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Determining Source Strength of Semivolatile Organic Compounds using Measured Concentrations in Indoor Dust

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Abstract

Consumer products and building materials emit a number of semivolatile organic compounds (SVOCs) in the indoor environment. Because indoor SVOCs accumulate in dust, we explore the use of dust to determine source strength and report here on analysis of dust samples collected in 30 U.S. homes for six phthalates, four personal care product ingredients, and five flame retardants. We then use a fugacity-based indoor mass-balance model to estimate the whole house emission rates of SVOCs that would account for the measured dust concentrations. Di-2-ethylhexyl phthalate (DEHP) and di-iso-nonyl phthalate (DiNP) were the most abundant compounds in these dust samples. On the other hand, the estimated emission rate of diethyl phthalate (DEP) is the largest among phthalates, although its dust concentration is over two orders of magnitude smaller than DEHP and DiNP. The magnitude of the estimated emission rate that corresponds to the measured dust concentration is found to be inversely correlated with the vapor pressure of the compound, indicating that dust concentrations alone cannot be used to determine which

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Supporting Information Available

Additional Supporting Information may be found in the online version of this article.

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compounds have the greatest emission rates. The combined dust-assay modeling approach shows promise for estimating indoor emission rates for SVOCs.

Keywords

dust; emission rates; modeling; flame retardants; personal care products; phthalates

1. Introduction

Consumer products and building materials emit phthalates, pesticides, flame retardants, and other personal care product ingredients such as synthetic musk and sun-blocking agents in indoor environments (Rudel et al., 2003; Wormuth et al., 2005, 2006). Exposure to many of these compounds results in adverse health effects, including asthma and endocrine disruption (Dodson et al., 2012). Over the past several decades, chemicals contained in consumer products and building materials have been measured in household air and dust (Rudel et al., 2010; Weschler, 2009; Weschler and Nazaroff, 2008). As a result, there has been increasing interest in assessing exposure to these indoor organic pollutants through inhalation, dermal uptake, and oral ingestion of settled dust (Shin et al., 2012). Once these are emitted into the indoor environment, estimation of reliable indoor exposure concentrations requires a detailed understanding of sources/sinks, transport, and distribution among gas- and particle-phases, settled dust, and surface compartments (e.g., carpet, vinyl flooring, and walls) (Liu et al., 2013; Salthammer and Bahadir, 2009; Weschler and Nazaroff, 2010). However, for most chemicals there is limited information available about how much their use in consumer products and building materials results in releases to indoor environments and then to human exposures. To address this gap we carried out research to explore the use of measured dust concentrations in combination with indoor chemical mass-balance models to predict the whole house emission rates.

The alternate method for deriving the whole house emission rate would be to sum emissions from various individual consumer products and building materials. Estimates of source strength from the use of consumer products such as cleaning products, cosmetics, and biocides depend on human activities and chemical concentrations in the products (Dodson et al., 2012; Wormuth et al., 2005). For example, the emission rates of personal care products such as shampoo, body wash, sun-blocking agents, and body lotion are calculated on the basis of the frequency and amount of product application, the chemical retention factor (i.e., the ratio of the amount remaining on the skin to the amount applied to the skin), the chemical concentration in products, and, for chemical evaporation estimates, the amount of time the user spends indoors between product application and wash-off.

Releases of compounds from building materials, home furnishings, and electronic devices as well as evaporation from paints and adhesives depend on the concentration of the compound in the material used, the diffusion rate of the compound through the product's material matrix, the area of source material within the room, indoor temperature (Wirts et al., 2003), and the air exchange rate (Xu et al., 2009). Using chamber experiments, researchers have published several studies that measured emission rates of organophosphate and brominated flame retardants from building and plastic materials (Kemmlein et al., 2003; Ni et al., 2007)

and of phthalates from polyvinyl chloride (PVC) flooring, wall coverings, and other materials (Afshari et al., 2004; Clausen et al., 2004; Uhde et al., 2001; Xu et al., 2012). Two studies (Xu et al., 2009; Xu and Little, 2006) also developed a model to predict emission rates of di-2-ethylhexyl phthalate (DEHP) from vinyl flooring and polymeric materials, accounting for the mass transfer within the boundary layer due to the concentration gradient, based on Clausen et al. (2004) experiments in both the FLEC (Field and Laboratory Emission Cell) and CLIMPAQ (Chamber for Laboratory Investigations of Materials, Pollution, and Air Quality) emission chambers.

As it is difficult to sum emissions for various products used in the home, an alternate method is needed to determine the whole house emission rate. One such approach utilized measured air concentrations and an indoor multimedia fugacity model to estimate emission rates of polybrominated diphenyl ethers (PBDEs) in an office (Zhang et al., 2009) and of both PBDEs and polychlorinated biphenyls (PCBs) in a residential indoor environment (Zhang et al., 2011). As an alternative to air concentrations, we hypothesized that dust measurements can be used to determine the source strength of semivolatile organic compounds (SVOCs) in the home. It may be more practical to use dust to estimate emissions, because dust can be collected in a single home visit, as opposed to air, which requires a second visit to retrieve the air sampler. Also, compounds with very low vapor pressure are more likely to have levels in dust that exceed the limit of detection of the method than in air, and thus more compounds can be analytically quantified. Moreover, dust is a reservoir for the compounds released into the home (Clausen et al., 2004; Shin et al., 2013), potentially reflecting chemical loading in the home over a longer period. Therefore, in this study, we utilized measured dust concentrations to predict the whole house emission rate as an alternate approach.

