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## **Growth of Organic Films on Indoor Surfaces**

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## S1. What about particles?

This section presents estimates of fine- and coarse-mode particle contributions to total mass as well as organic matter on vertical and horizontal surfaces. The results of this section are summarized in Table 3 of the main article.

**Indoor concentrations.** Data suggest that in US housing, the central tendency for the indoor concentration of fine-mode particles, absent smoking, is in the range of 10-20  $\mu\text{g}/\text{m}^3$ .<sup>1-4</sup> For coarse-mode particles, typically reported time-average concentrations are in the range 1-15  $\mu\text{g}/\text{m}^3$ .<sup>1-3,5</sup>

**Vertical surfaces, fine-mode particle accumulation.** Within the fine mode, most of the mass is expected to be present in particles between 0.1 and 1.0  $\mu\text{m}$  diameter. For walls, windows, and other vertically oriented surfaces, the deposition velocity,  $v_d$ , for these particles is anticipated to have a maximum value of  $10^{-4}$  cm/s (0.004 m/h); a reasonable estimate of the central tendency value is  $5 \times 10^{-5}$  cm/s (0.002 m/h).<sup>6</sup> The corresponding maximum rate at which fine-mode particles would accumulate on vertical indoor surfaces is therefore anticipated to be about  $20 \mu\text{g m}^{-3} \times 0.004 \text{ m/h} \sim 0.07 \mu\text{g}/(\text{m}^2 \text{ h})$  or  $\sim 2 \mu\text{g}/(\text{m}^2 \text{ d})$ ; the corresponding central tendency is estimated to be  $\sim 0.5\text{-}1 \mu\text{g}/(\text{m}^2 \text{ day})$ . If the fine mode particles are 50% organic matter by weight, these estimates correspond to estimated accumulation rates in the range  $0.2\text{-}1 \mu\text{g}/(\text{m}^2 \text{ d})$  for organics associated with fine-mode particles.

During cold weather, interior window surfaces are normally cooler than room air, especially if the window glass is single paned. Under such conditions, thermophoresis will substantially increase the deposition velocity of fine-mode particles. For a temperature difference between the window surface and air of 4 °C, predicted deposition velocities for particles in the size range 0.1-1  $\mu\text{m}$  diameter are approximately  $6 \mu\text{m s}^{-1}$  or about  $0.02 \text{ m h}^{-1}$ .<sup>7</sup>

This is 10× as high as the central tendency value used in the preceding estimate and leads to accumulation rate estimates of 5-10  $\mu\text{g}/(\text{m}^2 \text{ d})$  for fine mode total mass and 3-5  $\mu\text{g}/(\text{m}^2 \text{ d})$  for organics associated with fine-mode particles.

**Vertical surfaces, coarse-mode particle accumulation.** Lai and Nazaroff<sup>8</sup> measured deposition velocities of coarse-mode particles to vertical glass surfaces in a stirred chamber. The mean deposition velocity for 5  $\mu\text{m}$  particles was  $4.5 \times 10^{-4} \text{ cm/s}$  (0.016 m/h); for 7  $\mu\text{m}$  particles, the corresponding value was  $12 \times 10^{-4} \text{ cm/s}$  (0.043 m/h). Hence the estimated rate at which coarse-mode particles would soil vertical surfaces is in the approximate range 0.02-0.6  $\mu\text{g}/(\text{m}^2 \text{ h})$  or 0.4-15  $\mu\text{g}/(\text{m}^2 \text{ d})$ . If 20% of the coarse mode particle mass is organic, this range of rates would correspond to organic accumulation on vertical surfaces of 0.08-3  $\mu\text{g}/(\text{m}^2 \text{ d})$  as a consequence of coarse particle deposition.

**Upward horizontal surfaces, fine-mode particle accumulation.** The deposition of particles to the topside of horizontal surfaces is influenced by gravity; deposition velocities can be larger than for vertical surfaces. For particles between 0.1 and 1.0  $\mu\text{m}$ , the central tendency for  $v_d$  is 0.04 m/h.<sup>6</sup> Hence, a typical fine-mode flux to upward horizontal surfaces is roughly 0.4-0.8  $\mu\text{g}/(\text{m}^2 \text{ h})$  or 10-20  $\mu\text{g}/(\text{m}^2 \text{ d})$ . Assuming that the fine-mode particles are 50% organic by weight results in fine-mode particles contributing organics to horizontal surfaces at about 5-10  $\mu\text{g}/(\text{m}^2 \text{ d})$  under typical indoor conditions.

**Upward horizontal surfaces, coarse-mode particle accumulation.** The value of  $v_d$  for coarse-mode particles accumulating on horizontal surfaces is relatively large – approximately 5-30 m/h, depending on diameter and density.<sup>6,9-11</sup> For coarse-mode particle concentrations in the range of 1-15  $\mu\text{g}/\text{m}^3$ , this settling rate would correspond to fluxes in the range of 5-450  $\mu\text{g}/(\text{m}^2 \text{ h})$  or 120-11000  $\mu\text{g}/(\text{m}^2 \text{ d})$ . If 20% of the coarse mode particles were organic material, then the

corresponding organic mass accumulation on horizontal surfaces would be 20-2200  $\mu\text{g}/(\text{m}^2 \text{d})$  as a consequence of coarse particle deposition.

