Title

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Journal

Environmental Science & Technology Letters, 8(5)

ISSN

2328-8930 2328-8930

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Publication Date

2021-04-22

DOI

10.1021/acs.estlett.1c00265

Peer reviewed

Intake fractions for volatile organic compounds in two occupied California residences

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ABSTRACT

Experimental estimates of residential intake fractions for indoor volatile organic compound (VOC) releases are scarce. We evaluated individual intake fractions (iF_i, mass inhaled by an individual per unit mass emitted) using ~five months of time-resolved VOC measurements acquired at two residences. First, we directly estimated iF_i using inert tracer gases that were released at fixed rates. Tracer gas iF_i values were generally consistent between occupants and comparable across seasons. Furthermore, iF_i for sources released on different floors of a residence were statistically indistinguishable, suggesting that source location within the living space was not strongly influential. Emissions from living space sources (iF_i ~ 0.3% = 3000 ppm) contributed to occupant exposures at rates 2–4 times higher than crawl space sources (iF_i ~ 1000 ppm) and >40 times higher than attic sources (iF_i < ~70 ppm). Second, we indirectly estimated iF_i for 251 VOCs using net emission rates estimated by indoor-outdoor material balance. Although emission patterns varied between compounds, all VOC-specific iF_i estimates were clustered near the values of the living space tracer gases. These experimental observations substantiate the theoretical expectation that iF_i values are largely independent of analyte characteristics, a useful simplification for exposure assessments.

INTRODUCTION

Mitigation strategies for limiting adverse health effects of air pollution often focus on identifying the specific sources that contribute the most to exposures.[1,2] One metric that can assist in such efforts is the intake fraction (iF), defined as a ratio: pollutant intake by a population (by inhalation, ingestion, or dermal absorption) normalized by total pollutant emissions from a given source or source category.[3] Intake fractions for air pollutants can vary by orders of magnitude.[4,5] Typical intake fraction values for outdoor pollutant releases range from half of a ppm (i.e., 0.5 μg pollutant inhaled per g emitted) to a few hundred ppm.[4] Typical values for indoor pollutant releases are much larger, ranging from hundreds of ppm to a few percent (1% = 10,000 ppm). These high intake fractions for indoor releases mainly reflect the much lower rate of per-occupant pollutant removal by ventilation indoors as compared with the effective per-capita rate of pollutant transport by wind outdoors.[6]

Various forms of the intake fraction concept have been used in life cycle assessments (LCAs) to enable rapid evaluations of the magnitude of human exposure to pollutants in consumer and commercial products.[7-14] Indoor intake fractions have been estimated using material balance models.[5,6,14,15] For volatile organic compounds (VOCs), these models indicate that intake fractions are principally controlled by air-change rates and by occupant behaviors.[5,6] Empirical determinations of VOC intake fractions are scarce. One study of this type calculated intake fractions from surveys of air-change rates and activity pattern surveys[16] that assess time spent at home.[17] A critical untested conclusion of prior studies is that intake fractions of VOCs are independent of analyte temporal behavior, such as whether emissions are baseline dominated or spike dominated, and physical properties such as vapor pressure. While a few studies have utilized time-resolved experimental measurements to evaluate particulate matter intake

fractions,[18-20] we were unable to identify any such experimental studies for VOC intake fractions in residences.

In this study, we report individual intake fractions (iF_i) via inhalation in two normally occupied residences in northern California as determined over three measurement campaigns. Intake fractions were assessed using chemical measurements from an online fast-response mass spectrometer, time-activity budgets from daily logs, and assumed standard breathing rates. To assess the importance of source location for non-reactive VOCs, we report iF_i for inert tracer gases that were released continuously at fixed rates in the living space, crawl space, and attic at each residence. To assess iF_i variability among diverse sources with different temporal behaviors and physical properties, we also report iF_i for 251 distinct VOCs sampled across three field campaigns using time-resolved emissions data derived by material balance. This study expands upon prior reports of speciated VOC emissions[21] (H1 only) and indoor exposures (H1 and H2).[22]

