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Predicting Residential Exposure to Phthalate Plasticizer Emitted from Vinyl Flooring: A Mechanistic Analysis

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A two-room model is developed to estimate the emission rate of di-2-ethylhexyl phthalate (DEHP) from vinyl flooring and the evolving gas-phase and adsorbed surface concentrations in a realistic indoor environment. Because the DEHP emission rate measured in a test chamber may be quite different from the emission rate from the same material in the indoor environment, the model provides a convenient means to predict emissions and transport in a more realistic setting. Adsorption isotherms for phthalates and plasticizers on interior surfaces, such as carpet, wood, dust, and human skin, are derived from previous field and laboratory studies. Log-linear relationships between equilibrium parameters and chemical vapor pressure are obtained. The predicted indoor air DEHP concentration at steady state is $0.15 \mu\text{g}/\text{m}^3$. Room 1 reaches steady state within about one year, while the adjacent room reaches steady state about three months later. Ventilation rate has a strong influence on DEHP emission rate while total suspended particle concentration has a substantial impact on gas-phase concentration. Exposure to DEHP via inhalation, dermal absorption, and oral ingestion of dust is evaluated. The model clarifies the mechanisms that govern the release of DEHP from vinyl flooring and the subsequent interactions with interior surfaces, airborne particles, dust, and human skin. Although further model development, parameter identification, and model validation are needed, our preliminary model provides a mechanistic framework that elucidates exposure pathways for phthalate plasticizers, and can most likely be adapted to predict emissions and transport of other semivolatile organic compounds, such as brominated flame retardants and biocides, in a residential environment.

Introduction

Since the 1930s, phthalates have been used as plasticizers to enhance the flexibility of rigid polyvinylchloride (PVC)

products (1), with worldwide phthalate production exceeding 3.5 million tons/year (2). About 90% of phthalates are used as plasticizers in polymers (e.g., PVC) and are found in a wide range of consumer products including floor and wall covering, toys, car interior trim, clothing, gloves, footwear, and artificial leather (3). Di(2-ethylhexyl) phthalate (DEHP) is most widely used and accounts for more than 50% of total phthalate production (3). The main use of DEHP is in PVC products such as vinyl flooring, where it is typically present at concentrations of ~20–40% (w/w) (4, 5). Other common phthalates are dibutyl phthalate (DBP), benzyl butyl phthalate (BBP), di-isononyl phthalate (DINP), and di-isodecyl phthalate (DIDP).

Because phthalates are not chemically bound in polymers, slow emission from the products to air or other media usually occurs. Adverse health effects of phthalates are briefly reviewed in the Supporting Information (SI). Phthalate esters have been recognized as major indoor pollutants (3, 4, 6). By sampling in 120 homes and analyzing for 89 organic chemicals, Rudel et al. (7) revealed that phthalates are one of the most abundant contaminants in indoor air. In the recent EPA-sponsored CTEPP (Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants) study (8), concentrations of over 50 target compounds were measured in multimedia samples from the homes and daycare centers of 260 preschool age children. The two phthalates targeted in the CTEPP study were detected in residential air and house dust, and on interior surfaces and dermal wipe samples. As in Rudel et al.'s study, measured phthalate concentrations were among the highest of any of the targeted compounds, including pesticides, PAHs, and PCBs. Despite this, only a few studies of phthalate emission characteristics and exposure are available. Uhde et al. (9) measured emission of several phthalates from PVC-coated wall-coverings in test chambers under standard room conditions. Clausen et al. (5) measured emissions of DEHP from vinyl flooring for more than a year in both the FLEC (field and laboratory emission cell) and the CLIMPAQ (chamber for laboratory investigations of materials, pollution, and air quality). In addition, the effect of humidity on the emission of DEHP from vinyl flooring was studied for one year in the FLEC (10), emission of phthalates from different types of plasticized product was studied for 150 days in the CLIMPAQ (11), and emission of phthalates from different types of plasticized materials was studied using a passive flux sampler (12).

