the affinity of  
77 SVOCs for airborne particles is affected by both SVOC and particle composition for phthalate  
78 diesters<sup>22,23</sup> and for third-hand smoke (THS) species.<sup>24</sup> These findings have been supported by  
79 measurements in real indoor environments for specific compounds. For example, in a study of a  
80 university classroom, DeCarlo et al. reported that increases in particle mass concentration led to  
81 increases in the concentrations of airborne THS species; they inferred that surface-sorbed THS  
82 species were transported through the gas phase onto aqueous airborne particles, with particle  
83 capacity influenced by acid-base processes.<sup>25</sup> Similarly, increases in DEHP concentrations were  
84 found to be associated with increased airborne particle concentrations in a normally occupied  
85 residence.<sup>8</sup>

86  
87 Indoor environments are subject to dynamic changes in temperature and particle concentration as  
88 influenced by occupants, their activities, and building interactions with the outdoor environment.  
89 Indoor airborne SVOC concentrations may be modulated by both direct primary emissions and  
90 by indirect interactions with surfaces. Few studies have examined how airborne SVOC  
91 concentrations evolve in real indoor settings. In this paper, we report hourly SVOC  
92 concentrations from two field campaigns, H2 and HOMEChem, and we use these data to  
93 characterize SVOC volatility-dependent dynamics and partitioning. We focus on three specific

94 species groupings: (a) total SVOC concentrations binned by volatility, (b) phthalate diesters, and  
95 (c) cyclic siloxanes. These emphases are intended to explore SVOC behavior as a bounded class  
96 and as discrete compounds with varying vapor pressures. The specific objectives of the study  
97 are: (a) to quantitatively describe indoor SVOC dynamics as functions of volatility, temperature,  
98 and particle loading; (b) to connect observations with predictions from theory and laboratory  
99 experiments; and (c) to evaluate the impact of surface emissions on airborne SVOC  
100 concentrations.

## 101 **2 EXPERIMENTAL METHODS**

102 Data analysis from the H2 observational field campaign focuses on a period of vacancy in an  
103 otherwise normally occupied residence. At H2, SVOC concentrations are binned by volatility  
104 and compared against temperature and particle mass concentrations. The HOMEChem campaign  
105 was structured around controlled occupant activities in a research house. At HOMEChem, the  
106 physical behavior of two classes of compounds, phthalate diesters and cyclic siloxanes, is  
107 investigated in relation to their volatility.  
108

109 **Study Sites:** The H2 field campaign was conducted a single-family dwelling in Contra Costa  
110 County, California, from 7 December 2017 to 4 February 2018. In this residence, air temperature  
111 was regulated by a forced-air gas-fired furnace incorporating a MERV 13 filter that influenced  
112 indoor particle levels. The furnace operated under control of a programmable thermostat, with  
113 “on” periods occurring twice daily (06:45–07:15 and 17:45–22:00). The H2 site and field  
114 monitoring campaign are detailed elsewhere.<sup>7,8</sup> The analysis here focuses on a five-day period  
115 (22–27 December) during which the house was unoccupied (the H2 ‘vacant period’).  
116

117 The HOMEChem field campaign was conducted during June 2018 at the UTest House, a  
118 manufactured house located at the JJ Pickle Research Campus of the University of Texas at  
119 Austin. The three-bedroom, 111-m<sup>2</sup> facility was operated with an air-conditioning system set to  
120 maintain a constant indoor air temperature of ~25 °C (298 K). A series of controlled experiments  
121 was conducted over the one-month campaign. Experiments were designed to explore the  
122 influence of cooking, cleaning, and occupancy on indoor air chemical composition. Two  
123 categories of experiments were undertaken. During ‘sequential’ experimental days, repeated ‘stir  
124 fry cooking’ or ‘cleaning’ experiments were conducted with intermittent venting periods to reset  
125 indoor conditions. ‘Layered’ experimental days simulated ‘day-in-the-life’ conditions for a  
126 residence, with meal preparation and cleaning occurring in a sequential manner without venting.  
127 Two high-emission ‘layered’ days were conducted to simulate ‘Thanksgiving,’ with more  
128 intensive cooking and occupancy of the type typical of the holiday meal in an American  
129 household. A full description of the HOMEChem campaign experimental design and an  
130 overview of all instrumental data acquisition is reported elsewhere.<sup>26</sup>  
131

132 **Instrumentation and Measurement Methods:** The research reported here focuses on data  
133 acquired using semivolatile thermal desorption aerosol gas chromatography (SV-TAG), a dual-  
134 channel gas chromatograph mass spectrometer (GC-MS) that quantifies, with hourly time  
135 resolution, gas-phase and gas-plus-particle-phase concentrations of SVOCs with vapor pressures  
136 corresponding to alkanes between ~C14 and ~C35.<sup>27-29</sup> SV-TAG collected the PM<sub>2.5</sub> particle  
137 fraction, excluding larger particles by means of a cyclone (BGI by Mesa Labs, SCC 2.654). After  
138 each sampling period (15 minutes at H2, 20 minutes at HOMEChem), the captured analytes are  
139

140 thermally desorbed from the collection cells, separated based on volatility using a gas  
141 chromatograph (Agilent 7890A) and analyzed using a 70-eV electron ionization (EI) mass  
142 spectrometer (Agilent 5975C). Measurements are repeated automatically at hourly intervals. SV-  
143 TAG was deployed during the H2 and HOMEChem campaigns. Operating and sampling  
144 parameters during the H2 campaign are described in the SI (Table S1) and in prior  
145 publications.<sup>7,8</sup> Operating parameters during the HOMEChem campaign were unchanged from  
146 the H2 campaign. Sampling parameters during HOMEChem are described in Table S2 and in  
147 Farmer et al.<sup>26</sup> Characteristic ions of most species were integrated using the TERN software,  
148 normalized to relevant internal standards, and calibrated against authentic external standards.<sup>30</sup>  
149 Quantification of low-volatility siloxanes (Figure S1), for which authentic external standards  
150 were not available, is described in the SI.