MATERIALS AND METHODS

Site description: The H1 and H2 monitoring campaigns have been extensively described;[21-24] a brief summary is provided here. Detailed measurements of airborne organic compounds and particulate matter were acquired at two normally occupied California residences in the East Bay region of the San Francisco Bay Area over three monitoring campaigns (H1 summer, H1 winter, H2 winter). The residences are single-family, wood-frame houses built in the late 1930s (H1) and early 1950s (H2) with approximately 180 m^2 of floor area each. The H1 site was a two-level residence, whereas the H2 site was a single-story house. The H1 and H2 residences were respectively occupied by two persons (adults designated H1M1 and H1F1) and four persons (two adults designated H2M1 and H2F1, a teenager, and an adult present only for a fraction of the campaign). During the campaigns, residents were encouraged to maintain their regular behavioral patterns with regard to indoor activities. Individual intake fractions were assessed for each of the two main adult occupants in each residence.

Study design: This work focuses on VOC measurements acquired by a proton-transfer reaction time-of-flight mass spectrometer (PTR-ToF-MS)[21-23] and daily occupant activity logs. At each residence, VOC concentrations were monitored at one outdoor and five indoor locations on a 5-minute rotating sample cycle yielding measurements at six locations once every 30 minutes. The five indoor locations were selected to characterize both the general living space (kitchen, bedroom hallway; living room for H2 only) as well as coupled unoccupied spaces (crawl space, attic; basement for H1 only). The H1 residence is constructed on two levels, with a kitchen, living room, and dining room situated toward the front (lower level) and a half flight of stairs connecting three bedrooms and two baths toward the back (upper level). The H1 basement is a single room beneath one bedroom. The remainder of the house has a crawl space beneath the

floor. Both the H1 and H2 residences contain an attic above the main living space. The H1 and H2 floorplans with PTR-ToF-MS sampling locations have been reported previously.[22,23] Daily logs yielded basic time-activity information ('awake', 'asleep', 'away') with roughly 5-minute time resolution for each occupant.

Three non-reactive deuterated VOCs, propene-d6, propene-d3, and butene-d3 ("tracer gases"), were simultaneously released at fixed emission rates at different locations within the residences. The release of tracer gases served 1) to estimate time-resolved ventilation rates and indoor airflow patterns and 2) to simulate the release of continuous indoor sources in different indoor zones. Two tracer gas release schemes were used over the three monitoring campaigns. In the first scheme, the three tracer gases were emitted into the crawl space, the living space, and the attic at the H1 and H2 site to study air-change rates and interzonal flows throughout the entire residence. In the second scheme, the tracer gases were deployed in the crawl space, lower living space, and upper living space (H1) or the crawl space, kitchen and living room (H2) to investigate interzonal transport and mixing within coupled living spaces.

Analysis and calculations: In this work, we assess the individual intake fraction (iF_i), i.e. the ratio of pollutant mass inhaled by a single occupant to pollutant mass emitted. Inhalation masses were estimated using chemical measurements from an online fast-response mass spectrometer (30-minute time resolution), time-activity budgets from daily logs (5-minute time resolution), and age-bracket-specific standard breathing rates taken from the United States Environmental Protection Agency Exposure Factors Handbook.[26]

Emitted masses were determined using both direct and indirect experimental designs. First, we use the known emission rates of tracer gases as "direct estimates." These can be interpreted as surrogates for general inert emission sources released at constant rates in the living

space, crawl space, and attic of the respective residences. Based on the direct estimate approach, we report iF_i estimates with daily time resolution in this study. Second, we use a material balance model[21] to determine net indoor VOC emissions for 251 compounds with different temporal behavior and physical properties at 2-hour time resolution as "indirect estimates." A representative subset of compounds spanning orders-of-magnitude in vapor pressure is reported in Table S1. We report campaign-average iF_i values in this study design. A limitation of both study designs is that concentrations were measured in stationary locations as opposed to human breathing zones. As such, proximity effects, especially during occupant activities such as cooking, may produce actual individual intake fractions that are underestimated for some compounds.[25] The magnitude of the proximity effect is expected to be unimportant for continuously released sources due to efficient mixing in the residence living space.[23] A detailed description of our approach is reported in the supporting information.

RESULTS AND DISCUSSION

We report iF_i values for the H1 summer, H1 winter, and H2 winter monitoring campaigns (Figs. 1, S1, S2). For each campaign, we report direct estimates of iF_i for tracer gases that were released at fixed rates in the living space, crawl space, or attic of the studied residences (Table 1). These values represent direct iF_i estimates for continuously released sources and can be interpreted as proxies for a generic source release in those locations. We also estimated indoor emission rates for the 251 unique VOCs observed over the three monitoring campaigns. Using these emissions data, we report campaign-averaged indirect iF_i estimates for all 251 VOCs (Figure 2).