**Upward horizontal surfaces, accumulation estimated from occupant emissions.**

Because of the large variability (both owing to mass concentrations and size dependent settling velocities) it is worthwhile to consider alternative approaches to estimating the flux of coarse-mode particles to the upper side of horizontal surfaces. For example, for a seated human occupant with moderate movement, Licina et al.<sup>12</sup> report an emission rate of 170 ( $\pm 100$ )  $\mu\text{g}/\text{h}$  for particles larger than 1  $\mu\text{m}$  diameter. Consider a room with 12  $\text{m}^2$  of topside horizontal surfaces that contains such an occupant for 12 h/d. Assuming that all of the emitted particles are deposited on the upward surfaces, then the accumulation rate on these surfaces attributable to occupant emissions would be  $(170 \mu\text{g}/\text{h}) \times (12 \text{ h}/\text{d}) / (12 \text{ m}^2) = 170 \mu\text{g}/(\text{m}^2 \text{d})$ . Such emissions would include skin flakes and clothing fragments. Skin flakes are approximately 10% skin oil by weight;<sup>13</sup> assume that the percent semivolatile organics in clothing particles is also 10%. Then, the organic accumulation on upward horizontal surfaces as a consequence of coarse particle emissions from a human occupant is estimated to be 20  $\mu\text{g}/(\text{m}^2 \text{d})$ . This value is comparable to the minimum estimated in the previous paragraph; it would be larger if coarse-mode particles generated from resuspension and household activities were included. Note that the experimental approach of Licina et al.<sup>12</sup> was designed to emphasize particle emissions from the human body envelope. Contact surfaces were maintained in a clean state to minimize particle resuspension.

**Upward horizontal surfaces, accumulation estimated from dustfall.** Still another approach for estimating the flux of coarse-mode particles to horizontal surfaces derives from measurements of dustfall — the accumulation of dust particles on the upper side of horizontal surfaces. Edwards et al.<sup>14</sup> made such measurements in four New Jersey homes and reported mean

dustfall rates of  $3700 \mu\text{g}/(\text{m}^2 \text{ d})$  during summer and  $2200 \mu\text{g}/(\text{m}^2 \text{ d})$  during winter. In a more extensive study, designed to be a nationally representative random sample, Rasmussen et al.<sup>15</sup> measured dustfall for 1025 Canadian homes. Of the 559 homes that followed a protocol to not clean for  $7 \pm 1$  day, the measured median dustfall rate was  $10,000 \mu\text{g}/(\text{m}^2 \text{ d})$  with 10<sup>th</sup> and 90<sup>th</sup> percentile values of  $3000$  and  $43,000 \mu\text{g}/(\text{m}^2 \text{ d})$ . If the deposited dust was 20% organic, then the corresponding accumulation on horizontal surfaces as a consequence of routine dustfall would be in the approximate range  $600 - 9000 \mu\text{g}/(\text{m}^2 \text{ d})$ . These values overlap with the range of the results estimated using the deposition velocity approach.

**Table S1.** Approximate relationship between  $K_{oa}$ , saturation vapor pressure, and saturation concentration,  $C_{sat}$  (based on Xiao and Wania<sup>16</sup>).

$\log K_{oa}$	Vapor pressure <sup>a</sup>	$C_{sat}$ <sup>b</sup>	$C_{sat}$ <sup>c</sup>
$[-]$	<i>atm</i>	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{m}^3$
8	$4.11 \times 10^{-7}$	5040	6720
8.5	$1.28 \times 10^{-7}$	1570	2090
9	$3.99 \times 10^{-8}$	490	653
9.5	$1.25 \times 10^{-8}$	153	204
10	$3.88 \times 10^{-9}$	48	63
10.5	$1.21 \times 10^{-9}$	14.8	19.8
11	$3.77 \times 10^{-10}$	4.6	6.2
11.5	$1.18 \times 10^{-10}$	1.44	1.92
12	$3.67 \times 10^{-11}$	0.45	0.60
12.5	$1.14 \times 10^{-11}$	0.14	0.187
13	$3.57 \times 10^{-12}$	0.044	0.058
13.5	$1.11 \times 10^{-12}$	0.014	0.018
14	$3.47 \times 10^{-13}$	0.0043	0.0057

<sup>a</sup> Using Eq. 6 of Xiao and Wania<sup>16</sup>; at 25 °C,  $\log K_{oa} = -0.98784 \log V_p + 1.6914$ , where the units for vapor pressure,  $V_p$  (liquid state) are atmospheres.

<sup>b</sup> Assuming average molecular weight is 300 g/mol; 1 ppb = 12.3  $\mu\text{g}/\text{m}^3$ .

<sup>c</sup> Assuming average molecular weight is 400 g/mol; 1 ppb = 16.4  $\mu\text{g}/\text{m}^3$ .

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