Intake Fraction for the Indoor Environment: A Tool for Prioritizing Indoor Chemical Sources

Hyeong-Moo Shin,*† Thomas E. McKone,‡ and Deborah H. Bennett†

†Department of Public Health Sciences, University of California—Davis, Davis, California, United States
‡School of Public Health, University of California—Berkeley, Berkeley, California, United States

Supporting Information

ABSTRACT: Reliable exposure-based chemical characterization tools are needed to evaluate and prioritize in a rapid and efficient manner the more than tens of thousands of chemicals in current use. This study applies intake fraction (iF), the integrated incremental intake of a chemical per unit of emission, for a suite of indoor released compounds. A fugacity-based indoor mass-balance model was used to simulate the fate and transport of chemicals for three release scenarios: direct emissions to room air and surface applications to carpet and vinyl. Exposure through inhalation, dermal uptake, and nondietary ingestion was estimated. To compute iF, cumulative intake was summed from all exposure pathways for 20 years based on a scenario with two adults and a 1-year-old child who ages through the simulation. Overall iFs vary by application modes: air release (3.1 × 10⁻³ to 6.3 × 10⁻³), carpet application (3.8 × 10⁻³ to 6.2 × 10⁻³), and vinyl application (9.0 × 10⁻³ to 1.8 × 10⁻²). These iF values serve as initial estimates that offer important insights on variations among chemicals and the potential relative contribution of each pathway over a suite of compounds. The approach from this study is intended for exposure-based prioritization of chemicals released inside homes.

1. INTRODUCTION

Over the last several decades, there have been significant increases in the volume of chemicals used in the manufacture of residential consumer products, including plastics, clothing, electronics, and cosmetics.¹ Not only the volume of chemicals produced but also the number and type of chemicals have increased. However, information about potential exposure and adverse health effects in humans from residential uses is limited for most chemicals. Thus, the development of both toxicity- and exposure-based assessments is needed to evaluate and prioritize chemicals in a rapid and efficient manner.²,³

Approaches for systematically prioritizing chemicals require human exposure data, product use information, and modeled human behavior and must address both near-field (from consumer products used indoors) and far-field (from overall environmental dispersion) exposures.¹,³ However, most current prioritization approaches focus mainly on far-field exposure and their output impact metrics are not standardized.¹

Because individuals generally spend over 70% of their time indoors at home,⁴ both the residence times of chemicals in the indoor environment and residential human activity patterns are key inputs to chemical prioritization. Several studies report that the concentrations of volatile organic compounds (VOCs) are consistently higher indoors than outdoors.⁵,⁶ Moreover, the levels of pesticides found most frequently in carpet dust tend to be higher than those in outdoor soil, and many pesticides have been found to be much more persistent in the indoor environment relative to outdoors.⁷ For less volatile and more persistent compounds, dermal uptake or nondietary ingestion pathways are additional routes of exposure for young children.⁸ These factors all strengthen the need for quantifying exposures for compounds released to the indoor environment.

Intake fraction (iF) is defined as the integrated incremental intake of a pollutant per unit of emission,⁹,¹⁰ and we consider it a useful concept for expressing source-to-intake relationships. Because iF is unitless, it is considered a convenient standardized metric of exposure. A number of researchers have applied iF to particulate matter (PM) and other types of emissions from power plants and mobile sources to determine the relationship among emissions, human intake, and health impacts.¹¹,¹² Moreover, iF has been calculated for over 300 organic chemicals released into ambient air and water to examine the relationship between chemical properties and primary exposure routes outdoors.⁹

Three studies estimated iF through inhalation of PM from indoor sources¹¹,¹３ and pollutants from episodic indoor emissions.¹⁴ However, no other studies to date have included dermal and nondietary exposure pathways to quantify iF values over a wide range for organic compounds found in the indoor environment. Several studies report that exposure through incidental dust ingestion and dermal contact is important in predicting polybrominated diphenyl ether (PBDE) and pesticide body burdens,¹⁵—¹⁹ indicating the importance of

Received: May 7, 2012
Revised: August 17, 2012
Accepted: August 24, 2012
Published: August 25, 2012
these pathways. Quantifying exposure through dermal and nondietary pathways requires a number of assumptions regarding both the primary mechanisms for exposure and parameter values associated with quantifying intake from these mechanisms.

The objectives of this study are to (1) estimate iF for a suite of organic compounds from residential exposure via three release scenarios (air emission and carpet and vinyl flooring applications), (2) examine the relationship between intake through different exposure pathways and chemical properties by varying the magnitude of chemical volatility, and (3) address the data and model gaps and their impact for estimating dermal and nondietary ingestion exposure.

2. METHODS

2.1. Overview. Calculating iF for chemicals released to an indoor residence requires a number of steps. First, the release scenario is defined. Second, the resulting fate and transport of the chemical through multiple indoor compartments is determined by use of an indoor fugacity model. Third, exposure concentrations are calculated from the mass associated with any compartment. Fourth, exposure over time is calculated via multiple exposure models. Finally, the integrated incremental intake of a pollutant per unit of emission, iF, is calculated.