Direct individual intake fraction (iF_i) estimates via controlled emissions.

Concentration time series of tracer sources, daily iF_i values, and time-resolved air-change rates are shown in Figure 1 for the H1M1 occupant during the H1 summer campaign. Similar data are shown for the H1 winter and H2 winter seasons in the SI (Figs. S1, S2). Changes in the concentration time series of the inert tracer gases are primarily associated with ventilation patterns, which are influenced by interzonal flows and the living zone air-change rate. The air-change rate is influenced by the extent to which doors and windows are open, indoor-outdoor temperature differences, and outdoor wind speed.[23] In the summer, a diurnal pattern is observed with enhanced ventilation during daytime periods leading to decreased tracer concentration. During summer nights periods, indoor/outdoor temperature differences are smaller, and doors and windows are more commonly closed, leading to enhanced tracer concentrations. The increase in iF_i from enhanced nighttime tracer concentrations is partially mitigated during sleeping hours owing to lower occupant breathing rates. Considering time varying ventilation

rates and the occupants' time-activity patterns, daily iF_i for the living space and crawl space tracer releases spanned factors of 7 and 6 in range, respectively.

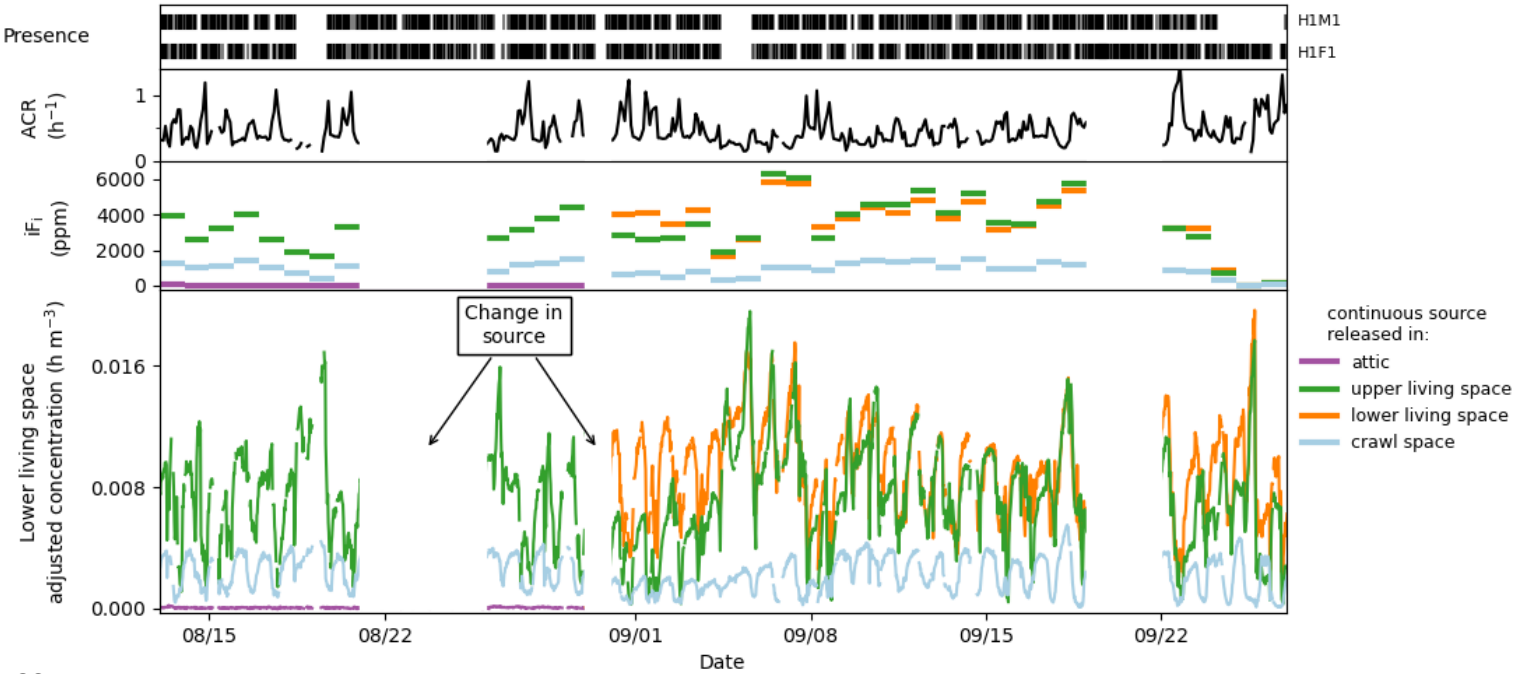


Figure 1. Time series of experimental data from study site H1 during the summer monitoring campaign. Times when occupants are present at the residence are marked in black in the uppermost panel. Time-resolved air-change rates are shown for the living space in the second panel. Daily individual intake fraction (iF_i) values for the continuously released tracer gas sources are shown for the H1M1 occupant in the third panel. The adjusted living room concentration (raw concentration [μg m^{-3}] divided by the mass release rate [μg h^{-1}]) of the continuously released tracer gas sources is shown in the bottommost source panel. Data proximate with changes to the tracer gas sources (location or release rate) are excluded.

In total, iF_i for living space sources were largely independent of source location, season, and occupant behavioral patterns. We highlight that iF_i for two sources released on different levels of the two-level H1 residence were statistically indistinguishable due to high rates of internal mixing (Table 1). This finding suggests that source location within the general living space is not a key