The objectives of this study are to (1) measure household dust concentrations of SVOCs released from consumer products and building materials from 30 U.S. homes, (2) apply a fugacity-based indoor mass-balance model to these data to estimate the whole house SVOC emission rates as a function of chemical properties and dust concentrations, (3) identify input parameters contributing most to the output uncertainty, and (4) compare the modeled whole house emission rates to those inferred from personal care product use behaviors and diffusive transfers from indoor surfaces to air.

2. Methods

2. 1. Overview

In this study, our first step is the analysis of 30 household dust samples to determine levels of 15 compounds primarily released from consumer products and indoor surfaces. We then derive generalized analytical solutions from an indoor fugacity model to estimate SVOC emission rates that account for the measured dust concentrations. Emission rates inferred from personal care product use behaviors and diffusive transfers are also estimated and compared to those from the fugacity model. We use these results to evaluate the feasibility of using dust measurements in combination with mass-balance models to infer SVOC emissions. The overview of this approach is also depicted in Figure S1 in the Supporting Information.

2. 2. SVOC Dust Samples

2.2.1. Selected Compounds—We selected 15 organic compounds, including one phthalate often used in a variety of personal care products [diethyl phthalate (DEP)], one phthalate often used in both PVC products and personal care products [di-iso-butyl phthalate (DiBP)], four phthalates commonly used in vinyl flooring and PVC plastics [di-n-butyl phthalate (DnBP), butylbenzyl phthalate (BBP), di-2-ethylhexyl phthalate (DEHP), and di-iso-nonyl phthalate (DiNP)] (Hauser and Calafat, 2005; Heudorf et al., 2007), two personal care product ingredients typically used as fragrances [7-acetyl-1,1,2,4,4,6-hexamethyl-tetralin (AHTN), 1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta-(g) 2-benzopyran (HHCB)], two sun-blocking agents [octyl dimethyl PABA (ODP), octyl methoxycinnamate (OMC)], and five flame retardants mostly used in plastics, couch foams, and textiles [bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (TBPH), 2-ethylhexyl-2,3,5,6-tetrabromobenzoate (TBB), tris(2-chloroethyl) phosphate (TCEP), triphenyl phosphate (TPP), and tris(1,3-dichloro-2-propyl) phosphate (TDCPP)]. Phthalates, synthetic musk in fragrances, and sun-blocking agents have been shown to have endocrine-disrupting potential (Dodson et al., 2012). In addition, DEHP in house dust has been associated with asthma (Bornehag et al., 2004). Organophosphorus flame retardants are also known to have adverse health effects: TPP can cause dermatitis in humans (WHO, 1991) and TCEP has been found to have carcinogenic potential in rats and mice (Beth-Hubner, 1999).

2.2.2. Sample Collection—Indoor dust samples (n=30) were collected from residences located in Northern California, Southeast Pennsylvania, and Northeast Maryland. Study participants were women participating in the Early Autism Risk Longitudinal Investigation (EARLI) (Newschaffer et al., 2012). These women are mothers who had a child diagnosed with an Autism Spectrum Disorder (ASD) and who were pregnant with another child at the time of sample collection. Samples were collected from the main living room of the home between 2009 and 2010 by research study staff. The main living room was selected as the room where the mother typically spent most of her time. Dust was collected with a Eureka Mighty-Mite vacuum cleaner equipped with the standard crevice tool attachment (Model 3670), modified to capture dust in a 19×90 mm cellulose extraction thimble (Whatman Inc), using a standardized collection protocol (Allen et al., 2008; Rudel et al., 2003). The equivalent of the entire floor-surface area of the room was vacuumed. Dust was not collected from under furniture or in crevices between cushions. The thimbles containing the dust samples were wrapped in pre-cleaned aluminum foil, placed in 50 ml polypropylene vials, shipped in a cooler to the central repository, and stored at −20 °C until analysis. The dust was shaken from each thimble into a 100-mesh stainless steel sieve, and sieved to obtain the fraction of dust smaller than 150 µm.

2.2.3. Sample Analysis—A 0.5 g aliquot of each dust sample was weighed out and spiked with two surrogate recovery compounds (SRSs) that are used to assess method performance on a sample-by-sample basis: 1.25 µg of d₄-di-n-butyl phthalate (SRS for the phthalates) and 0.25 µg of d₁₀-phenanthrene (SRS for personal care product ingredients). The dust was extracted for 15 minutes by ultra-sonication in 12.0 mL of 1:1 hexane: acetone (Colt et al., 2008; Harnley et al., 2009). A 1.0 mL aliquot was removed, spiked with the internal standard (IS) 4, 4'-dibromobiphenyl, and analyzed directly using electron impact

gas chromatography mass spectrometry (EI GC/MS) for the phthalate esters. The remainder of the extract was cleaned using solid phase extraction, concentrated to 1.0 mL and spiked with the same IS prior to analysis using both conventional EI GC/MS for the majority of the analytes and negative chemical ionization (NCI) GC/MS for the flame retardants of TBPH and TBB. Samples were analyzed concurrently with multi-point calibration curves that spanned the anticipated range of sample concentrations. Samples that exceeded the highest calibration point by more than 15% were diluted, respiked proportionally with IS, and reanalyzed. Samples were analyzed using an Agilent 6890 GC/5973N MS, with methane as the reagent gas for the NCI analyses. EI GC/MS analyses for phthalates and other personal care product ingredients were performed using a moderate polarity GC column (Restek Rtx-5MS, 30 m $\times$ 0.25 mm id $\times$ 0.25 μ m film), programmed from 50–100 $^{\circ}$ C @ 8 $^{\circ}$ /min and then 100–300 $^{\circ}$ C @ 10 $^{\circ}$ /min. NCI GC/MS analyses for selected electro-negative compounds were performed using a similar column (J&W Scientific DB-5MS, 15 m $\times$ 0.25 mm id $\times$ 0.10 μ m film), programmed from 50–300 $^{\circ}$ C @ 10 $^{\circ}$ /min. The IS method of quantification was used with calibration curves derived from linear least-squares models.