151  
152 At HOMEChem, particle number concentrations were quantified by two separate scanning  
153 mobility particle sizers, with one implementing a nano differential mobility analyzer (4-105 nm:  
154 TSI 3080 EC + 3085 nano-DMA + 3788 water CPC) and one implementing a long differential  
155 mobility analyzer (105-532 nm: TSI 3080 EC + 3081 long-DMA + 3787 water CPC). Size-  
156 resolved concentrations of larger particles (diameter > 542 nm) were determined by an  
157 aerodynamic particle sizer (TSI 3321). At H2, particle number concentrations were quantified by  
158 a Grimm 11-A optical particle counter reporting time-resolved measurements in 31 size-  
159 segregated bins with particle diameters ranging from 0.25 to 32  $\mu\text{m}$ . For the HOMEChem study,  
160 an assumed particle density of  $1 \text{ g cm}^{-3}$  (similar to the density of cooking-related aerosol) was  
161 used for consistency with other work.<sup>31,32</sup> The present work similarly assumed a particle density  
162 of  $1 \text{ g cm}^{-3}$  at the H2 site for internal consistency. (A density of  $1.67 \text{ g cm}^{-3}$  was assumed in prior  
163 published work at the H2 site.<sup>8,33,34</sup>) The presence of particle-bound siloxanes was confirmed  
164 with supporting measurements from a high-resolution time-of-flight aerosol mass spectrometer  
165 (HR-ToF-AMS), with experimental details available in the SI (Figure S2).

166  
167 **SVOC Integration:** The chromatographic signal from SV-TAG was converted into total SVOC  
168 concentrations following the approach of Kristensen et al.<sup>7</sup> In the present work, the  
169 chromatogram was subdivided into bins where bin borders were defined by the midpoints of the  
170 retention times of adjacent alkanes. Then, each bin was normalized to the closest alkane internal  
171 standard in retention time and quantified using calibration curves prepared for the closest alkane  
172 in retention time (Figure 1). Normalization by internal standards is needed to account for a  
173 general decline in ion-source response (restorable by source-cleaning) and for matrix loading  
174 effects. Alkane-internal standards are used as the closest surrogate for each SVOC-bin in  
175 volatility space but may not be fully representative for highly polar compounds contained within  
176 each bin. The retention time is closely related to compound volatility, although other parameters,  
177 such as polar interactions with the column, can influence retention. Summing all bins and  
178 subtracting blank ‘internal-standard only’ measurements yielded the ‘total SVOC’ concentration.  
179 SV-TAG incorporates in-situ derivatization in its analytical procedure, which may affect the  
180 retention time of derivatized compounds. The derivatization agent, N-methyl-N-  
181 (trimethylsilyl)trifluoroacetamide (MSTFA), reacts with hydroxy groups such as carboxylic  
182 acids, alcohols, sugars, and similar analytes, and replaces active hydrogen atoms with  
183 trimethylsilyl groups. Silylation occurs for only a small subset of captured analytes and generally  
184 shifts the retention time by no more than 1-2 alkane-equivalent volatility bins. Most of the  
185 volatility bins are expected to be captured by the collection and thermal desorption (CTD) cell

186 with negligible loss through the collection and transfer processes. Collection efficiencies of gas-  
187 phase organics on the CTD cell are expected to be high (80-100% for most measured  
188 compounds). Transfer efficiencies off the CTD cell and focusing trap were > 95% for the C15 –  
189 C26 alkanes and decreased to 50% by the C32 alkane, while the transfer efficiency of the C14  
190 alkane was 40%. The uncharacterized C13 alkane transfer efficiency is expected to be lower.<sup>27</sup>  
191 Quantitative corrections for transfer losses were made using deuterated alkane internal standards  
192 deposited on the CTD cell and analyzed with every sample.

193  
194 **Modeling Temperature Dependence of Gaseous SVOCs:** Past models have been developed to  
195 estimate gas-phase SVOC concentrations in two separate cases.<sup>35</sup> In one case, SVOCs are  
196 emitted from surfaces when additive SVOCs are released from a parent material, such as in the  
197 case of plasticizers. Then, equation (1) describes the gas-phase concentration  $y$ , where  $h$  is the  
198 convective mass-transfer coefficient over the emission surface,  $y_0$  is the gaseous SVOC  
199 concentration immediately adjacent to the surface,  $A$  is the source surface area, and  $Q^*$  is the  
200 “equivalent ventilation rate,” a parameter related to the ventilation rate that accounts for particle-  
201 associated removal and is described more thoroughly in the SI.

$$202 \quad y = \frac{h \times y_0 \times A}{h \times A + Q^*} \quad (Eq. 1)$$

203  
204  
205 In another case, SVOCs can be emitted from surfaces when a previously sorbed or otherwise  
206 deposited SVOC is released from that surface, such as in the case of pesticide or water-repellant  
207 treatments. This process occurs over two phases. For each phase, equation (2) describes the gas-  
208 phase SVOC concentration, where  $A_s$  refers to the sorbing interior surface area and  $h_s$  is the  
209 convective mass-transfer coefficient over interior surfaces.

$$210 \quad y = \frac{h \times y_0 \times A}{h \times A + h_s \times A_s + Q^*} \quad (Eq. 2)$$

211  
212  
213 Under variable temperature, changes in  $y$  across the observed temperature range (288 – 292 K)  
214 are assumed to be governed by changes in the equilibrium constant controlling  $y_0$ , as described  
215 in equation (3) where  $[SVOC_{surf}]$  is the condensed-phase SVOC concentration at the surficial  
216 interface.

$$217 \quad K_{eq}(T) = \frac{y_0}{[SVOC_{surf}]} \quad (Eq. 3)$$

218  
219  
220 Such changes in equilibrium partitioning are anticipated to be similar to those described by the  
221 van't Hoff equation as shown in equation (4), where  $T$  is temperature,  $\Delta S$  is the entropy of  
222 vaporization,  $R$  is the gas constant, and  $k$  is a constant described by  $-\Delta H/R$ , where  $\Delta H$  is the heat  
223 of vaporization.<sup>18</sup>

$$224 \quad K_{eq}(T) = \exp \left[ \frac{\Delta S}{R} + k \left( \frac{1}{T} \right) \right] \quad (Eq. 4)$$