determinant for occupant exposures at the H1 site, where all interior doors were intentionally left open during the monitoring campaigns. Similarly, we note that iF_i values were comparable between occupants at the H1 site and occupants at the H2 site. Male iF_i values were slightly larger than female iF_i values, primarily due to larger assumed inhalation rates. Male iF_i values are statistically indistinguishable from female iF_i values when identical inhalation rates are assumed. We stress that these results were obtained at only two sites with four occupants. If generalizable to the broader population, these findings suggest that temporal differences in occupant behavior may not be key determinants of iF_i values. Estimated iF_i values for the two living space sources were slightly higher in the H1 winter season than in the H1 summer season, largely due to lower air-change rates during the winter period.[23] Estimated iF_i of living space sources for the H2 winter season were slightly lower than those observed at the H1 summer and H1 winter seasons. While mean ventilation rates were comparable between the two sites (H1 summer = 160 $m^3\ h^{-1}$, H1 winter = 120 $m^3\ h^{-1}$, H2 winter = 170 $m^3\ h^{-1}$), ventilation rates during periods of occupancy were higher during the H2 winter campaign, leading to the discrepancy.

Table 1: Summary statistics (mean ± standard deviation) of daily individual intake fractions (iF_i) for tracer gases released in different locations during the H1 summer, H1 winter, and H2 winter campaigns.[a]

	H1 summer			H1 winter			H2 winter		
Source released in:	**sample size (days)**	**H1M1 iF_i (ppm)**	**H1F1 iF_i (ppm)**	**sample size (days)**	**H1M1 iF_i (ppm)**	**H1F1 iF_i (ppm)**	**sample size (days)**	**H2M1 iF_i (ppm)**	**H2F1 iF_i (ppm)**
crawl space	*n* = 36	900 (± 400)	800 (± 200)	*n* = 25	1100 (± 300)	900 (± 200)	*n* = 27	1300 (± 400)	900 (± 300)
living space[b]	*n* = 24	3500 (± 1500)	3100 (± 900)	*n* = 25	3900 (± 1100)	3200 (± 800)	*n* = 27	2400 (± 800)	1700 (± 700)
upper living space	*n* = 36	3300 (± 1500)	2800 (± 900)	*n* = 6	3400 (± 900)	3000 (± 500)	-	-	-
attic	N/A [c]	< 30	<30	N/A [c]	< 80	< 70	N/A [c]	< 70	< 60

[a] The number of daily determinations for each category is *n*.

[b] The "living space" refers to the "lower living space" at the two-level H1 residence and the "general living space" at the single story H2 residence.

[c] An upper bound campaign-average iF_i value is reported in place of summary statistics for daily iF_i values.