2.2.4. Quality Assurance—The 30 samples were analyzed in two sample sets. Each set included 1 solvent method blank, 15 field samples, one sample selected at random for duplicate sample analysis, and that same sample also selected for spiking with all analytes (effective spike level of 20 μ g/g for phthalates and 0.5 μ g/g for all other analytes), plus the 7 calibration curve standards. Percent recovery of the spike was calculated after subtracting the average analyte level in the duplicates. The analyte-specific limit of detection (LOD) is listed in Table S1. Personal care product ingredients and flame retardants were not detected in the solvent method blanks; trace levels of most phthalates were detected in solvent method blanks with concentrations equivalent to 0.02–0.12 μ g/g; DiNP was not detected in these blanks (see Table S1). The phthalate with the lowest level in dust (i.e., DEP) had an effective solvent method blank level that was 10-fold lower than the lowest sample level and thus samples were not blank corrected. Duplicate analyses showed very good agreement; the average relative percent difference was 9% (range: 0 – 41% for individual pairs of replicate samples; see Table S1). Analyte recoveries from spiked samples averaged 81% (range of 56% for HHCB to 128% for TDCPP; see Table S1). Analyte recoveries for DEHP and OMC could not be determined due to significantly higher (10–100 times) native levels than spike levels. Structural similarity to other analytes suggests that recoveries for these compounds would be similar to the other analytes (Dodson et al., 2012). The average recoveries of the SRSs in the samples were 93% for d₄-di-n-butyl phthalate and 78% for d₁₀-phenanthrene.

2. 3. Development of SVOC Emission Rate Model

2.3.1. Mass Balance—In order to relate measured indoor dust concentrations to the emission rate of SVOCs from indoor sources, we used a fugacity-based indoor mass-balance model that accounts for chemical partitioning and transfers between air and surface compartments (Bennett and Furtaw, 2004; Shin et al., 2012, 2013) (see Supporting Information for summary of model). The model includes four compartments that serve as potential indoor reservoirs of a chemical: air, carpet, vinyl flooring, and walls. The air compartment is comprised of gas- and particle-phases. Therefore, we computed the fugacity capacity of each phase and added those for the total fugacity capacity of the air

compartment. We quantified advective mass transport driven by particle resuspension and deposition. We also quantified mass transfers driven by fugacity difference between air and surface compartments, traditionally referred to as diffusive mass transport, using a fugacity-based mass-transfer coefficient. Ideally, resistance to mass transfer depends on the diffusivity of the compound and airflow conditions in the room. A simplified model is used in the approach.

Assuming that contributions from outdoors are negligible, the mass balance equation for the air compartment is

$$\frac{dM_a}{dt} = S + M_c \cdot T_{c \rightarrow a} + M_v \cdot T_{v \rightarrow a} + M_w \cdot T_{w \rightarrow a} - M_a \cdot (a + k_a + T_{a \rightarrow c} + T_{a \rightarrow v} + T_{a \rightarrow w}) \quad (1)$$

where M_i is the mass associated with compartment i (a for air, c for carpet, v for vinyl, and w for wall) (mg), S is the emission rate from indoor sources such as consumer products and building materials (mg/day), a is the air exchange rate (1/day), k_a is the degradation rate constant in air ($= C_{OH} \times k_{OH}$) (1/day), C_{OH} is the OH radical concentration in air (mol/cm^3), k_{OH} is the chemical-specific OH radical reaction constant ($\text{cm}^3/\text{mol}\cdot\text{day}$), and T values are the transfer factors (1/day). We defined $T_{c \rightarrow a}$ as transfers from carpet to air, $T_{v \rightarrow a}$ as transfers from vinyl to air, $T_{w \rightarrow a}$ as transfers from wall to air, $T_{a \rightarrow c}$ as transfers from air to carpet, $T_{a \rightarrow v}$ as transfers from air to vinyl, and $T_{a \rightarrow w}$ as transfers from air to wall. We considered only oxidation by the OH radical as the degradation mechanism in indoor air, because other degradation mechanisms such as photolysis and chemical and biological degradation are negligible in indoor air (Weschler and Nazaroff, 2008).

Assuming that SVOCs are directly emitted from sources (e.g., consumer products, furniture, vinyl flooring, and PVC plastics) to indoor air with no emissions to the other compartments, the mass balance equations for the carpet, vinyl, and wall compartments are

$$\frac{dM_c}{dt} = M_a \cdot T_{a \rightarrow c} - M_c \cdot k_c \cdot \theta_c - M_c \cdot T_{c \rightarrow a} \quad (2)$$

$$\frac{dM_v}{dt} = M_a \cdot T_{a \rightarrow v} - M_v \cdot k_v \cdot \theta_v - M_v \cdot T_{v \rightarrow a} \quad (3)$$

$$\frac{dM_w}{dt} = M_a \cdot T_{a \rightarrow w} - M_w \cdot T_{w \rightarrow a} \quad (4)$$

where k_c and k_v are the dust removal rate through cleaning of carpet and vinyl flooring, respectively (1/day), θ_c is the fraction of the compound in the mobile phase in the carpet compartment ($= M_{\text{carpet dust}} / (M_{\text{carpet dust}} + M_{\text{carpet}})$), and θ_v is the fraction of the compound in the mobile phase in the vinyl compartment ($= (M_{\text{vinyl dust}} + M_{\text{film}}) / (M_{\text{vinyl dust}} + M_{\text{film}} + M_{\text{vinyl}})$) (Shin et al., 2013). We note that in actuality, vinyl flooring may emit compounds to air and dust sitting on the vinyl, which will eventually transfer to air. Using dust collected on