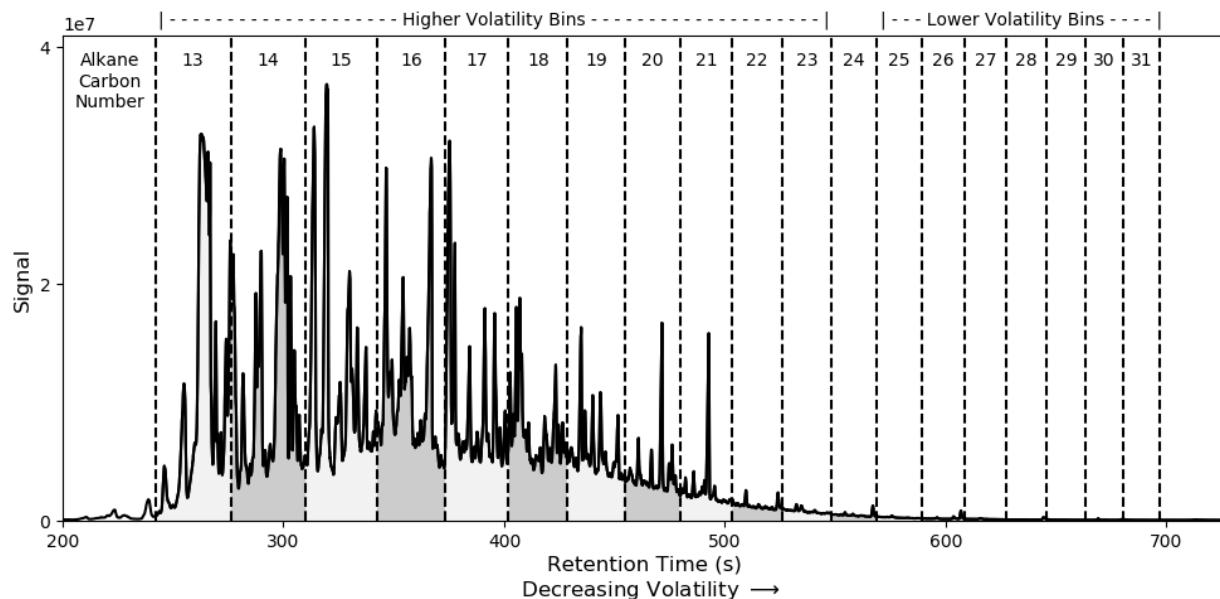
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227 We present a model describing gas-phase SVOC concentrations based on the framework  
 228 developed in equations (1-4). Then,  $y$  as a function of temperature is described by equation (5),  
 229 where  $a$  is a variable containing terms that are generally independent of temperature. A full  
 230 derivation of equation (5) is available in the SI. Because the van't Hoff equation applies to  
 231 equilibrium partitioning of a pure material and the studied indoor physical system consists of a  
 232 complex mixture, we refer to experimentally derived  $k$  values as  $k^*$ .

$$y(T) = a \times \exp\left[k^* \left(\frac{1}{T}\right)\right] \quad (\text{Eq. 5})$$

### 236 3 RESULTS AND DISCUSSION

237 At H2, SVOC concentrations were determined for chromatographic bins corresponding to the  
 238 volatility of the nearest alkane in retention time ('alkane-equivalent volatility bin'). Estimated  
 239 vapor pressures and saturation concentrations ( $C^*$ ) for each bin are reported in Table S3.  
 240 Concentrations are reported for the vacant period when no occupant activities, such as cooking  
 241 or cleaning, occurred. Departures from steady-state conditions during the vacant period are likely  
 242 to occur with (1) controlled indoor temperature changes related to the home heating system and  
 243 (2) changes in indoor air PM<sub>2.5</sub> concentrations. During the period of vacancy, indoor primary  
 244 particles are expected to originate only from infiltration of outdoor particles and not from any  
 245 indoor source.<sup>8</sup> Observed dynamic behavior of SVOCs was separated into two categories based  
 246 on volatility: the C13-C23 bins, which are predominantly gaseous and display dependence on  
 247 temperature, and the C25-C31 bins, which partition appreciably into airborne particles and  
 248 display dependence on particle concentration.



250 **Fig. 1.** Quantified bins of the SV-TAG chromatogram are displayed for a typical chromatogram.  
 251 Bins were subdivided and quantified based on the closest alkane in retention time.

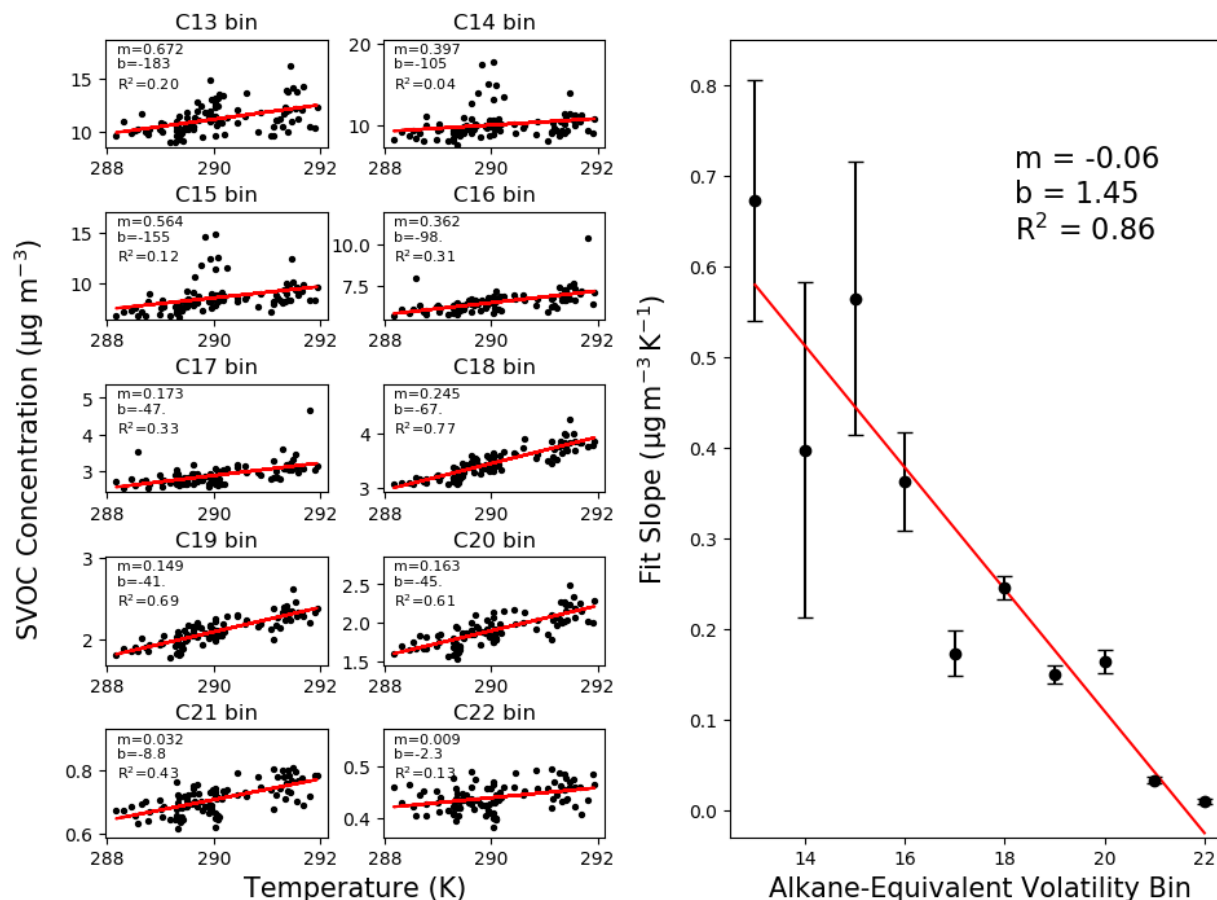
252  
 253 **Abundance of Higher Volatility SVOCs is Associated with Temperature.** During the vacant  
 254 period at H2, observed total (gas-plus-particle) SVOC concentrations of the C13-C23 bins  
 255 generally decreased with decreasing volatility. Average concentrations and associated summary  
 256