Based on the Clausen et al. (5) experiments, Xu and Little (13) developed a model to predict the emission rate of phthalates from polymer materials. Their analysis revealed that emissions of the very low volatility semivolatile organic compounds (SVOCs) (such as DEHP) are subject to "external" control (partitioning into the gas phase, the convective mass-transfer coefficient, and adsorption onto chamber surfaces). The tendency of phthalates to adsorb strongly to surfaces is most likely similar to that of other SVOCs. Gebefügi (14) showed that SVOCs were sorbed by cotton and Van Loy et al. (15) found that more than 99% of recovered nicotine was adsorbed to the walls of their stainless steel chamber. Compared to these experimental chamber systems, the indoor environment has many other types of surface that will adsorb SVOCs such as DEHP to different extents. The emission rate measured in a test chamber may therefore be quite different from the emission rate from the same material in the indoor environment.

The model developed by Xu and Little (13) provides a convenient means to estimate the emission rate and gas phase

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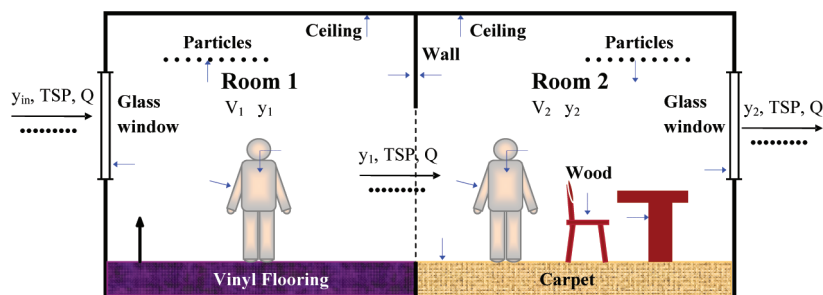


FIGURE 1. Schematic representation of the two-room model.

TABLE 1. Conditions for Two-Room Model

	room 1	room 2
volume (m × m × m)	3 × 3 × 3	3 × 3 × 3
ventilation rate (m ³ /h)	13.3	13.3
area of vinyl flooring, A _v (m ²)	9	
area of carpet, A _c (m ²)		9
area of glass window, A _g (m ²)	1.7 ^a	1.7 ^a
area of furniture, A _f (m ²)		20.3 ^a
area of ceiling and wall, A _{cw} (m ²)	41	41
total suspended particles, TSP (μg/m ³)	20 ^b	20 ^b

^a According to typical surface to volume ratio in residences (16). ^b Typical TSP in residential environment (17).

and adsorbed surface concentrations likely to occur in more realistic indoor environments. In this paper we both simplify and extend the Xu and Little model to investigate potential emission and distribution of DEHP in a residential environment. Field data collected in the CTEPP study as well as recent laboratory data are used to parameterize the extended model. The model is then used to estimate the emission rate and gas-phase DEHP concentration following the installation of vinyl flooring in a room. Finally, we examine the influence of two key parameters (air exchange rate and airborne particle concentration) on DEHP emissions, and estimate the potential exposure through inhalation, dermal absorption, and oral ingestion of dust.

Two-Room Model

To better estimate DEHP emissions in a residential environment the SVOC emissions model (13) was extended from a one-compartment description of an experimental chamber to a two-compartment representation of two adjacent rooms in a home (Figure 1). Vinyl flooring, the only source of DEHP considered, is placed in room 1, while carpet and wooden furniture are arranged in room 2. The room conditions are provided in Table 1.