Calculating iF, rather than predicting exposure from a broad range of specific source input scenarios, allows simplified release scenarios because the magnitude of iF is not strongly dependent on the details of the release scenario beyond the release compartment. This is critical if the model is to be used to screen chemicals. Compounds were compared on the basis of a pulse-release (i.e., one-time release at time = 0) into three primary household media: air, carpet, and vinyl. A pulse-release may not capture all of the details of different modes of application for the chemicals modeled, but it allows chemical prioritization when calculating iF. For example, if a constant number of individuals is assumed, for a continuous airborne release of a volatile compound, such as naphthalene from a mothball, iF is the same as for a pulse input. When chemicals are screened in a prioritization framework, it is unlikely that the emission scenario would be known, particularly if the chemical was assessed prior to its use in commerce. This model can also be used to predict time-varying exposures if the emission profile in the indoor residential environment is known.

Ideally, a detailed quantitative uncertainty assessment should be applied to the model results to account for uncertainty in chemical property values, variability and uncertainty in housing characteristics, and model uncertainty resulting from various assumptions related to dermal and nondietary exposure pathways. Insufficient information exists to reliably quantify the uncertainty of parameter input values, and there are vast differences between various models available for dermal and nondietary exposure. Therefore, we focused on relatively large modeling uncertainty by determining the sensitivity of results to the selection of two alternative dermal and nondietary models.

2.2. Indoor Fate and Transport Model. 2.2.1. Model Description and Input Parameters. We used a fugacity-based mass-balance model to simulate the fate and transport of compounds in the indoor environment\textsuperscript{21} (see Supporting Information for summary of the model). The model includes four compartments that capture the major indoor reservoirs for chemicals: air, carpet, vinyl flooring, and walls. Each compartment in the model is composed of multiple phases, such as gases and particles in the air compartment. We used multiple size fractions of particles, including 0–2.5, 2.5–10, and 10–150 \( \mu m \), because of the unique physical properties of each particle size fraction. We assumed that each phase in a given compartment was in chemical equilibrium with all other phases in that compartment. However, the compartments are not required to be in chemical equilibrium with each other because there are driving diffusive mass transfers between compartments. Specifically, a compound can partition to carpet after being released to air and then slowly volatilize back out into the air. Since semivolatile organic compounds (SVOCs) tend to partition to dust, advective transport pathways of dust, including resuspension and deposition, were considered. A compound can be removed from the indoor system through ventilation of gas and particle phases or through surface cleaning. A set of differential equations accounts for gains and losses in each compartment as well as transfers between compartments. Input parameters and the properties of particles in different size fractions are listed in Tables S1 and S2 of the Supporting Information. In applying the model, we first defined

### Table 1. Physicochemical Properties of 15 Studied Chemicals

| Chemical                  | MW (g/mol) | \( K_{ow} \) | VP (Pa·m\(^3\)/mol) | \( H^d \) (Pa·m\(^3\)/mol) | \( D_{aw} \) (m\(^2\)/day) | \( k_{OH} \) (cm\(^3\)/mol·day) |
|---------------------------|------------|---------|-----------------|----------------|-----------------|----------------|-----------------|
| formaldehyde              | 30.0       | 2.2     | 4.6 \times 10\(^2\) | 7.0 \times 10\(^1\) | 1.4             | 8.1 \times 10\(^{-7}\) |
| naphthalene               | 128.2      | 2.3 \times 10\(^1\) | 2.1 \times 10\(^1\) | 1.9 \times 10\(^2\) | 5.2 \times 10\(^{-1}\) | 1.9 \times 10\(^{-4}\) |
| diethyl phthalate         | 222.2      | 5.1 \times 10\(^2\) | 2.2 \times 10\(^1\) | 9.3 \times 10\(^{-1}\) | 2.3 \times 10\(^{-1}\) | 3.0 \times 10\(^{-2}\) |
| galaxolide                | 238.2      | 1.1 \times 10\(^2\) | 5.5 \times 10\(^{-2}\) | 5.9 \times 10\(^{-3}\) | 3.0 \times 10\(^{-1}\) | 1.9 \times 10\(^{-4}\) |
| tris(2-chloroethyl) phospate| 284.0    | 3.0 \times 10\(^1\) | 1.4 \times 10\(^{-2}\) | 5.5 \times 10\(^{-4}\) | 2.1 \times 10\(^{-1}\) | 2.3 \times 10\(^{-4}\) |
| diazinon                  | 304.0      | 5.9 \times 10\(^1\) | 9.4 \times 10\(^{-2}\) | 1.3 \times 10\(^{-1}\) | 1.8 \times 10\(^{-1}\) | 1.9 \times 10\(^{-4}\) |
| chlorpyrifos              | 350.6      | 1.0 \times 10\(^1\) | 2.2 \times 10\(^{-3}\) | 4.7 \times 10\(^{-1}\) | 3.3 \times 10\(^{-1}\) | 7.9 \times 10\(^{-6}\) |
| PBDE-47                   | 485.8      | 4.8 \times 10\(^1\) | 5.6 \times 10\(^{-2}\) | 1.1             | 2.7 \times 10\(^{-1}\) | 2.3 \times 10\(^{-4}\) |
| bis(2-ethylhexyl) phthalate| 390.5    | 3.3 \times 10\(^1\) | 5.3 \times 10\(^{-2}\) | 1.8             | 1.5 \times 10\(^{-1}\) | 1.9 \times 10\(^{-4}\) |
| triphenyl phosphate       | 326.1      | 3.9 \times 10\(^1\) | 1.7 \times 10\(^{-3}\) | 7.5 \times 10\(^{-3}\) | 5.0 \times 10\(^{-1}\) | 2.3 \times 10\(^{-4}\) |
| butyl benzyl phthalate    | 312.0      | 8.1 \times 10\(^1\) | 9.5 \times 10\(^{-3}\) | 3.9 \times 10\(^{-3}\) | 1.5 \times 10\(^{-1}\) | 9.5 \times 10\(^{-4}\) |
| PBDE-99                   | 564.7      | 2.0 \times 10\(^1\) | 3.3 \times 10\(^{-2}\) | 3.0 \times 10\(^{-1}\) | 1.9 \times 10\(^{-1}\) | 2.3 \times 10\(^{-4}\) |
| permethrin                | 391.3      | 4.4 \times 10\(^1\) | 9.9 \times 10\(^{-2}\) | 1.0 \times 10\(^{-2}\) | 1.7 \times 10\(^{-1}\) | 2.8 \times 10\(^{-4}\) |
| benzo[a]pyrene            | 252.3      | 1.5 \times 10\(^1\) | 2.5 \times 10\(^{-3}\) | 3.4 \times 10\(^{-2}\) | 4.1 \times 10\(^{-1}\) | 4.3 \times 10\(^{-4}\) |
| PBDE-209                  | 959.2      | 2.8 \times 10\(^1\) | 2.2 \times 10\(^{-2}\) | 1.5 \times 10\(^{-3}\) | 1.7 \times 10\(^{-1}\) | 2.3 \times 10\(^{-4}\) |