257 statistics of each alkane-equivalent volatility bin are reported in Table S3. Concentrations in the  
258 C23 bin were roughly fifty times lower than concentrations in the C13 bin. In Figure 2, SVOC  
259 concentrations are compared binwise against indoor air temperature, with ordinary least squares  
260 regression lines superimposed. For the C13-C23 bins (C13-C22 shown), SVOC concentrations  
261 showed statistically significant positive variation with temperature. Furthermore, the magnitude  
262 of the fitted slopes tended to decrease with decreasing bin volatility. Significant positive  
263 variation with temperature was not observed beyond the C23 bin. Time series and comparisons  
264 against temperature for each bin are shown in the SI (Figures S3, S4).

265  
266 Observed temperature dependencies can be connected to the model described in equation (5).  
267 Predicted  $k^*$  values were calculated from literature values for alkane heats of vaporization,  
268 which correlate with vapor pressure.<sup>37,38</sup> Experimental  $k^*$  values are within 50% of predicted  $k^*$   
269 values, a remarkably close correspondence (Figure S5). Discrepancies between the two values  
270 may arise because the vapor pressures of the measured organics differ from alkane vapor  
271 pressures and because the studied system involves vaporization from a complex mixture rather  
272 than from a pure condensed phase. Furthermore, the application of equation (5) requires that  
273 emissions from surfaces are the dominant source process. Larger differences between the  
274 experimental and predicted values of  $k^*$  are observed for the C21 and C22 alkane-equivalent  
275 volatility bins. Other factors, such as appreciable interactions with airborne particles, are  
276 expected to partially account for this observation.

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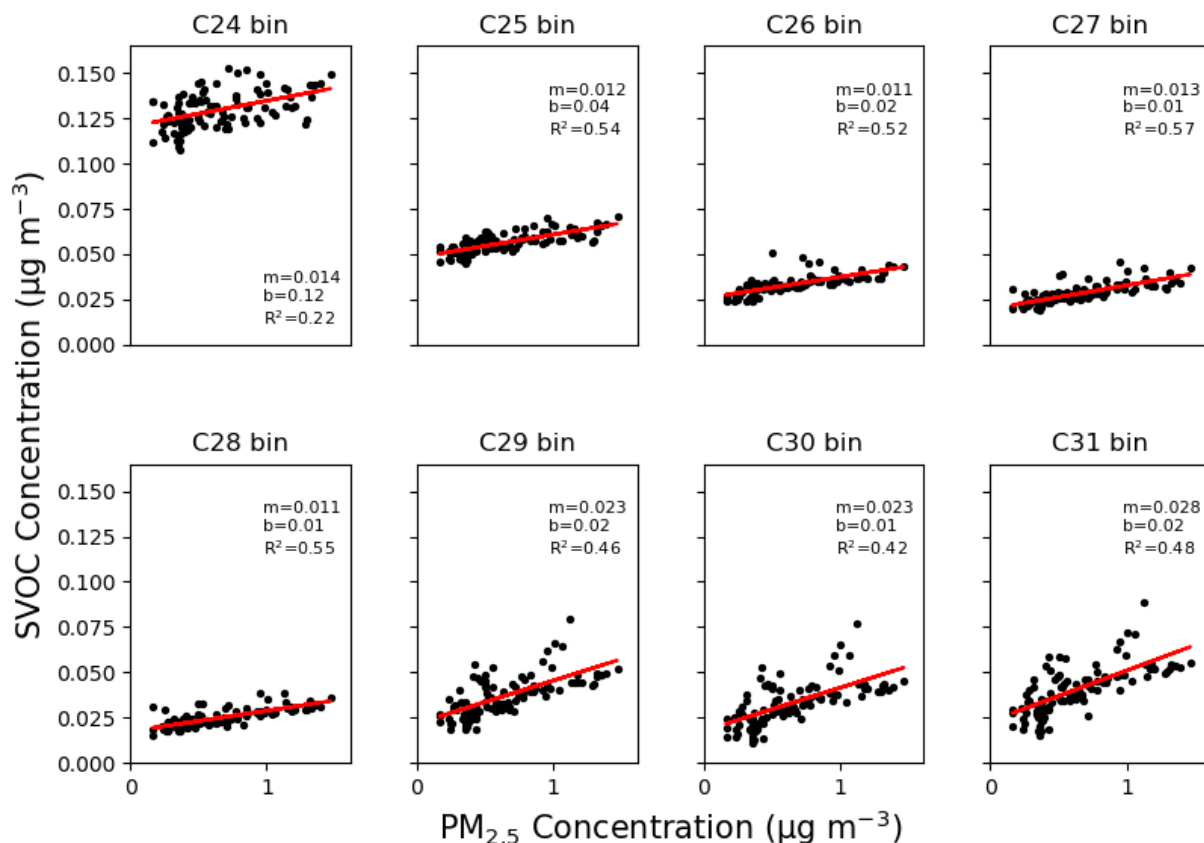
**Fig. 2.** In the left panel, total (gas-plus-particle) SVOC concentrations ( $\mu\text{g m}^{-3}$ ) are compared against temperature. Each measured point represents a 15-minute sample collection period with hourly replication during the observational period. Units of measure for the linear fit slope and intercept are  $\mu\text{g K}^{-1} \text{m}^{-3}$  and  $\mu\text{g m}^{-3}$ , respectively. In the right panel, the magnitude of the linear fit slope is compared against the corresponding alkane-equivalent volatility bin.