carpet, the original source of the compound in the model is indistinguishable. Assuming that the mass in each compartment is at steady-state (i.e., $dM_i/dt=0$), Equations 1–4 become

$$S=M_a \cdot (a+k_a+T_{a-c}+T_{a-v}+T_{a-w})-M_c \cdot T_{c-a}-M_v \cdot T_{v-a}-M_w \cdot T_{w-a} \quad (5)$$

$$M_a=\frac{M_c \cdot (k_c \cdot \theta_c+T_{c-a})}{T_{a-c}} \quad (6)$$

$$M_v=M_a \cdot \frac{T_{a-v}}{(k_v \cdot \theta_v+T_{v-a})}=\frac{M_c \cdot (k_c \cdot \theta_c+T_{c-a})}{T_{a-c}} \cdot \frac{T_{a-v}}{(k_v \cdot \theta_v+T_{v-a})} \quad (7)$$

$$M_w=M_a \cdot \frac{T_{a-w}}{T_{w-a}}=\frac{M_c \cdot (k_c \cdot \theta_c+T_{c-a})}{T_{a-c}} \cdot \frac{T_{a-w}}{T_{w-a}} \quad (8)$$

Inserting Equations 6–8 into 5 yields

$$S=M_c \cdot \frac{(k_c \cdot \theta_c+T_{c-a})}{T_{a-c}} \left[a+k_a+\frac{k_c \cdot \theta_c \cdot T_{a-c}}{(k_c \cdot \theta_c+T_{c-a})}+\frac{k_v \cdot \theta_v \cdot T_{a-v}}{(k_v \cdot \theta_v+T_{v-a})} \right] \quad (9)$$

To determine the mass in the carpet based on the known concentration in the carpet dust (C_{cp} , $\mu\text{g/g}$ of dust), we used the fugacity principle, $M=f \cdot Z \cdot V$, where M is the mass of the compound in the compartment (mol), f is the fugacity (Pa), Z is the fugacity capacity ($\text{mol/m}^3 \cdot \text{Pa}$), and V is the volume of the compartment (m^3). Fugacity can be regarded as the partial pressure or the tendency of a chemical to leave or escape from a given state or compartment (Bennett and Furtaw, 2004) (see Supporting Information for more information on the definitions of fugacity and fugacity capacity). Assuming that each phase in a given compartment is in chemical equilibrium (e.g., fugacity in the carpet fiber = fugacity in the carpet dust), the application of this principle to the carpet fiber and carpet dust in the carpet compartment results in the following equation.

$$M_c=M_{cf}+M_{cp}=(f \cdot Z_{cf} \cdot V_{cf}+f \cdot Z_{cp} \cdot V_{cp}) \cdot MW \cdot (10^3 \text{mg/g}) \quad (10)$$

where M_{cf} and M_{cp} are the mass in the carpet fiber and carpet dust (mg), respectively, Z_{cf} and Z_{cp} are the fugacity capacity of the carpet fiber and carpet dust ($\text{mol/m}^3 \cdot \text{Pa}$), respectively, V_{cf} and V_{cp} are the volume of the carpet fiber and carpet dust (m^3), respectively, and MW is the molecular weight (g/mol). Using the fact that the mass in the carpet dust is also the product of the concentration (C_{cp}) and volume (V_{cp}) of the dust yields

$$M_{cp}=f \cdot Z_{cp} \cdot V_{cp} \cdot MW \cdot (10^3 \text{mg/g})=C_{cp} \cdot V_{cp} \cdot \rho_{\text{dust}} \quad (11)$$

where ρ_{dust} is the dust density in the carpet compartment (kg/m^3). Rearranging Equation 11 to solve for f yields

$$f = \frac{C_{cp} \cdot \rho_{dust}}{Z_{cp} \cdot MW \cdot (10^3 \text{mg/g})} \quad (12)$$

Inserting Equation 12 to Equation 10, we get

$$M_c = \frac{C_{cp} \cdot \rho_{dust}}{Z_{cp}} \cdot (Z_{cf} \cdot V_{cf} + Z_{cp} \cdot V_{cp}) \quad (13)$$

Inserting Equation 13 into Equation 9, we get

$$S = \left[\frac{C_{cp} \cdot \rho_{dust}}{Z_{cp}} \cdot (Z_{cf} \cdot V_{cf} + Z_{cp} \cdot V_{cp}) \cdot \frac{(k_c \cdot \theta_c + T_{c-a})}{T_{a-c}} \right] \cdot \left[a + k_a + \frac{k_c \cdot \theta_c \cdot T_{a-c}}{(k_c \cdot \theta_c + T_{c-a})} + \frac{k_v \cdot \theta_v \cdot T_{a-c}}{(k_v \cdot \theta_v + T_{v-a})} \right] \quad (14)$$

Environmental input parameters such as ρ_{dust} , V_{cf} , V_{cp} , a , k_c , and k_v and the properties of airborne particles in different size fractions are listed in Tables S2 and S3 of the Supporting Information. Derivations of Z_{cf} , Z_{cp} , T_{c-a} , T_{a-c} , T_{v-a} , T_{a-v} , θ_c , and θ_v are based on an existing indoor fugacity model (Bennett and Furtaw, 2004; Shin et al., 2012, 2013) and described in detail in Tables S4, S5, and S6 of the Supporting Information. These values depend on chemical properties, which are listed in Table 1.