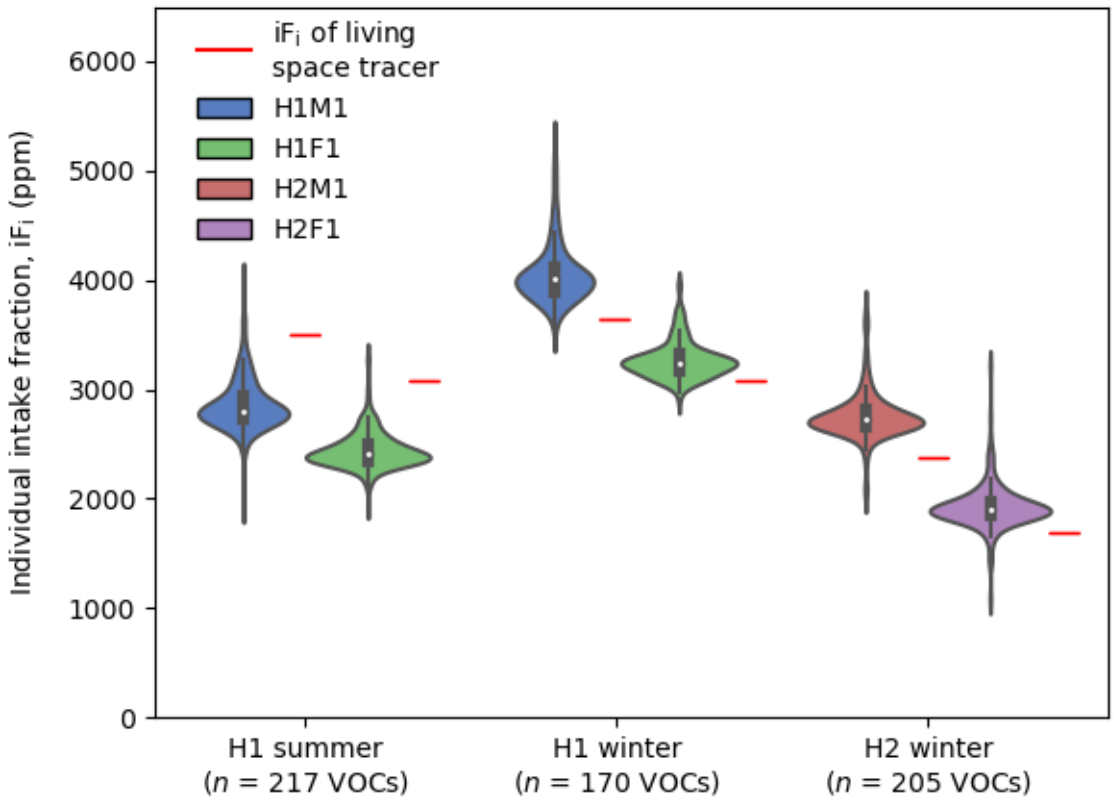
Emissions into the general living space were considerably more impactful for occupant exposures than emissions in hidden coupled spaces. At the H1 and H2 sites, sources released in living spaces (iF_i = 1700–3900 ppm) reached human receptors at average rates 2–4 times higher than crawl space sources (700–1300 ppm) and >40 times higher than attic sources (iF_i < ~70 ppm, season dependent). The attic tracer was often near or below the limit of detection (defined as 3.3 sigma of blank measurements). During calculations of iF_i, we replaced non-detect measurements with the limit of detection value; attic iF_i values therefore represent an upper bound estimate. Although the air-change rate during occupancy was higher at the H2 site than at the H1 site, the H2 crawl space source entered the living space at higher efficiency leading to a minor increase in iF_i for crawl-space emissions at H2 as compared to H1.

To contextualize iF_i values reported in this work, we note that the time-averaged population intake fraction for distributed ground-level outdoor pollutant releases in the San Francisco Bay Area is estimated to be 38 parts per million.[4] That value, which accounts for inhalation intake of non-reactive airborne pollutants by the entire urban population, is roughly 100 times smaller than iF_i values reported in this work for inhalation intake by individuals for pollutant releases within the normally occupied space of their residences. As theoretically anticipated,[1,2] indoor pollutant releases contribute much more to exposures than do outdoor pollutant releases per unit mass emitted.