288 **Abundance of Lower Volatility SVOCs is Associated with Particle Concentration.** During  
 289 the vacant period at H2, consistent associations between total airborne SVOC concentrations and  
 290 particle mass concentration were observed for the C25-C31 bins (Figure 3). We stress that SV-  
 291 TAG samples only the  $\text{PM}_{2.5}$  particle fraction; coarse-mode particles (as observed during  
 292 resuspension events) likely have a different chemical composition than  $\text{PM}_{2.5}$ , which may alter  
 293 equilibrium partition behavior. Furthermore, equilibration time scales are slower for larger  
 294 particles and faster for smaller particles. As such, equilibration is less likely to be achieved  
 295 during the time of indoor suspension for coarse particles as compared to fine particles.<sup>36</sup> Time  
 296 series and comparisons against particle mass concentrations for each bin are shown in the SI  
 297 (Figures S3, S6). These bins had substantial fractions ( $\sim 0.4$  to  $\sim 1.0$ ) of airborne mass in the  
 298 particle-phase (Table S3). Conversely, positive associations between SVOC concentrations and  
 299 particle mass concentration were not observed for the C13-C23 bins. SVOCs contained in these  
 300 higher volatility bins were predominantly in the gas-phase and are not expected to strongly  
 301 partition to particles. The intercepts of the ordinary least-squares fits displayed in Figure 3 are

302 related to the airborne gas-phase concentration in the absence of particles. As SVOC volatility  
303 decreases, the background gas-phase SVOC concentration generally decreases. Gas-particle  
304 partitioning by bin is displayed in Figure S7. As expected, the observed particle fraction,  $F_p$ ,  
305 generally increases with increased particle mass concentration and with decreased vapor  
306 pressure.

307  
308 The strong correlation ( $R^2 > 0.4$ ) between  $PM_{2.5}$  and the airborne SVOC concentrations in the  
309 C25-C31 bins suggests a net outcome of surface-particle partitioning as SVOCs are transported  
310 from condensed-phase reservoirs. We infer that the gas-phase acts as a transport medium  
311 between condensed-phase SVOC surface reservoirs and airborne particles. Gas-phase SVOC  
312 concentrations were relatively constant under variable particle concentrations considering  
313 experimental uncertainty (Figure S8). Correlations between  $PM_{2.5}$  and SVOC concentrations for  
314 bins below C25, which are predominantly in the gas-phase, were weaker ( $R^2 < \sim 0.3$ ) or not  
315 observed (Figure S6).

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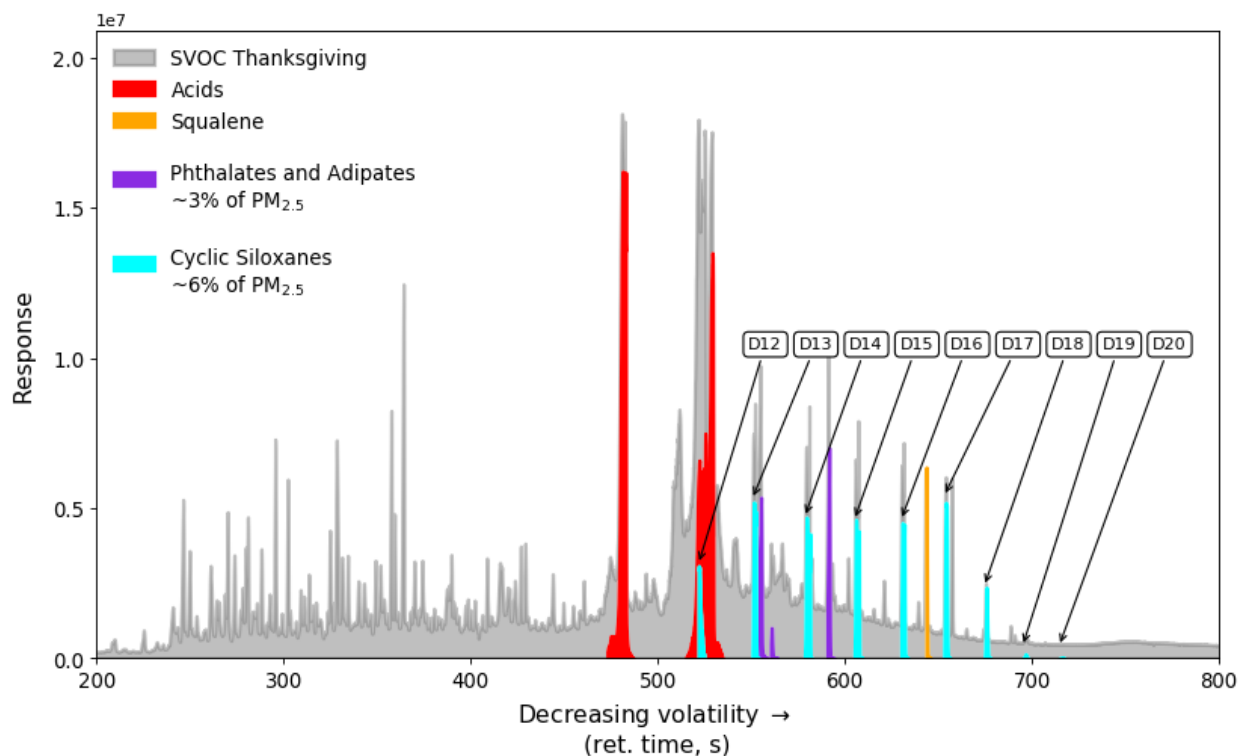
321  
322 **Fig. 3.** Binned total (gas-plus-particle) SVOC concentrations ( $\mu\text{g m}^{-3}$ ) are compared against  
323  $PM_{2.5}$  mass concentrations ( $\mu\text{g m}^{-3}$ ) at H2 during the vacant period. The linear fit slope ( $m$ ) is  
324 dimensionless; the intercept ( $b$ ) has units of  $\mu\text{g m}^{-3}$ . Each measured point represents a 15-minute  
325 sample collection period with hourly replication during the observational period.