\*Molecular mass. \*Octanol–water partition coefficient (unitless). \*Vapor pressure. \*Henry’s law constant. \*Diffusion coefficient in pure air. \*OH radical reaction rate constant.
the fugacity capacity for each compartment, including all phases (Table S3, Supporting Information) and transfer factors between compartments (Table S4, Supporting Information). Then, we solved ordinary differential equations to compute the time-dependent mass in each compartment from the mass balance equations (Table S5, Supporting Information) using MATLAB (Mathworks, Natick, MA).

2.2.2. Chemical Release Scenarios. We selected to study 15 chemicals based on concern for their effects on human health and their large range in vapor pressure values (see Table 1 for chemical properties). The selected chemicals were the following: formaldehyde, naphthalene, galaxolide (a synthetic musk), benz[a]pyrene, three pesticides commonly found in the indoor environment (diazinon, chlorpyrifos, and permethrin), three common polybrominated diphenyl ether (PBDE) congeners as flame retardants (PBDE-47, PBDE-99, and PBDE-209), two flame retardants newly detected in houses [tris(2-chloroethyl) phosphate and triphenyl phosphate], and three plasticizers [diethyl phthalate, bis(2-ethylhexyl) phthalate, and butyl benzyl phthalate]. We assumed that each chemical was emitted or applied as a pulse—release to the air, carpet, and vinyl compartments, and we computed time-dependent concentration profiles for 20 years.

2.3. Exposure Model. We chose a representative exposure scenario, consisting of two adults and a 1-year-old child who ages through the simulation, based on U.S. Census Bureau data published in 2011, in which the average household was reported to consist of 1.95 adults and 0.63 children.22 In our exposure scenario, the 1-year-old child is tracked for 20 years, during which age-specific exposure factors, including inhalation uptake rates, hand-to-mouth frequencies, and surface area of hands, change over time. A conservative assumption was made that the occupants were present indoors when the chemicals were released and that both adults and the child were exposed through inhalation and dermal uptake. We estimated exposure through nondietary ingestion only for the young child, as the frequency of mouthing activities after surface contact is considerably lower for adults, although we note that this may underestimate adult exposure.23

2.3.1. Inhalation Exposure. For each individual, daily intake (micrograms per day) via inhalation is based on the following equation:

\[ I_{inh} = (C_g + C_p)R_{inh}\frac{f_{res}}{f_{res}} \]

where \( C_g \) and \( C_p \) are the exposure concentrations in the gas and particle phases, respectively, of the air compartment (micrograms per cubic meter), \( R_{inh} \) is the inhalation uptake rate (cubic mass per day), and \( f_{res} \) is the fraction of time spent at the residence (unitless).