Mass Balance. With reference to Figure 1, the accumulation of phthalate in room 1 obeys the following mass balance:

$$\frac{dy_1(t)}{dt} \cdot V_1 = Q \cdot y_{in}(t) - A_g \cdot \frac{dC_g(t)}{dt} - A_{cw} \cdot \frac{dC_{cw}(t)}{dt} - V_1 \cdot \frac{dF_1(t)}{dt} + A_v \cdot \dot{m}(t) - Q \cdot y_1(t) - QF_1(t) \quad (1)$$

where y_1 (μg/m³) is the gas-phase concentration as a function of time, V_1 (m³) is the room volume, Q (m³/h) is the ventilation rate, y_{in} (μg/m³) is the gas-phase concentration coming in from outside, F_1 (μg/m³) is the particle-phase concentration, \dot{m} (μg/m²h) is the vinyl flooring emission rate per unit area, A_v (m²) is the vinyl flooring surface area, A_g (m²) and A_{cw} (m²) are the surface area of glass and ceiling/wall respectively,

and C_g (μg/m³) and C_{cw} (μg/m³) are the corresponding surface concentrations. The accumulation of phthalate in room 2 is given by:

$$\frac{dy_2(t)}{dt} \cdot V_2 = Q \cdot y_1(t) + QF_1(t) - A_g \cdot \frac{dC_g(t)}{dt} - A_{cw} \cdot \frac{dC_{cw}(t)}{dt} - A_c \cdot \frac{dC_c(t)}{dt} - A_f \cdot \frac{dC_f(t)}{dt} - V_2 \cdot \frac{dF_2(t)}{dt} - Q \cdot y_2(t) - QF_2(t) \quad (2)$$

where y_2 (μg/m³) is the gas-phase concentration, V_2 (m³) is the room volume, F_2 (μg/m³) is the particle-phase concentration, A_c (m²) and A_f (m²) are the surface area of carpet and furniture respectively, and C_c (μg/m²) and C_f (μg/m²) are the corresponding surface concentrations. The gas-phase and particle-phase concentrations leaving room 1 are the same as those entering room 2, and represent the only source of DEHP in room 2.

DEHP Emission Rate. A detailed description of the mechanisms governing the emissions process is given by Xu and Little (13). While DEHP is present at a very high concentration in the vinyl flooring, as an SVOC with a very low vapor pressure, it emits very slowly. In fact, calculations show that even after 1 year only 0.003% of the total mass of DEHP has come out of the vinyl flooring. To simplify the model, we therefore assume that the vinyl flooring has an effectively constant DEHP concentration, and ignore the diffusion of DEHP within the vinyl flooring (Figure S1) (Table and Figure numbers preceded by an "S" are in the Supporting Information). Assuming a linear equilibrium relationship exists between the vinyl flooring and the gas phase, and considering the mass transfer within the boundary layer due to the concentration gradient, the emission rate is:

$$\dot{m}(t) = h_m \cdot (y_0 - y) \quad (3)$$

where

$$y_0 = C_0 / K \quad (4)$$

In equations 3 and 4, y (μg/m³) is the bulk gas-phase concentration, y_0 (μg/m³) is the concentration in the air immediately adjacent to the vinyl flooring surface, h_m (m/h) is the convective mass-transfer coefficient, K is the DEHP vinyl flooring/air partition coefficient, and C_0 is the constant DEHP concentration in the vinyl flooring.

Surface Adsorption. Although the partitioning behavior of phthalates between air and indoor surfaces is not known, partitioning of VOCs, phenanthrene, and nicotine has been studied (17–19). Assuming a linear equilibrium relationship for phthalates, the ratio of the concentration of a chemical on a surface to its concentration in the gas phase is equal to the surface/air partition coefficient, K_{surf} (m), or