We also highlight that first-order approximations are comparable to the time-resolved estimates provided in this work.[5] Using mean ventilation rates for the H1 summer (160 $m^3\ h^{-1}$), H1 winter (120 $m^3\ h^{-1}$), and H2 winter (170 $m^3\ h^{-1}$) campaigns, age-specific daily breathing rates of 14.2 $m^3\ d^{-1}$ (H1) and 15.7 $m^3\ d^{-1}$ (H2),[26] and a population average of 69% time spent in a residence,[16] the first-order estimate of iF_i values at the three residences would be 2600 ppm,

3400 ppm, and 2700 ppm, respectively. These estimates based on theoretical expectations are within 30% of the direct experimental estimates determined from controlled tracer release. We note that the mean ventilation rates for this analysis were derived via material balance of the same time-resolved tracer gas concentrations as the "direct estimate" approach. Therefore, the resemblance between the time-resolved "direct estimate" approach and the time-averaged "first-order approximation" approach is partially attributable to methodological overlap.

Indirect individual intake fraction (iF_i) estimates for VOCs. Time-resolved indoor emission rates were evaluated by material balance for 251 distinct VOCs observed in the H1 summer, H1 winter, and H2 winter campaigns. These VOCs originated from diverse source processes, including continuous emissions from the building and its contents, episodic emissions from occupants and their activities such as cooking, cleaning, use of personal care products,[21] and indoor chemistry,[27] resulting in substantial variability in mean concentration, temporal variability, and analyte physical properties. The emissions data were used to estimate the iF_i values as summarized graphically in Figure 2. The iF_i values tended to cluster and were comparable to values more directly estimated by tracer release.



229

230 **Figure 2.** Mean individual intake fraction (iF_i) values for all observed VOCs during the H1
231 summer (217 compounds), H1 winter (170 compounds), and H2 winter (205 compounds)
232 measurement campaigns are summarized in violin plots for two occupants in each house (H1M1,
233 H1F1, H2M1, H2F1). The expected value as determined experimentally by continuous tracer-
234 release is shown in red. Values corresponding to $C_7H_4ClF_2^+$, a compound of predominantly
235 outdoor origin, are not shown.

A key assumption in some exposure assessment methods is that intake fraction values are largely analyte independent, as theory predicts.[5] Assuming that this assumption is correct, exposures in microenvironments with known emission patterns can be estimated using compound-independent intake-fraction values. In congruence with theoretical expectations, our results provide direct empirical evidence that the temporal behavior of a source (as for instance, a continuously released emission from the building or an episodically released emission from an occupant activity) does not strongly affect iF_i for a broad suite of mainly organic compounds measured by PTR-ToF-MS (Figure S3). Limitations of this work include that it did not conduct personal monitoring or consider the influence of variable occupant breathing rates beyond a binary awake/asleep distinction. The approach used in this work would also underestimate emissions for strongly sorbing compounds or for compounds that undergo chemical degradation at fast time scales in the indirect experimental design, resulting in an overestimate of the respective iF_i value. These latter influences are expected to be minor. Another important consideration is the small sample size — only two households and four occupants were considered in this study. Additional work is needed to demonstrate applicability of the results to larger populations.

This study reports individual intake fraction (iF_i) values in two normally occupied residences via time-resolved VOC measurements. We find that iF_i values for VOC sources within the living space were consistent with past estimates and largely independent of source location, occupant behavioral patterns, and season. However, iF_i values for sources in the crawl space and especially for sources in the attic were smaller. Ultimately, this work corroborates theoretical expectations that indoor inhalation intake fractions are principally influenced by the ventilation patterns and generally independent of analyte characteristics.

Supporting Information: Data analysis procedures, tracer gas iF_i during H1 winter campaign, tracer gas iF_i during H2 winter campaign, impact of episodic emissions on iF_i, iF_i data table for selected compounds.

Acknowledgements: The H1 and H2 occupants gave informed consent for this study, which was conducted under a protocol approved in advance by the Committee for Protection of Human Subjects for the University of California, Berkeley (Protocol #2016 04 8656). The authors thank the H1 and H2 occupants for cooperation in allowing their homes to be studied and in maintaining daily log sheets of occupancy and activities. We thank Robin Weber for technical assistance. This work was supported by the Alfred P. Sloan Foundation Program on Chemistry of Indoor Environments via Grants 2016-7050 and 2019-11412. D. L. acknowledges support from the National Science Foundation (Grant No. DGE 1752814). K. K. acknowledges support from the Carlsberg Foundation (Grant No. CF16-0624). The authors declare no competing financial interests.

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