2.3.2. Uncertainty Analysis—We performed a Monte Carlo uncertainty analysis to compute the distribution of predicted emission rates and then determined the relative contribution of input parameters to the output uncertainty of the emission rate for each studied compound. Distributions of input variables used for the uncertainty analysis, including the mean, coefficient of variation (CV), and the type of distribution, are provided in Table S2. Also, we used a log-normal distribution for all physicochemical properties with the following CVs recommended by McKone (1993): VP = 0.38; H = 0.45; K_{ow} = 0.37; K_{oa} = 0.10; K_p = 0.1; k_{OH} = 1.0; D_{air} = 0.1. Due to the large number of input variables ($n = 46$), we used a stepwise method to perform a multilinear regression on the emission rate and included only the variables whose regression coefficients have a p-value less than 0.05 in the final model. Then, we calculated pairwise correlation coefficients between the output variable and each of the included inputs and computed the percent contribution of the included input parameters to output (emission rate) uncertainty.

2. 4. Alternate Methods to Estimate Emissions from Personal Care Products and Building Materials

2.4.1. Emission Rates from Personal Care Products—For compounds often used as personal care products (two phthalates (DEP, DiBP), synthetic musk compounds, and sun-blocking agents), we used the following equation to estimate the emission rate from the use of personal care products.

$$S_{pcp} = C_i \cdot A_a \cdot f_{\text{retention}} \cdot F_{\text{evap}} \cdot (g/10^6 \mu g) \quad (15)$$

where S_{pcp} is the emission rate of compounds from personal care products (mg/day), C_i is the chemical concentration in personal care products ($\mu\text{g/g}$), A_a is the daily application rate (mg/day), which is the product of the applied amount per use (mg/use) and the frequency of product application (uses/day), $f_{\text{retention}}$ is the retention factor (unitless), which is the ratio of the amount that remains on the body to the amount applied (e.g., body lotion=1 and body wash=0.05), and F_{evap} is the fraction of the compound that evaporates after application (unitless).

We used the concentrations of the phthalates, synthetic musk compounds, and sun-blocking agents in personal care products from four published studies, averaging concentrations when reported in more than one study (Dodson et al., 2012; Koniecki et al., 2011; Roosens et al., 2007; Wormuth et al., 2006) (see Supporting Information, Table S7). The Dodson et al. paper (2012) reports chemical concentrations in a product category as either 1–100 $\mu\text{g/g}$, 100–1,000 $\mu\text{g/g}$, or above 1,000 $\mu\text{g/g}$. Thus, for compounds within one of the lower two ranges, we used the geometric mean of the end points of the range, and for chemical concentrations greater than 1,000 $\mu\text{g/g}$, we used the geometric mean (3,162 $\mu\text{g/g}$) between 1,000 and 10,000 $\mu\text{g/g}$, acknowledging that this assumption might result in over- or under-estimation of chemical concentrations in the products. We estimated the average daily application rate of personal care products from five published studies (Hall et al., 2007; Loretz et al., 2006, 2008; Neale et al., 2002; Wormuth et al., 2005) (see Supporting Information, Table S8). We applied the retention factor of personal care products from Wormuth et al. (2005) (see Supporting Information, Table S8).

We used the following equation to compute the fraction of evaporation from skin after application (Kasting and Saiyasombati, 2001).

$$F_{\text{evap}} = x_r / (k + x_r) \quad (16)$$

where k , the transdermal permeation coefficient, is an experimentally determined parameter that depends on the air velocity over the skin and skin temperature and x_r , the evaporation potential, is determined using the following equation (Kasting and Saiyasombati, 2001):

$$x_r = \frac{VP_r \cdot MW_r^{2.7}}{(K_{ow} \cdot S_w)_r} \quad (17)$$

where VP_r is the dimensionless vapor pressure ($= VP/133 \text{ Pa}$), MW_r is the dimensionless molecular weight ($= MW/100 \text{ g/mol}$), K_{ow} is the octanol-water partition coefficient (unitless), S_w is the solubility in water (g/L), and $(K_{ow} \cdot S_w)_r$ is the dimensionless value ($= (K_{ow} \cdot S_w)/1000 \text{ g/L}$) for computational convenience. Increasing x_r results in a larger fraction of evaporation to indoor air and a smaller fraction of absorption through skin. The physicochemical properties for estimating x_r are listed in Table 1. Although the parameter k is chemical specific and influenced by experimental conditions, including wind velocity and skin temperature, we used a value of 0.15, the average of two values derived from perfume raw materials without fragrance fixative reported in Kasting and Saiyasombati (2001).

2.4.2. Emission Rates from Building Materials—For compounds commonly used in building materials and home furnishings, two studies (Xu et al., 2009; Xu and Little, 2006) developed a model to predict emissions of DEHP from vinyl flooring, accounting for the mass transfer within the boundary layer due to the concentration gradient. In our study, we adapted the Xu and Little model to predict emissions of our studied compounds released from various indoor surfaces. We applied Fick's law of diffusion to compute the diffusive flux (J) across the interface between air and surface and multiplied by the surface area of sources to estimate the emission rate of phthalates and flame retardants from indoor surfaces such as home furnishings and building materials using Equation 18 below.

$$S_{\text{surf},i} = J_i \cdot A_i = D_a \frac{(C_{s,\text{air}} - C_{\text{air}})}{\delta_{\text{bl}}} \cdot A_i \quad (18)$$

where $S_{\text{surf},i}$ is the emission rate of compounds (mg/day) from surface i (e.g., vinyl surfaces, electronics, couch), J_i is the diffusive flux (mg/m²-day) from surface i , A_i is the area of surface i (m²), D_a is the diffusion coefficient in pure air (m²/d), $C_{s,\text{air}}$ is the saturation concentration at the interface between surface materials and the air compartment (μg/m³), C_{air} is the concentration of the compound in the gas phase (C_g) of the air compartment (μg/m³), and δ_{bl} is the effective boundary-layer thickness of surface i (m).