For inhalation of air particles, we included chemical mass only for particles less than 10 µm in diameter.24 Human activities, such as walking, folding blankets, and vacuuming, can resuspend dust from surface compartments. Ferro et al.25 measured the mean PM2.5 and PM10 concentrations for the three activities mentioned above and reported that personal exposures to PM2.5 and PM10 were respectively 1.4 and 1.6 times as high as direct exposure to average indoor concentrations. Thus, we accounted for both resuspended particles from human activities and the fraction of human activity time at residence in computing exposure through particle inhalation. We estimated the fraction of human activity time at residence by subtracting the fraction of time spent sleeping/napping (\( f_{slp} \)) from the fraction of time spent at residence (\( f_{res} \)). The age-specific values for \( f_{slp} \) and \( f_{res} \) were obtained from the U.S. Environmental Protection Agency (EPA) Exposure Factors Handbook (EFH)26 and are listed in Table 2. The average inhalation uptake rates for male and female adults (Table 3) were applied from the EFH. Those for children under 12 years of age were applied from the Child-Specific Exposure Factors Handbook (CSEFH).23

### Table 2. Age-Specific Exposure Parameter Values for Inhalation and Nondietary Ingestion

<table>
<thead>
<tr>
<th>age (years)</th>
<th>inhalation rate(^a) (m(^3)/day)</th>
<th>hand-to-mouth frequency(^a) (events/day)</th>
<th>object-to-mouth frequency(^a) (events/day)</th>
<th>surface area of hands(^b) (m(^2))</th>
<th>fraction of time spent at residence(^c)</th>
<th>fraction of time sleeping and napping(^d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>5.4</td>
<td>190</td>
<td>240</td>
<td>0.024</td>
<td>0.77</td>
<td>0.53</td>
</tr>
<tr>
<td>1</td>
<td>8.0</td>
<td>200</td>
<td>100</td>
<td>0.030</td>
<td>0.74</td>
<td>0.54</td>
</tr>
<tr>
<td>2</td>
<td>8.9</td>
<td>130</td>
<td>100</td>
<td>0.028</td>
<td>0.68</td>
<td>0.49</td>
</tr>
<tr>
<td>3</td>
<td>10.1</td>
<td>150</td>
<td>100</td>
<td>0.037</td>
<td>0.66</td>
<td>0.47</td>
</tr>
<tr>
<td>4</td>
<td>10.1</td>
<td>150</td>
<td>100</td>
<td>0.037</td>
<td>0.66</td>
<td>0.47</td>
</tr>
<tr>
<td>5</td>
<td>10.1</td>
<td>150</td>
<td>100</td>
<td>0.037</td>
<td>0.66</td>
<td>0.47</td>
</tr>
<tr>
<td>6</td>
<td>12.0</td>
<td>70</td>
<td>10</td>
<td>0.051</td>
<td>0.62</td>
<td>0.43</td>
</tr>
<tr>
<td>7</td>
<td>12.0</td>
<td>70</td>
<td>10</td>
<td>0.051</td>
<td>0.62</td>
<td>0.43</td>
</tr>
<tr>
<td>8</td>
<td>12.0</td>
<td>70</td>
<td>10</td>
<td>0.051</td>
<td>0.62</td>
<td>0.43</td>
</tr>
<tr>
<td>9</td>
<td>12.0</td>
<td>70</td>
<td>10</td>
<td>0.051</td>
<td>0.62</td>
<td>0.43</td>
</tr>
<tr>
<td>10</td>
<td>12.0</td>
<td>70</td>
<td>10</td>
<td>0.051</td>
<td>0.62</td>
<td>0.43</td>
</tr>
</tbody>
</table>


### Table 3. Exposure Parameter Values

<table>
<thead>
<tr>
<th>parameter</th>
<th>symbol</th>
<th>child</th>
<th>adult</th>
</tr>
</thead>
<tbody>
<tr>
<td>inhalation uptake rate (m(^3)/day)</td>
<td>( R_{inh} )</td>
<td>15.5</td>
<td>26</td>
</tr>
<tr>
<td>fraction of time spent at residence</td>
<td>( F_{res} )</td>
<td>0.65</td>
<td>26</td>
</tr>
<tr>
<td>fraction of time sleeping/napping</td>
<td>( F_{slp} )</td>
<td>0.36</td>
<td>26</td>
</tr>
<tr>
<td>transfer efficiency of carpet-to-hand</td>
<td>( T_{E_c} )</td>
<td>0.034</td>
<td>33</td>
</tr>
<tr>
<td>transfer efficiency of vinyl-to-hand</td>
<td>( T_{E_v} )</td>
<td>0.033</td>
<td>36</td>
</tr>
<tr>
<td>transfer efficiency of hand-to-mouth</td>
<td>( T_{E_{h-m}} )</td>
<td>0.5</td>
<td>40</td>
</tr>
<tr>
<td>transfer efficiency of object-to-mouth</td>
<td>( T_{E_{o-m}} )</td>
<td>0.5</td>
<td>43</td>
</tr>
<tr>
<td>surface area of hands contacted with surface or object (m(^2)/event)</td>
<td>( S_{A_h} )</td>
<td>0.098</td>
<td>26</td>
</tr>
<tr>
<td>surface area of object or hand that is mouthed (m(^2)/event)</td>
<td>( S_{A_o} )</td>
<td>0.005</td>
<td>45</td>
</tr>
<tr>
<td>exposure frequency of hand-to-carpet (events/h)</td>
<td>( E_{F_{hc}} )</td>
<td>11</td>
<td>34, 36, 38</td>
</tr>
<tr>
<td>exposure frequency of hand-to-vinyl (events/h)</td>
<td>( E_{F_{hv}} )</td>
<td>12</td>
<td>34, 36, 38</td>
</tr>
<tr>
<td>exposure frequency of hand-to-object (events/h)</td>
<td>( E_{F_{ho}} )</td>
<td>115</td>
<td>35, 36, 37</td>
</tr>
<tr>
<td>fraction absorbed through skin</td>
<td>( A_{B_k} )</td>
<td>0.02</td>
<td>44</td>
</tr>
<tr>
<td>fraction absorbed through gut</td>
<td>( A_{B_g} )</td>
<td>0.70</td>
<td>41</td>
</tr>
</tbody>
</table>
resulting equations for exposure concentrations for all compartments are listed in Table S6 of the Supporting Information.