326  
327 Infiltration of outdoor gas- and particle-phase SVOCs as well as changes in source emission rates  
328 may also influence observed concentrations in indoor air. Indoor-outdoor concentration ratios of  
329 the alkane-equivalent volatility bins were slightly below unity with moderate variability (Figure  
330 S9). However, because outdoor particle mass concentrations were greater than indoor particle  
331 mass concentrations, SVOC concentrations per particle mass were greater indoors than outdoors.  
332 As indoor particles during the H2 vacant period are expected to be primarily of outdoor origin,<sup>8</sup>  
333 this evidence suggests the occurrence of net partitioning of indoor SVOCs to outdoor particles  
334 upon transport to the indoors. Particle-normalized indoor/outdoor ratios of alkane-equivalent  
335 volatility bins display substantial variability, whereas indoor SVOC concentrations were highly  
336 regular. The regular correspondence between SVOC concentrations and PM<sub>2.5</sub>, coupled with  
337 high normalized indoor/outdoor ratios, suggests that partitioning processes may be modulating  
338 observed indoor SVOC concentrations at time scales comparable to or faster than the air-  
339 exchange rate. Under classical partitioning theory, it is assumed that SVOCs partition by  
340 absorption into the organic PM<sub>2.5</sub> fraction rather than by adsorption to bulk particle surfaces.<sup>15</sup>

341  
342 **Indirect Surface Emissions Contribute to Indoor Particle Mass.** During the first  
343 HOMEChem ‘Thanksgiving’ experiment, substantial proportions of airborne PM<sub>2.5</sub> mass  
344 originated from primary cooking emissions. Among primary species emitted are palmitic acid,  
345 stearic acid, and other carboxylic acids with varying degrees of unsaturation. Such species have  
346 been reported to be directly emitted from cooking in laboratory studies.<sup>40-42</sup> We also found that  
347 squalene emissions were strongly associated with oven usage on Thanksgiving; the expected  
348 source is volatilization of oils from the skin of the roasting turkey. An additional source could be  
349 from human skin oils present on heated surfaces or cookware. Squalene was observed during  
350 other cooking events at roughly one-to-two orders of magnitude smaller abundance.

351  
352 Surprisingly, we also observed on this day elevated indoor particle-phase concentrations of  
353 specific SVOCs that should not originate from the food itself, in particular phthalates, adipates,  
354 and siloxanes (Figure 4, Figures S1, S2). Phthalates and adipates are plasticizers commonly used  
355 in vinyl flooring.<sup>43</sup> Low-volatility siloxanes are used as thermally stable lubricating greases and  
356 sealants. Remarkably, plasticizer and siloxane concentrations accounted for approximately 10%  
357 of airborne particle mass at certain times on this experimental day, suggesting that partitioning  
358 processes between particles and condensed-phase reservoirs significantly influenced the  
359 observed airborne concentrations. Moreover, these observations highlight difficulties in  
360 determining exact SVOC emission profiles of events in a real-world setting: surface emissions  
361 that were indirectly stimulated by primary event-driven particle emissions contributed sizeable  
362 fractions of indoor PM<sub>2.5</sub> mass.

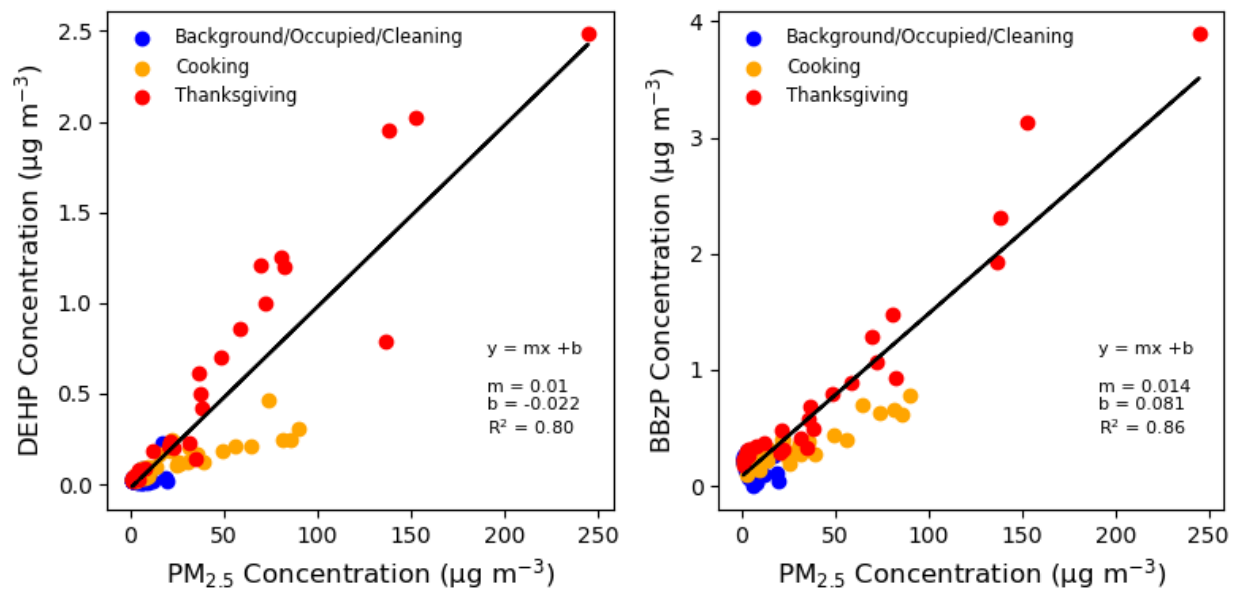
363



364  
 365 **Fig. 4.** A particle-phase chromatogram from the first ‘Thanksgiving’ experiment day ( $PM_{2.5}$   
 366 concentration =  $245 \mu\text{g m}^{-3}$ ) on June 18 at 3:05 PM is displayed. Direct emissions attributable to  
 367 cooking (carboxylic acids and squalene) are highlighted in red and orange, respectively. Indirect  
 368 emissions likely attributable to the building composition are highlighted in purple (plasticizers)  
 369 and teal (siloxane lubricants and heat-transfer materials).

