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### Authors

Weschler, CJ  
Nazaroff, WW

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### Organic Films on indoor surfaces

Charles J. Weschler<sup>1,2\*</sup> and William W Nazaroff<sup>3</sup>

<sup>1</sup>Rutgers University, Piscataway, NJ 08854, USA

<sup>2</sup>Technical University of Denmark, DK-2800 Lyngby, Denmark

<sup>3</sup>University of California, Berkeley, CA 94720-1710, USA

\*Corresponding email: [weschlch@rwjms.rutgers.edu](mailto:weschlch@rwjms.rutgers.edu)

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#### 1 Introduction

When new and clean, surfaces exposed to indoor environments can possess distinctive chemical properties. Over time, organic films form on indoor surfaces via sorption of gas-phase organics and deposition of airborne particles. On vertical surfaces in typical indoor environments, calculations suggest that the former process occurs at a much faster rate than the latter. In what follows, we illuminate the dynamics of organic film formation, exploring the important emergent properties associated with the growth of such films on indoor surfaces. This study has several specific aims: (1) to apply knowledge of the physics and chemistry of semivolatile organic compounds (SVOCs) so as to construct a conceptual model of the dynamics of surface film growth focusing on uptake of gas-phase SVOCs with varying  $K_{oa}$  values; (2) to reconcile and interpret what we know from experiments with knowledge of physical and chemical behavior of SVOCs; and (3) to add new insights to the collective scientific understanding of indoor surface film occurrence and growth.

#### 2 Methods

In an indoor setting, one might anticipate that gas-phase SVOCs initially *adsorb* to impermeable indoor surfaces. This process is envisioned to occur quickly, but to only form a thin layer, perhaps of the order of a monolayer thick. Soon SVOCs will begin to *absorb* into the organics previously accumulated. As the sorption process proceeds, the thickness of this nascent surface film grows. One can describe

the process governing the absorption and growth of this organic film in terms of partitioning of SVOC species between the gas-phase and the surface film. Gas-phase SVOCs possessing lower values of octanol-air partition coefficients ( $K_{oa}$ ) equilibrate (by partitioning into these films) faster than those with larger  $K_{oa}$  values. Consequently, the fraction of SVOCs in surface films possessing larger  $K_{oa}$  values is expected to increase over time. We have formulated a mathematical model of the growth and evolution of organic films on impermeable surfaces. The formulation is based on a blended treatment of SVOCs in which species can be modeled as individual SVOCs or clustered into aggregate groupings. The groupings are organized around a *basis set* of  $\log K_{oa}$  values; this treatment has parallels to the volatility basis set concept that has been developed for understanding the partitioning of SVOCs between the gas and particle phase (Donahue et al., 2006). The model accounts for the coupled dynamics of film growth and changes in the surface abundance (mass/film area) and volumetric concentration (mass/film volume) for SVOCs sorbed into the film.

#### 3 Results and Discussion

The mathematical model predicts several features of organic film growth on impervious indoor surfaces that conform to published experimental observations. Predictions indicate that under typical indoor conditions, the organic constituents would be dominated by species with intermediate  $\log K_{oa}$  values ( $\sim 9.5$ - $12.5$ ).

Species with smaller  $\log K_{oa}$  values do not partition strongly to surface films from the gas phase; species with larger  $\log K_{oa}$  values approach equilibrium only slowly.

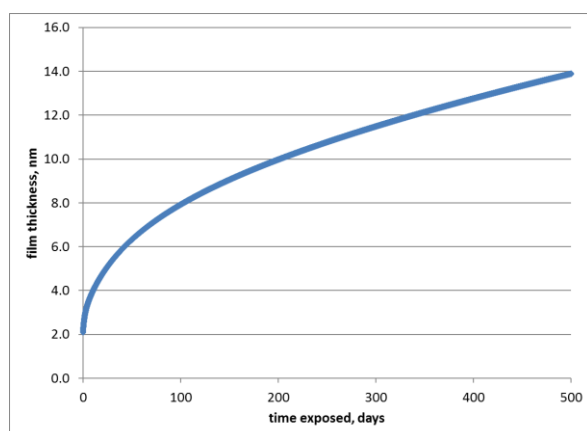


Figure 1: Evolution of the thickness of an organic surface film predicted using a multicomponent gas/surface film-partitioning model with initial film thickness of 2 nm.

Using estimates of typical indoor concentrations for SVOCs with  $\log K_{oa}$  values between 8 and 13, Figure 1 shows modeled organic film thickness versus time over a span of 500 days, at which time the thickness has grown from an initial 2 nm to 14 nm. The model predicts progressive enrichment of the film toward species with higher  $\log K_{oa}$  values. In the early stages of film growth ( $\sim 10$  h), the dominant SVOCs in surface films have  $\log K_{oa}$  values between 9 and 11; at 100 hours, between 10 and 11, and beyond 1000 hours, between 11 and 12.

Although the specifics will vary, the model indicates that organic surface films grow at a faster rate in the initial days and subsequently grow at a diminished and almost constant rate over time scales on the order of a hundred days. For common indoor conditions, the estimated growth rates are roughly  $0.05 - 0.1 \text{ nm day}^{-1}$  at 100 days and  $0.03 - 0.06 \text{ nm day}^{-1}$  at 200 days.

The above predictions are consistent with results reported for the organic surface films based on sampling from windows in actual indoor environments (Bennett et al. 2015; Huo et al. 2016; Li et al. 2010; Liu et al. 2003; Melymuk et al. 2016; Pan et al. 2012; Venier et al. 2016), from windows, plates and mirrors in a test house (Bi et al. 2015), and from Petri dishes and aluminum foils exposed to indoor air for different time intervals (Wallace et al. 2017).

## 4 Conclusions

Growing knowledge on this topic will contribute to improved understanding of multiple indoor processes – from surface-mediated indoor chemistry to dermal uptake following contact with organic-covered surfaces.

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