$$K_{surf} = C_{surf} / y_{0,surf} \quad (5)$$

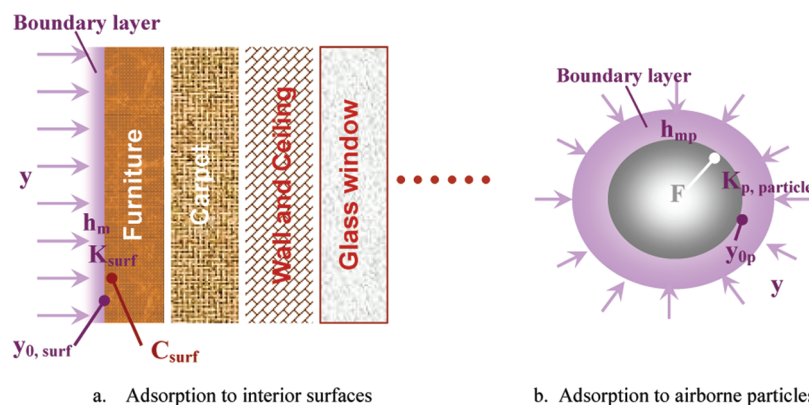


FIGURE 2. Schematic of sorption process. Note that the four individual materials shown for illustrative purposes in Figure 2a do not comprise a layered structure.

where C_{surf} is the surface concentration ($\mu\text{g}/\text{m}^2$) and $y_{0,\text{surf}}$ ($\mu\text{g}/\text{m}^3$) is the gas-phase concentration immediately adjacent to the surface (Figure 2a). Assuming a boundary layer exists adjacent to the adsorption surfaces, the amount of phthalate accumulated on the surface is equal to the total mass transferred through the boundary layer from the gas phase, or

$$dC_{\text{surf}}/dt = h_m(y - y_{0,\text{surf}}) \quad (6)$$

Equations 5 and 6 are assumed to apply to phthalate transferring between the bulk air and all exposed interior surfaces, such as carpet, furniture, glass window, ceiling/wall, and even human skin.

Sorption to Airborne Particles. For airborne particles (Figure 2b), an equation that has been used successfully to parameterize particle/air partitioning (20) is:

$$K_{p,\text{particle}} = (F/\text{TSP})/y_{0p} \quad (7)$$

where $K_{p,\text{particle}}$ ($\text{m}^3/\mu\text{g}$) is the particle/air partition coefficient for a given compound; F and y_{0p} are the particle and gas-phase concentrations ($\mu\text{g}/\text{m}^3$) of the compound, respectively; and TSP ($\mu\text{g}/\text{m}^3$) is the total suspended particulate material concentration. Thus, F/TSP is the fractional concentration of a given organic compound on the particles (e.g., μg of compound per g of particles). It is analogous to the fractional concentration of a compound in dust, in which case $K_{p,\text{dust}}$ is the dust/air partition coefficient. Similar to adsorption to the other surfaces, and assuming a boundary layer surrounding each particle, the accumulated particle-phase concentration is equal to the total mass transferred through the boundary layer, or

$$dF/dt = h_{mp} \cdot (y - y_{0p}) \cdot A_p \quad (8)$$

where h_{mp} (m/h) is the mass-transfer coefficient for particles, and A_p is the particle surface area (m^2/m^3 air). Equations 7 and 8 are assumed to apply to phthalate transfer between the bulk air and the airborne particles. The two-room model describing phthalate emissions and subsequent interaction with different interior surfaces and airborne particles is obtained by combining eqs 1 through 8. The vinyl flooring functions as a source only, but all other gas/surface interactions are fully reversible, and embody both equilibrium and kinetic mechanisms.

Sorption Isotherms for DEHP in Residential Environment
Estimation of Sorption Equilibrium Parameters on Interior Surfaces. To estimate partition coefficients for phthalate and different surfaces, data collected in a residential field study as well as data collected in a laboratory chamber study were used. In the CTEPP field study (8), 48-hour integrated samples

were collected simultaneously at each child's day-care center and at the child's home in either North Carolina or Ohio. The samples were collected from residential air, house dust, interior surfaces, and dermal hand-wipes. Only two phthalates (BBP and DBP) were measured in the CTEPP study. We therefore used the top three high-concentration pollutants (BBP, DBP, and bisphenol A (BPA)) in our analysis. BPA is also a semivolatile organic compound, which is typically present in commercial polycarbonate. Clausen et al. (5) conducted experiments in three CLIMPAQ chambers to study DEHP uptake by dust on PVC flooring. The DEHP concentrations in the dust and gas-phase will be used in the following analysis of DEHP partitioning between dust and air.