370  
 371 In previous analyses of data from the H2 field campaign, correlations between particle mass  
 372 concentration and airborne DEHP concentration suggested that particulate matter rapidly  
 373 acquired surface-laden DEHP from reservoirs such as organic surface films and dust.<sup>8</sup> Prior  
 374 analysis of airborne DEHP concentrations at the UTest House characterized the role of  
 375 temperature over a 9 K range.<sup>44</sup> In that case, on a long-term basis, a 9 K increase in temperature  
 376 approximately doubled airborne DEHP and butyl benzyl phthalate (BBzP) concentrations.  
 377 However, there was no systematic examination in that study of particle-dependent variations in  
 378 phthalate concentrations. In the current work at the UTest House, with temperature  
 379 approximately constant at 298 K, strong associations between particle concentration and total  
 380 airborne SVOC concentrations were observed for both DEHP and BBzP (Figure 5).

381  
 382 Substantial portions of the UTest House vinyl flooring are known to contain phthalates.<sup>44</sup>  
 383 Because both DEHP and BBzP are expected to originate from the composition of the flooring  
 384 material, and because airborne phthalate concentrations are strongly associated with particle  
 385 concentration for many different source events, it is similarly inferred that transport through the  
 386 gas phase from condensed-phase stationary reservoirs to airborne particles is the principal source  
 387 of observed particulate phthalate concentrations during the HOMEChem campaign. DEHP  
 388 resides in the C25 alkane-equivalent volatility bin and BBzP resides in the C24 alkane-  
 389 equivalent volatility bin based on chromatographic retention time.



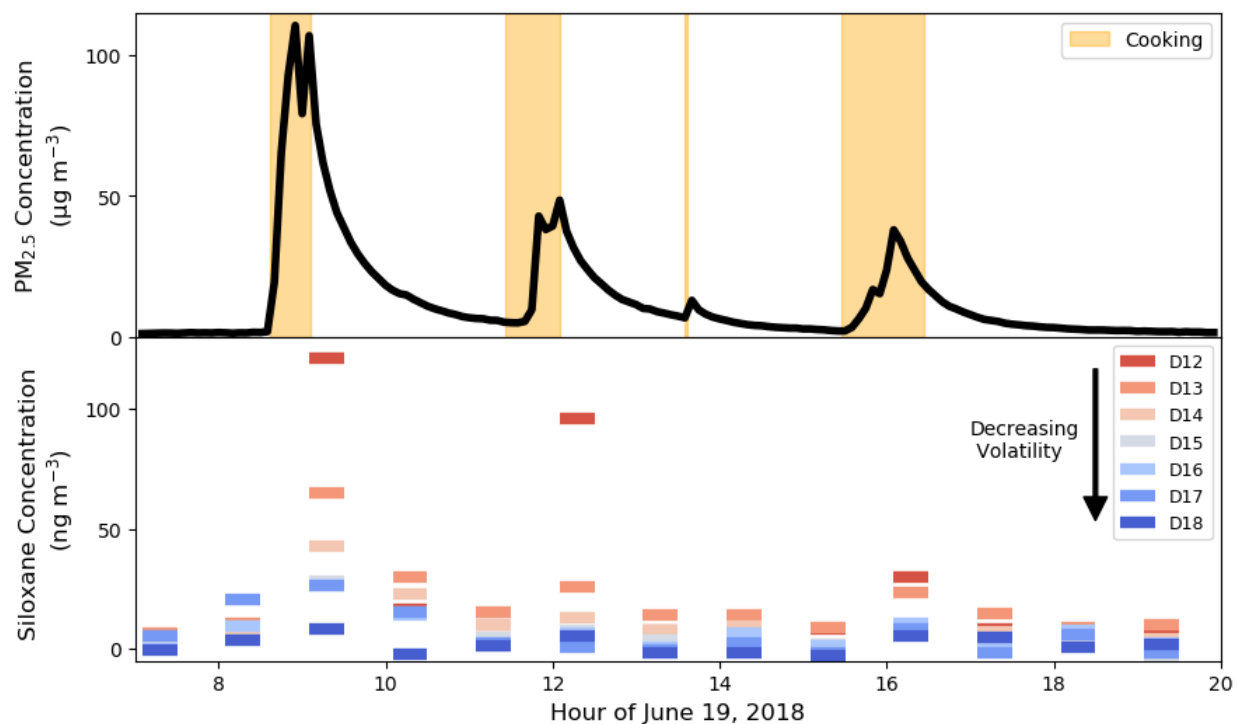
391  
 392 **Fig. 5.** Total (gas-plus-particle) concentrations ( $\mu\text{g m}^{-3}$ ) of two phthalates, DEHP and BBzP, are  
 393 compared against particle mass concentration ( $\mu\text{g m}^{-3}$ ) for measurements from the HOMEChem  
 394 campaign. Units of measure for the fit slope ( $m$ ) and intercept ( $b$ ) are unitless and  $\mu\text{g m}^{-3}$ ,  
 395 respectively.

396  
 397 **Lower Volatility Siloxanes Exhibit Ongoing Emissions after High Emission Event.**  
 398 Surprisingly high concentrations of low-volatility siloxanes (D13-D20 cyclic and L13-L19 linear  
 399 siloxanes) were observed during a particle loading event associated with the HOMEChem  
 400 ‘Thanksgiving’ experiment day on June 18 in association with cooking and oven use. We  
 401 hypothesize that the low-volatility siloxane source is the oven, an appliance that likely contains  
 402 heat-transfer compounds and thermally stable lubricants. Commercially available products  
 403 containing siloxanes have been recommended for such uses in ovens.<sup>45</sup> In principle, high  
 404 temperatures attained throughout the oven during cooking could have driven appreciable  
 405 amounts of low-volatility siloxanes into the gas-phase, which subsequently condensed onto  
 406 airborne particles as air exited the oven and cooled. Although the oven was approximately  
 407 thirteen years old, it had been operated only three times prior to the June 18 Thanksgiving event.  
 408 Peak concentrations of low-volatility siloxanes during the ‘Thanksgiving’ experiment day are  
 409 reported in Table S4. Minor siloxane enhancements were observed in the morning (stovetop  
 410 breakfast preparation) and evening (oven cooking) of the June 8 layered day, but not during the  
 411 lunch-time stir-fry event.