Because D_a was not available in *EPI Suite 4.1* (EPA, 2012), we used a chemical-properties estimation tool available online (<http://www.envmodels.com/>) to calculate D_a based on reported values of VP , MW , and density from *EPI Suite 4.1*. For $C_{s,\text{air}}$, Clausen et al. found that the concentration immediately adjacent to the vinyl flooring is equal to the vapor pressure of pure DEHP (2012). Thus, the ideal gas law was employed to determine the saturation concentration for compound A associated with vapor pressure (Ramaswami et al., 2005).

$$C_{s,\text{air}}^A = \frac{M^A}{V} = \frac{P^A \cdot MW^A}{R \cdot T} \cdot \frac{10^6 \mu\text{g}}{\text{g}} \quad (19)$$

where $C_{s,\text{air}}^A$ is the saturation concentration of compound A at the interface between the surface materials and the air compartment (μg/m³), M^A is the mass of compound A (μg), V is the volume of the gas (m³), P^A is the saturation vapor pressure of compound A in air (Pa), MW^A is the molecular weight of compound A (g/mol), R is the ideal gas constant (8.314 Pa·m³/mol·K), and T is the absolute environmental temperature (298 K).

We computed the average concentration in the gas phase of the air compartment (C_g , μg/m³) from dust concentrations using the following ratio (K_{dg} , m³/g) of an SVOC's mass fraction in dust (C_{dust} , μg/g) to its gaseous concentration (Weschler and Nazaroff, 2010).

$$K_{\text{dg}} = \frac{C_{\text{dust}}}{C_g} = \frac{f_{\text{om-dust}} \times K_{\text{oa}}}{\rho_{\text{dust}}} \quad (20)$$

where $f_{\text{om_dust}}$ is the fraction of the dust that is organic matter (unitless) and K_{oa} is the octanol-air partition coefficient (unitless). We used reported values of $f_{\text{om_dust}}$ (=0.2) and ρ_{dust} ($=2.0 \times 10^3 \text{ kg/m}^3$) from Hunt et al. (1992).

Although the boundary-layer thickness is specific to both factors related to the material and the airflow in the room, such as friction velocity and smoothness of the material, we used the value 0.033 m from Bennett and Furtaw (2004), which was based on experimental values derived by Morrison and Nazaroff (2002) for a variety of surfaces and room conditions. Here, D_a/δ_{bl} is equivalent to the convective mass-transfer coefficient (h_m) as noted in the Xu et al. model (2009, 2012). The computed value of D_a/δ_{bl} of DEHP in our study ($=0.00015 \text{ m/s}$) is comparable to h_m ($=0.00041 \text{ m/s}$) calculated from Xu et al. (2012). We estimated the average surface area of a couch (sources for TBPH, TBB, TDCPP) based on measurements from 66 couches ($=8.9 \text{ m}^2$) (see Supporting Information for the description of methods). For PVC plastics (sources for DnBP, TCEP, TPP), we assumed that the surface area of a 42 inch television, one computer set (monitor, keyboard, desktop, and mouse), and other electronics is on the order of 1 m^2 and summed across different PVC plastic products. We used the value of surface area for vinyl flooring (sources for BBP, DEHP, DiNP) ($=24.5 \text{ m}^2$) from Bennett and Furtaw (2004). This material-specific area of surface was used to determine the emission rate of compounds from surface materials (S_{surf}) using Equation 18.

3. Results and Discussion

3.1. Dust Concentrations

A total of six phthalates, four personal care product ingredients, and five flame retardants were analyzed in household dust from 30 homes in three different regions in the U.S. Limited measurements of phthalate (Guo and Kannan, 2011; Rudel et al., 2003) and flame retardant (Meeker and Stapleton, 2010; Stapleton et al., 2008) concentrations in dust are available for U.S. homes and no studies to date have measured fragrance ingredients (AHTN, HHCB) and sun-blocking agents (ODP, OMC) in U.S. household dust. In our results, all compounds, except ODP, were above the limit of detection in all 30 dust samples. A box-plot and a summary of measured concentrations are provided in Figure 1 and Table S1. DEHP and DiNP, which have the lowest VP and highest K_{oa} values of the phthalates, were two most abundant compounds in these dust samples. It is noteworthy that the median dust concentration of OMC, a sun-blocking agent, is larger than that of DnBP, a plasticizer.

We also added median dust concentrations of our studied compounds from other published studies in Figure 1. Overall, the dust concentrations measured in our study tend to be lower than or similar to measurements reported in other U.S. and European studies. There are three studies that report exceptionally higher or lower concentrations. For example, three phthalate concentrations (DEP, DEHP, and DiNP) reported in Bulgaria (▲, green triangle) are consistently higher than other studies (Kolarik et al., 2008). For AHTN and HHCB, the concentrations reported in China (▼, green upside down triangle) (Lu et al., 2011) are about one order of magnitude lower than those from our study and Fromme et al. (2004). For most of the flame retardants, the concentrations in New Zealand dust samples were about 1 to 2 orders of magnitude lower than our dust samples (Ali et al., 2012).