2.3.2. Dermal Exposure. There are multiple methods available to estimate intake through dermal exposure. Cohen Hubal et al.\textsuperscript{27} integrated dermal uptake based on a series of discrete transfers from each contact with a contaminated medium. Weschler and Nazaro\textsuperscript{28} estimated air-to-skin transdermal uptake of SVOCs by using two permeability coefficients: (1) skin lipid and (2) indoor air transdermal. Zartarian et al.\textsuperscript{6} developed the residential stochastic human exposure and dose simulation model for pesticides (residential-SHEDS) to estimate dermal exposure and dose by use of microlevel time—location—activity information.

We selected to use both the Cohen Hubal and Weschler and Nazaro models for calculating dermal exposure and comparing the results as a sensitivity analysis. The Cohen Hubal model served as the baseline approach. Because the stochastic approach required to use SHEDS makes it difficult to evaluate pathway contributions over long periods of time, and our goal is to provide a screening-level assessment model, we elected not to include the SHEDS model in our final comparison among alternative modeling approaches. The details for the Cohen Hubal model are presented below, while those for the Weschler and Nazaro model are presented in the Supporting Information.

We used the following equation to estimate the intake (micrograms per day) from dermal exposure using the “microactivity” approach from the model of Cohen Hubal et al.\textsuperscript{27}

\[
I_{dem} = C_i(SA)(EF)(TE_i)(ABS_i)
\]

(2)

where \(C_i\) is the exposure concentration on surface \(i\) (carpet or vinyl) (micrograms per square meter), \(SA\) is the surface area of the hand contacted (square meters per event), \(EF\) is the exposure frequency of contacts per day for surface \(i\) (events/day), \(TE\) is the efficiency of the contaminant transferring from surface \(i\) to the hand (unitless), and \(ABS\) is the fraction of mass absorbed through skin (unitless).

For the exposure concentration associated with the carpet compartment (\(C_c\)), we assumed that only dust was transferred from carpet and that only a portion of the contaminant residue in that dust was available for transfer to the skin. Dislodgable fractions of compounds from carpet are often measured by using a polyurethane foam (PUF) roller.\textsuperscript{29} Thus, we multiplied chemical concentrations on carpet dust by the average value of dislodgeable fraction (1.5%) as measured by a PUF roller from Nishioka et al.\textsuperscript{30} and Fortune et al.\textsuperscript{31} For the exposure concentration associated with the vinyl compartment (\(C_v\)), we assumed that only dust was transferred from vinyl and that only a portion of the contaminant residue in that dust was available for transfer to the skin. Dislodgable fractions of compounds from vinyl are often measured by using a polyurethane foam (PUF) roller.\textsuperscript{29} Thus, we multiplied chemical concentrations on vinyl dust by the average value of dislodgeable fraction (1.5%) as measured by a PUF roller from Nishioka et al.\textsuperscript{30} and Fortune et al.\textsuperscript{31}

For the exposure concentration associated with the carpet compartment (\(C_c\)), we estimated the average frequency of contacts over one day for carpet, vinyl floor, and objects from five published studies\textsuperscript{34−38} to obtain the values 11, 12, and 115 events/day, respectively, for use in our model (see Supporting Information, Table S7). Although the transfer efficiency from surfaces to human skin is chemical-specific and influenced by other factors, including condition of skin on the palm, we used the average transfer efficiency of chlorpyrifos from carpet to hand (\(TE_c = 3.35\%\)) from Lu and Fenske\textsuperscript{33} and Cohen Hubal et al.\textsuperscript{39} as our default value for carpet and the average of all analytes from vinyl flooring to hand (\(TE_v = 3.15\%\)) from Clothier\textsuperscript{40} and Cohen Hubal et al.\textsuperscript{39} as our default value for vinyl. For the absorption fraction through skin (\(ABS\)), we assumed that 2% of the chemical in soil or dust was absorbed through skin.\textsuperscript{41}

In addition to carpet and vinyl flooring, children contact numerous other objects such as toys and furniture, which can contain the chemicals selected for this study. There has been little work to determine the relationship between the concentration on carpet/vinyl surface compartments and the surface concentration on other objects. The existing work is difficult to interpret because toy concentrations were based on an extraction technique rather than a wipe sample.\textsuperscript{42} In this study, we assumed that the concentrations on the surface of other objects and the transfer efficiency from other objects to hands can be represented by area-weighted values of carpet and vinyl surfaces, acknowledging that this is an uncertain parameter.