412  
 413 Small enhancements of D18 were observed during cooking events on the June 17 sequential stir-  
 414 fry day; enhancements of other siloxanes were not observed. In contrast, D12-D14 siloxanes  
 415 were strongly enhanced during cooking events on the June 19 ‘layered’ experiment day. A  
 416 plausible explanation for these observations is that siloxanes emitted during the June 18  
 417 ‘Thanksgiving’ experiment day were deposited on surfaces throughout the residence and were  
 418 subsequently reemitted during high particle emission events (Figure 6). Reemission of the  
 419 semivolatile siloxanes occurred more readily for smaller homologues, which have  
 420 correspondingly higher vapor pressures. Lower-volatility siloxanes are expected to preferentially  
 421 partition to airborne particles; however, airborne concentration enhancements may not be

422 observed for species with especially low volatility, for which equilibration time scales are  
423 significantly longer than the particle residence time indoors.

424  
425 The D12-D19 siloxanes reside in the C22-C31 alkane-equivalent volatility bins, suggesting that  
426 their physicochemical properties may be well suited for reemission from surfaces. Vapor  
427 pressures and octanol-air partition coefficient ( $K_{oa}$ ) values for D12-D19 were calculated using  
428 SPARC and are reported in Table S5. Using the method of Weschler and Nazaroff<sup>1</sup> and  
429 assuming a condensing-particle diameter of 100 nm and a gas-phase diffusivity of  $0.03 \text{ cm}^2 \text{ s}^{-1}$ ,  
430 gas-particle equilibration time scales for D12 are expected to be on the order of 8 days while  
431 equilibration time scales for D19 are expected to be effectively infinite ( $\sim 10^9 \text{ y}$ ). A mechanical  
432 ventilation system was operated to maintain an air-exchange rate of  $\sim 0.5$  per hour during the  
433 HOMEChem campaign.<sup>26</sup> The calculated vapor pressure of D12 ( $10^{-12.1} \text{ atm}$ ) is similar in  
434 magnitude to that of DEHP ( $10^{-11.85} \text{ atm}$ ), a compound that displayed prominent gas-particle  
435 interactions in a normally occupied residence.<sup>8</sup> The calculated vapor pressure of D19 is eight  
436 orders of magnitude lower. In summary, airborne concentration enhancements may occur for  
437 D12 in association with increased  $\text{PM}_{2.5}$  loading but are not expected to occur for D19 without an  
438 additional stimulus, such as the inference that the high-temperature event associated with oven-  
439 use drove D19 into the gas-phase. Linear siloxanes, which were emitted at far lower  
440 concentrations during the June 18 Thanksgiving emission event, were not observed significantly  
441 above background during the June 19 layered day. Small siloxane enhancements were observed  
442 for D12-D16 siloxanes on the June 12 sequential stir-fry day. These observations may be related  
443 to deposition events from oven use on June 5 and June 8.  
444



445  
446 **Fig. 6.** Total (gas-plus-particle) airborne concentrations ( $\mu\text{g m}^{-3}$ ) of a siloxane homologous series  
447 superimposed on  $\text{PM}_{2.5}$  concentrations ( $\mu\text{g m}^{-3}$ ) for a ‘layered’ experiment day on June 19  
448 following the June 18 ‘Thanksgiving’ experiment day.

449  
450 **Implications.** In the absence of episodic emission sources, key driving factors influencing  
451 variability of indoor airborne SVOC concentrations are volatility and partitioning phenomena.  
452 Airborne concentrations of the higher volatility (C13-C23 bins; predominantly gaseous) SVOCs  
453 are mainly sensitive to surface temperatures. Concentrations of lower volatility (C25-C31 bins;  
454 substantially particle-phase) SVOCs are sensitive to airborne particle concentrations. This work  
455 suggests that, at the H2 residence, the dynamic behavior of specific SVOCs can be predicted if  
456 their volatility is known. Ultimately, if demonstrated to be generalizable, such understanding  
457 would contribute to improving exposure assessment and mitigation strategies.

458  
459 Emissions of low-volatility siloxanes and phthalates from surfaces are inferred to have been  
460 indirectly stimulated by event-driven emissions of particles. Analysis of low-volatility siloxane  
461 concentrations suggests that SVOCs can be deposited throughout a residence and then reemitted  
462 during subsequent particle loading events. This effect was most important for siloxanes with  
463 significant particle-bound fractions and appreciable gas-phase fractions. Despite similar total  
464 (gas-plus-particle) airborne concentrations during the initial source event, smaller concentrations  
465 of lower volatility siloxanes were observed during reemission events compared to higher  
466 volatility siloxanes. Because indoor air is the transporting medium between condensed-phase  
467 surface reservoirs and airborne particles, the lowest-volatility siloxanes are expected to remain in  
468 condensed-phase reservoirs when considering kinetic transport limitations. These lowest-  
469 volatility siloxanes were observed during primary emission events involving oven use but not  
470 appreciably during reemission episodes.

471  
472 These observations of airborne SVOC concentrations are consistent with, but not fully  
473 demonstrative of prior modeling and laboratory results. More work is needed to strengthen  
474 confidence in current models by connecting speciated measurements of surface-sorbed organics  
475 to airborne SVOC concentrations over longer time scales and in other indoor spaces. Future  
476 analysis would benefit from chemically differentiating primary event emissions and indirect  
477 event emissions where surface-sorbed species are enhanced in indoor air by primary particles.  
478 The same primary emission event could produce significantly different airborne SVOC  
479 concentrations, and ultimately occupant exposures, in indoor environments with different  
480 condensed-phase reservoirs.

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491 by the Committee for Protection of Human Subjects for the University of California, Berkeley  
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493

494 **6 SUPPORTING INFORMATION**

495 SV-TAG sampling conditions; SV-TAG data analysis; HR-ToF-AMS instrument operation;  
496 gaseous SVOC modeling; SVOC bin summary statistics; SVOC bin time series; SVOC bin  
497 versus temperature and PM<sub>2.5</sub> concentrations, SVOC bin gas-particle partitioning, SVOC bin  
498 indoor/outdoor ratios, HOMEChem siloxane concentrations, siloxane physicochemical  
499 properties  
500

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