3.2. Estimated Emission Rates Derived from the Fugacity Model

In addition to knowing the dust concentrations, we would like to know source emissions to the home as there is a dearth of data on indoor emission rates. To fill this gap we applied a modeling analysis to the dust measurements. Figure 2 provides the distribution of SVOC emission rate estimates from the Monte Carlo simulation of the fugacity-based indoor model and measured dust concentrations. For all phthalates and three personal care product ingredients (AHTN, HHCB, and OMC), the estimated median emission rates range from 0.7 to 1.6 mg/day, although the measured dust concentrations of these compounds vary over 2 orders of magnitude. In contrast to the results for phthalates and personal care product ingredients, for chemicals commonly used as flame retardants, our estimated median emission rates vary over 4 orders of magnitude, ranging from 0.001 to 48.5 mg/day, while all of the measured median dust concentrations are within approximately one order of magnitude. In order to understand the relationship between dust concentrations, estimated emission rates, and chemical properties, we plotted $\log(VP)$ versus the ratio of the measured dust concentration to the estimated emission rate. Figure 3 shows that the ratio is inversely correlated with $\log(VP)$, which indicates that the required whole house emission rate to reach the dust concentration (y-axis) for compounds with low VP values (e.g., TBB, TBPH, TDCPP, etc.) is estimated to be small, while that for compounds with high VP values (e.g., DEP, DiBP, AHTN, HHCB, etc.) is expected to be large.

Figure S4 illustrates the relative contribution of each input parameter to the output distribution from the multilinear regressions analysis. Overall, the area of house, the air exchange rate, and the concentration in the dust are the three most influential parameters on the emission rate. For compounds commonly used in building materials, couch foam, and PVC plastics (e.g., DEHP, DiNP, TBPH, TBB, and TDCPP), the dust-related parameters, including the removal rate from carpet cleaning, the embedded dust ratio (the ratio of the mass of total dust loading in the carpet to the mass removable by standard vacuuming), and the dust loading on the carpet, are also important input parameters for the value of the emission rate. For compounds commonly used in personal care products and other compounds with relatively small K_{oa} values, these dust-related parameters do not significantly influence the emission rate.

3.3. Estimated Emission Rates from Product Use and Building Materials Composition

As an alternate approach to the fugacity model, we also made emissions estimates based on personal care product use behaviors and building materials composition. Figure 2 shows estimated SVOCs emission rates inferred from personal care product use behaviors (●, red circle) and diffusive transfers from indoor surfaces to air (▲, blue triangle). For compounds commonly used as personal care product ingredients, the estimated median emission rates from the application and evaporation model (●, red circle), Equations 15 and 16, are within 0.5 orders of magnitude (= a factor of about 3) of the estimated median emission rates from the fugacity model (line inside boxes). For ODP, we could not apply the application and evaporation model because concentrations in the personal care products were not available. For compounds commonly used in home furnishings and PVC plastics, the simple diffusive model associated with diffusion coefficient and VP (referring to Equation 18) results in values that in many cases were much higher or lower than those from the fugacity model.

The fugacity model likely under-estimated emission rates for DnBP and BBP because these compounds did not reach steady-state even after 20 years of simulation. Another reason for discrepancy between model results might be limited information on the surface area of sources in the home. In addition, assuming that the concentration in the air at the surface materials reaches vapor pressure of the compound may be a significant over-estimate for some compounds. Because the content of some plasticizers (DnBP, BBP) in PVC products is generally low and certain flame retardants in polyurethane foam exist as solids at room temperature, these compounds do not behave as a pure liquid (Little et al., 2012). Thus, approximation of $C_{s,air}$ by vapor pressure could result in overestimates of emissions. Although there are uncertainties with both the evaporation and simple diffusive models, this approach still yields a first estimate of emissions and thus they are left as point values.

3.4. Comparison with Other Studies

In order to provide further evaluation of our emissions estimates approach, we compared estimated emission rates from home furnishings and building materials to the reported value in two studies (Xu et al., 2009; Xu and Little, 2006) (see Figure 2). Although other environmental conditions are different from our study homes and Xu et al.'s experimental settings (2009), under the same environmental conditions as our study (i.e., $\alpha = 0.54 \text{ day}^{-1}$ and total suspended particles (TSP) = $20 \mu\text{g}/\text{m}^3$), the estimated emission rate of DEHP is approximately $2 \mu\text{g}/\text{m}^2\cdot\text{hour}$ after one year. Multiplying the surface area of vinyl ($A_{\text{vinyl}} = 24.5 \text{ m}^2$) used in our study by the emission rate becomes $1.1 \text{ mg}/\text{day}$ (■, green square), which is comparable to our estimate, $0.74 \text{ mg}/\text{day}$.

3.5. Implications/Limitations

There have been extensive efforts to estimate emission rates of chemicals from individual indoor surfaces (Afshari et al., 2004; Clausen et al., 2004; Kemmlein et al., 2003; Xu et al., 2009; Xu and Little, 2006; Zhang et al., 2009, 2011). However, to date there has been a lack of understanding about how household air and dust concentrations are related to the whole house emission rate of chemicals released from a variety of consumer products and building materials. This study provides an alternate approach for estimating the source strength of a suite of organic compounds from a range of indoor sources in a whole house context where the individual sources of emissions are unknown. By combining direct measurements of dust concentrations with evaluation tools that include indoor mass-balance models, product-use-behavior emissions estimates, and diffusive flux from indoor surfaces, we present results from different approaches in this study. Applying these diverse tools to the same set of residential environments offers more insight on the range and reliability of the emissions estimates. Despite the lack of measurements from indoor sources for each house, the emission estimates from the fugacity model combined with measured dust concentrations provide results that are comparable to those inferred from personal care product use behaviors, highlighting that the household dust concentration is an effective indicator of source strength of indoor chemicals.