2.3.3. Nondietary Ingestion Exposure. As with dermal exposure, there are multiple methods for calculating nondietary ingestion. Cohen-Hubal et al.\textsuperscript{27} integrated nondietary uptake based on a series of discrete transfers from hand- and object-to-mouth activities. Ozkaynak et al.\textsuperscript{43} estimated 68 mg/day soil and dust ingestion rate via hand and object mouthing, using the residential-SHEDS model. This ingestion rate is similar to the EPA’s recommended soil and dust ingestion rate (= 60 mg/day) for children.\textsuperscript{53} Thus, chemical intake through soil and dust ingestion can be estimated by multiplying soil/dust ingestion rate (milligrams per day) by chemical concentrations in soil/dust (micrograms of chemical/milligram of dust and soil).\textsuperscript{44} We calculated nondietary exposure using both the Cohen Hubal and Ozkaynak models for comparison, with the Cohen Hubal model as the baseline approach for nondietary exposure.

We considered two nondietary ingestion pathways: object-to-mouth and hand-to-mouth. In both cases, the equation to estimate the daily intake (micrograms per day) from nondietary ingestion is based on the approach from the model of Cohen Hubal et al.:\textsuperscript{27}

\[
I_{nd} = C_x(SA_x)(EF)(TE_x)(ABS_x)
\]

(3)

where \(x\) is the hand or object that is mouthed, \(C_x\) is the exposure concentration on hand or object placed in the mouth (micrograms per square meter), \(SA_x\) is the surface area of hand or object that is mouthed (square meters per event), \(EF\) is the exposure frequency of the mouthing event for the hand or object (events/day), \(TE_x\) is the transfer efficiency from hand or object to mouth (unitless), and \(ABS_x\) is the fraction of mass absorbed through gut (unitless).

For the exposure concentration on hand (\(C_h\)), we multiplied area-weighted exposure concentrations on carpet (\(C_c\)) or vinyl (\(C_v\)) by transfer efficiencies from carpet to hand (\(TE_c\)) or vinyl to hand (\(TE_v\)). For the exposure concentration on objects (\(C_o\)), we used area-weighted exposure concentrations on carpet or vinyl because objects are inserted directly into the mouth. We assumed that the surface area of an object that is mouthed was...
the same as the surface area of a finger (i.e., 10% of hand) that is mouthed.\textsuperscript{45} We assumed that the transfer efficiency from hand or object to mouth (TE\textsubscript{m}) was 0.5, acknowledging that this is an uncertain parameter.\textsuperscript{45} For the absorption fraction through gut (ABS\textsubscript{g}), we assumed that 70% of an administered dose was absorbed through the gut.\textsuperscript{44} For children under 12 years of age, we obtained age-specific exposure parameters, including hand-to-mouth frequency, object-to-mouth frequency, and the surface area of hand (Table 3) from the CSEFH.\textsuperscript{23}

3. RESULTS AND DISCUSSION

3.1. Exposure Profile. We estimated intake for 15 organic compounds commonly found in indoor environmental samples using the fugacity model time-dependent mass distribution among indoor compartments, human activity patterns, our inhalation model, and microactivity approaches of Cohen Hubal et al.\textsuperscript{27} for dermal and nondietary ingestion exposure. The chemicals were grouped into three categories on the basis of their relative chemical properties. Formaldehyde, naphthalene, and diethyl phthalate were categorized as “volatile” compounds. Galaxolide, tris(2-chloroethyl) phthalate, diazinon,
and chlorpyrifos were categorized as “minimally volatile” compounds. The remaining compounds, benzo[a]pyrene, permethrin, PBDE-47, PBDE-99, PBDE-209, triphenyl phosphate, bis(2-ethylhexyl) phthalate, and butyl benzyl phthalate, were categorized as “nonvolatile” compounds. The primary exposure pathway for volatile compounds is always gas-phase inhalation regardless of application type. Minimally volatile compound exposure is dependent on the release scenario, with gas-phase inhalation dominating for releases to air and nondietary ingestion dominating for applications to surfaces. For nonvolatile compounds, inhalation of particles is important for releases to air and applications to carpet, while dermal and nondietary ingestion are significant for releases to air and applications to carpet and are the dominant exposure pathways for applications to vinyl.

Figure 1 illustrates the relative contribution of each exposure pathway to the total intake and cumulative intake for chlorpyrifos (a, b, and c) and PBDE-209 (e, f, and g) as representative minimally volatile and nonvolatile compounds, respectively. Gas-phase inhalation is a dominant exposure...
pathway for chlorpyrifos released into air, whereas particle-phase inhalation accounts primarily for PBDE-209 exposure. When applied to carpet, more than 95% of chlorpyrifos exposure is driven by gas-phase inhalation because it moves from carpet to air compartment due to high diffusive and advective transfer rates from carpet to air, while particle-phase inhalation, dermal uptake, and nondietary ingestion account for about 50%, 10%, and 40% of PBDE-209 exposure, respectively. When applied to vinyl, about 50% of chlorpyrifos is taken up from gas-phase inhalation and the rest of the exposure is from dermal and nondietary ingestion. In contrast, dermal uptake and nondietary ingestion account for about 30% and 70% of PBDE-209 exposure, respectively. For this application mode, the contribution of dermal exposure increases for both compounds because the dislodgeable fraction (100%) of the compound associated with the vinyl compartment is much greater than the dislodgeable fraction (1.5%) of the compound associated with the carpet compartment, resulting in higher exposure concentrations on vinyl surfaces. The exposure profile for a compound depends on the actual application compartment. For example, if chlorpyrifos was applied to a vinyl surface in a kitchen or bathroom, a common application room, dermal and nondietary exposure would be important, as has been predicted in detailed exposure studies.43,44

In considering the overall persistence of chemicals in the indoor environment, we noted that with respect to the three application modes, both chlorpyrifos and PBDE-209 remained the longest indoors when applied to carpet. This occurs because a significant portion of the compound is in surface reservoirs, such as carpet fibers or foam materials, resulting in long indoor persistence. In contrast, when these same two compounds are applied to vinyl, it takes about 90 days for them to be removed.

### 3.2. Overall Intake Fractions for Each Chemical

Figure 2 shows the relative contribution of intake from each exposure pathway to the total intake (left) and the absolute iF (right) for the three chemical application modes. In the case of air release, overall baseline iF ranges from $3.1 \times 10^{-3}$ to $6.3 \times 10^{-3}$. For volatile and minimally volatile compounds, the total intake is dominated by inhalation of the gas phase. For nonvolatile compounds, the contribution of particle-phase inhalation, dermal uptake, and nondietary ingestion becomes significant to the total intake. In the case of application to carpet, iF is estimated from $3.8 \times 10^{-5}$ to $6.2 \times 10^{-3}$. The overall pattern of contribution of each exposure pathway to the total intake is similar to the results for air release. In the case of application to vinyl, iF is estimated from $9.0 \times 10^{-5}$ to $1.8 \times 10^{-2}$. The pattern of exposure pathway contribution for vinyl is clearly different from air and carpet chemical application modes. This arose because of higher dislodgeable fractions of vinyl dust and organic film compared to the carpet compartment and the higher transfer rates from vinyl to air. Because there are no data available to determine the dislodgeable fraction of compounds from vinyl dust and organic carbon to hands, we used an approach that tends toward higher rather than lower exposure estimates.

In evaluating the results in Figure 2, we observed that exposure to volatile compounds was primarily driven by gas-phase inhalation due to high transfer rates from vinyl to air, while exposure to nonvolatile compounds was driven by dermal and nondietary ingestion due to high exposure concentrations.
on vinyl surfaces, associated primarily with $K_{ow}$ values. However, tris(2-chloroethyl) phosphate, diazinon, and chlopyrifos have lower transfer rates from vinyl to air, resulting in relatively lower exposure concentrations in air. These chemicals also have lower $K_{ow}$ values, resulting in lower exposure concentrations on vinyl surfaces, implying that chemical properties determine both total intake and primary exposure pathways.

Ventilation, surface cleaning, and degradation due to OH radical reactions are considered potentially important removal pathways for the selected compounds. When released to air, ventilation is a primary removal pathway for all compounds due to the large air exchange rate (Figure S2a, Supporting Information). As the vapor pressure decreases, the contribution of ventilation to the total removal rate decreases while that of surface cleaning increases. When a compound is applied to carpet, ventilation is still a primary removal pathway (>85%) for volatile and minimally volatile compounds (Figure S2b, Supporting Information). However, surface cleaning accounts for more than 70% of total removal for nonvolatile compounds. When a compound is applied to vinyl, ventilation is a dominant removal pathway for formaldehyde and naphthalene, whose vapor pressure values are greater than 10 Pa. On the other hand, diethyl phthalate is removed almost equally by ventilation and surface cleaning, and other chemicals are dominantly removed by surface cleaning (Figure S2c, Supporting Information).

### 3.3. Sensitivity Analysis: Dermal and Nondietary Ingestion

We calculated $iF$ from the various dermal and nondietary models, which are fundamentally different in underlying assumptions and processes, and found large differences, indicating that model choice is likely one of the greatest contributors to output uncertainties. In order to explore the dependence of $iF$ on chemical properties and alternative modeling approaches, we carried out a sensitivity analysis for four of the compounds (diethyl phthalate, chlopyrifos, permethrin, and benzo[a]pyrene). Figure 3 illustrates the results of the sensitivity analysis for air release, providing estimates of $iF$ via dermal uptake and nondietary ingestion from different methods. For diethyl phthalate, which has a relatively large inhalation intake, model selection for dermal and nondietary ingestion pathways was not significant on the total $iF$. For chlopyrifos and benzo[a]pyrene, although changing a model for dermal uptake (from a touch model to a dust ingestion model) resulted in increased $iF$ by approximately 0.5–2 orders of magnitude, the total $iF$ did not increase significantly. However, the importance of dermal uptake and nondietary ingestion became comparable to inhalation intake for these compounds. For permethrin, the total $iF$ increased approximately 1.5 orders of magnitude upon changing from a touch model to a dust ingestion model for nondietary ingestion, making nondietary ingestion the driving exposure pathway. Results from carpet and vinyl application are provided in Figures S3 and S4 of the Supporting Information.