With respect to the useful insights for future work, the information on the proportion and type of indoor surfaces, including PVC plastics, couch, and vinyl flooring, needs to be collected from the field studies. In addition, for compounds commonly used in personal care

products, model predictions would be improved if the product use behaviors, including application rate and product type, are used as predictors of source strength. Furthermore, multiple measurements of dust samples in a single house over a long period of time could reduce uncertainty and variability in emission rate estimates that arise from having only a single measurement. Although foam-based furniture and fabrics are not included in the model, they are considered to be an important indoor sink in residential houses. Assuming that foam-based furniture has the same partition coefficient as carpet, increasing fugacity capacity of carpet by 50% results in decrease of emission rates from 4% (e.g., DEHP) to 33% (e.g., DEP), depending on the relative importance of carpet on the total indoor fugacity capacity. Thus, accounting for additional model compartments may improve the emission estimates. Our model assumes equilibrium between gas- and particle-phases, but recent studies have indicated gas-particle partitioning is not instantaneous for less volatile compounds, which may also impact our results (Liu et al., 2013; Shi and Zhao, 2012; Weschler and Nazaroff, 2008). The approach in this study offers insight on how to fill some of the large gaps in understanding emission rates for indoor chemicals in a whole house context.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Practical Implications

The combined dust-assay modeling approach in this study can be used to predict the source strength of indoor released compounds, integrating emissions from consumer products, building materials, and other home furnishings. Our findings show that estimated emission rates are closely related to not only the level of compounds on dust, but also the vapor pressure of the compound. Thus, a fugacity-based indoor mass-balance model and measured dust concentrations can be used to estimate the whole house emission rates from all sources in actual indoor settings, when individual sources of emissions are unknown.

Figure 1.

Box plots of measured dust concentrations ($\mu\text{g/g}$) of 15 studied compounds from 30 U.S. homes. Box lines indicate median concentrations, box edges correspond to the 25th and 75th percentile, and lines extend to minimum and maximum values. Measured dust concentrations are plotted on log 10 scale. Additional symbols represent the median from other published studies: ● USA (Guo and Kannan, 2011); ▼ Sweden (Bergh et al., 2011); ▲ Denmark (Langer et al., 2010); ■ Germany (Abb et al., 2009); ▲ Bulgaria (Kolarik et al., 2008); ▲ Sweden (Bornehag et al., 2005); ■ Germany (Fromme et al., 2004); ■ USA (Rudel et al., 2003); ▼ China (Lu et al., 2011); ▲ New Zealand (Ali et al., 2012); ▲ Belgium (van den Eede et al., 2011); ● USA (Meeker and Stapleton, 2010); ● USA (Stapleton et al., 2008); ■ Spain (Garcia et al., 2007); ▲ Germany (Wensing et al., 2005).

Figure 2.

Box plots of predicted emission rates in log₁₀ scale (mg/day). Box lines indicate median concentrations, box edges correspond to the 25th and 75th percentile, and lines extend to minimum and maximum values. Additional symbols represent the mean emission rate from different approaches and reported value: ● from personal care product use behaviors (Equation 15); ▲ from diffusive transfers of building materials to air (Equation 18); ■ reported value (Xu et al., 2009).

Figure 3.

Log (Vapor Pressure) versus the ratio of the measured dust concentration to the estimated emission rate. Regression results: $Y = -41.1X - 36.1$, $R^2 = 0.74$.

Table 1

Chemical Properties from EPI Suite

Product type	Abbr.	MW ^a	VP ^b	H ^c	S ^d	logK _{ow} ^e	logK _{ad} ^f	K _p ^g	k _{OH} ^h	D _{air} ^{i,j}
Phthalates	DEP	222.2	2.8E-01	6.2E-02	1.1	2.4	7.0	2.6E-06	3.0E-07	0.50
	DiBP	278.3	3.2E-01	6.5E-02	6.2E-03	4.1	8.4	6.3E-05	8.3E-07	0.43
	DnBP	278.4	2.7E-03	1.8E-01	1.1E-02	4.5	8.6	1.1E-03	8.0E-07	0.43
	BBP	312.4	1.1E-03	1.3E-01	2.7E-03	4.7	9.0	2.6E-04	9.6E-07	0.48
	DEHP	390.6	1.9E-05	2.7E-02	2.7E-04	7.6	12.6	8.9E-01	1.9E-06	0.42
Personal care product ingredients	DiNP	418.6	7.2E-05	1.5E-01	2.0E-04	9.4	13.6	9.4	2.0E-06	0.35
	AHTN	258.4	6.8E-02	1.4E+01	1.3E-03	5.7	7.9	2.2E-05	1.5E-06	0.34
	HHCB	258.4	7.3E-02	1.4E+01	1.8E-03	5.9	8.2	3.6E-05	3.3E-06	0.44
	ODP	277.4	4.7E-03	4.1E-01	2.0E-04	5.8	9.6	8.8E-04	1.2E-05	0.42
	OMC	290.4	1.8E-03	8.6E-01	1.5E-04	5.8	9.9	3.0E-04	4.5E-06	0.41
Flame retardants	TBPH	706.1	2.3E-09	3.1E-02	2.0E-09	12.0	16.9	1.8E+04	1.9E-06	0.32
	TBB	549.9	9.7E-08	5.6E-04	5.9E-07	7.2	13.8	1.7E+01	1.5E-06	0.39
	TCEP	285.5	1.3E-02	2.6E-03	7.0	1.4	5.3	5.0E-08	1.9E-06	0.49
	TPP	326.3	8.4E-04	4.0E-03	1.9E-03	4.6	8.5	7.0E-05	9.4E-07	0.42
	TDCPP	430.9	3.8E-05	2.6E-04	7.0E-03	3.7	10.6	1.0E-02	1.6E-06	0.41

^a Molecular weight (g/mol)

^b Vapor pressure (Pa)

^c Henry's law constant (Pa·m³/mol)

^d Solubility (g/L)

^e Octanol-water partition coefficient in log 10 (unitless)

^f Octanol-air partition coefficient in log 10 (unitless)

^g Partition coefficient between particles and air (m³/μg)

^h OH radical reaction rate constant (cm³/mol·day)

ⁱ Diffusion coefficient in pure air (m²/day)