Using all available models, the minimum (red triangle) and maximum (blue diamond) $iF$ values are shown in Figure 2, indicating that, depending on the chemical application mode, nonvolatile compounds have uncertainties of about 0.5–2 orders of magnitude in the estimates of intake as a result of the uncertainties in dermal and nondietary ingestion. A complete breakdown of $iF$ for both adults and children from all pathways and models, including the percent dermal uptake directly from air versus uptake from surface contact in the Weschler and Nazaroff model, can be found in Table S8 of the Supporting Information. Use of the alternate model generally increased total $iF$ for emissions to air and applications to carpet. With the baseline models, $iF$ was found to be slightly lower for the less-volatile compounds than the more volatile compounds, as less-volatile compounds are more rapidly removed from the air, decreasing inhalation exposure, while dermal and nondietary exposure are not increased accordingly. With the alternate models, increases in dermal and nondietary exposures for the less-volatile compounds far exceeded the decrease in exposure through inhalation resulting from partitioning out of air. This results in the less volatile compounds having a higher $iF$ than the more volatile compounds. Thus, the choice of model for dermal and nondietary intake results in different absolute values of total $iF$.

In addition, the model choice dramatically alters the importance of dermal and nondietary exposure, altering the primary exposure pathway for compounds such as chlopyrifos, permethrin, and benzo[a]pyrene. As a result, the relative ranking of $iF$ shifts. This implies that exposure models should be carefully selected in screening large sets of compounds. Thus, further research is needed to ensure this pathway is properly quantified.

### 3.4. Implications/Conclusions

It is well established that $iF$ is greater for releases to the indoor environment than for releases to the outdoor environment. This work is an effort to build more capacity for addressing indoor exposures. However, there has been a lack of understanding about how $iF$ values for indoor releases vary among a broad range of chemical compounds and the relative importance of various exposure pathways for indoor releases. This paper addresses this critical need by providing a first attempt to estimate multipathway residential indoor $iF$ values for a suite of organic compounds. The reported indoor $iF$ values range from $10^{-3}$ to $10^{-1.4}$, highlighting the critical need to understand releases to the indoor environment relative to the outdoor environment, as $iF$ values for releases to the outdoor environment range from $10^{-8}$ to $10^{-4.5}$. The 2 orders of magnitude range in indoor $iF$ values also points to the importance of understanding variability between compounds.

Estimating dermal and nondietary ingestion via direct observation is a necessary adjunct to this work. However, direct observation is costly and labor-intensive and has many challenges (e.g., different residue-to-skin transfer efficiency with repeated contacts). Thus, it is vital to provide a modeling framework to help guide and interpret these observational studies. In addition, the difficulties or technical issues of collecting urine and blood samples from a young child make it challenging to validate model results associated with exposures driven by dermal and nondietary ingestion. Although the estimates of total intake from these pathways are uncertain, the positive correlations between PBDE biomarkers and indoor dust concentrations suggest that dermal and nondietary ingestion are potentially important pathways contributing to human body burden for PBDEs. Moreover, although intake estimates from dermal and nondietary pathways have significant uncertainties, excluding these pathways fails to provide a comprehensive estimation of exposure. For example, Xu et al. reported that the total exposure to diethyl phthalate emitted from vinyl flooring was primarily from the oral ingestion of dust.
Environmental Science & Technology

Despite the lack of model evaluation by case studies, the modeled \( iF \) from this study offers key insights on the role of chemical properties and chemical application modes in controlling human exposure to chemicals released indoors. We believe this fugacity model can be useful in screening large numbers of chemicals. \( iF \) for a pulse–release scenario can be applied to large numbers of chemicals, provided chemical properties are available. Ultimately, for chemical prioritization, this near-field model needs to be integrated with a far-field model, and the portion of the mass of the chemical released to the indoors as compared to the outdoors must be known. Furthermore, models that account for direct consumer product use, such as personal care products applied directly to the skin, also need to be included for complete prioritization. This model fills an important gap in understanding chemical exposures occurring in the indoor environment.

**ASSOCIATED CONTENT**

**Additional Information**

- Additional text, eight tables, and four figures with input parameters, fugacity capacities, exposure concentration equations, and mass balance equations of an indoor fugacity model; contribution of each removal pathway to total removal amount; and results of this sensitivity analysis for surface applications.

This material is available free of charge via the Internet at http://pubs.acs.org.

**AUTHOR INFORMATION**

**Corresponding Author**

*E-mail: hmshin@ucdavis.edu; phone: 949.648.1614; fax: 530.752.5300.*

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

This research is funded by the American Chemistry Council (Grant 3-DBACC01).

**REFERENCES**


(22) Average number of people per household, by race and Hispanic origin, marital status, age, and education of household; U.S. Census Bureau, Washington, DC, 2011.


(24) PM 2.5 objectives and history; U.S. Environmental Protection Agency, Washington, DC, 2008.