To establish surface/air equilibrium relationships for the three chemicals (BBP, DBP, and BPA) and the various indoor surfaces, we plotted the CTEPP data (e.g., DBP on adult skin and dust), as shown in Figure 3. Results for the other two chemicals and other surfaces are shown in the SI. The results suggest that simple linear relationships suffice to describe surface/air equilibrium in all cases. The statistical results for the regressions are summarized in Table S1. Based on eq 5, the intercept was set to zero. In all the linear regressions, the p -value was less than 0.05, with 95% confidence that the partition coefficient is different from zero. Furthermore, the p -value of the Shapiro test was larger than 0.05, with 95% confidence that the regression residues are normally distributed.

Unfortunately, according to the sampling method of the CTEPP study, the measured concentrations in the indoor air "y" are actually the total airborne concentrations; that is, the sum of the gas phase and airborne particle concentration "y + F". To obtain a more representative gas/surface partition coefficient, the equilibrium relationships need to be modified, or

$$K_{\text{surf}} = K_{\text{surf,CTEPP}} \cdot (y + F)/y \quad (9)$$

where $K_{\text{surf,CTEPP}}$ is the equilibrium coefficient in Table S1 derived from the CTEPP study, and K_{surf} is the desired gas/surface partition coefficient. As discussed in the SI, the modified K_{surf} for each compound is listed in Table S4 in bold. Because K_{surf} is derived from field measurements, it includes the contribution of chemicals sorbed to airborne particles that subsequently deposit on surfaces.

We now have approximate equilibrium relationships for BBP, DBP, and BPA between indoor air and various surfaces (hardwood floor, carpet, child skin, adult skin, and dust), as well as the directly measured equilibrium relationship for DEHP between indoor air and dust (5). What we need, however, are the equilibrium relationships between DEHP and hardwood floor, carpet, and human skin. To get these,

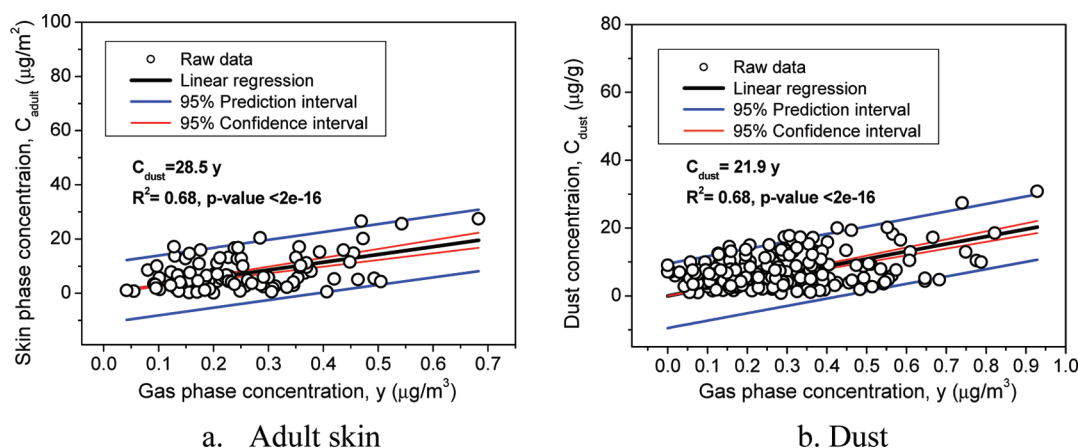


FIGURE 3. Linear regression results for DBP.

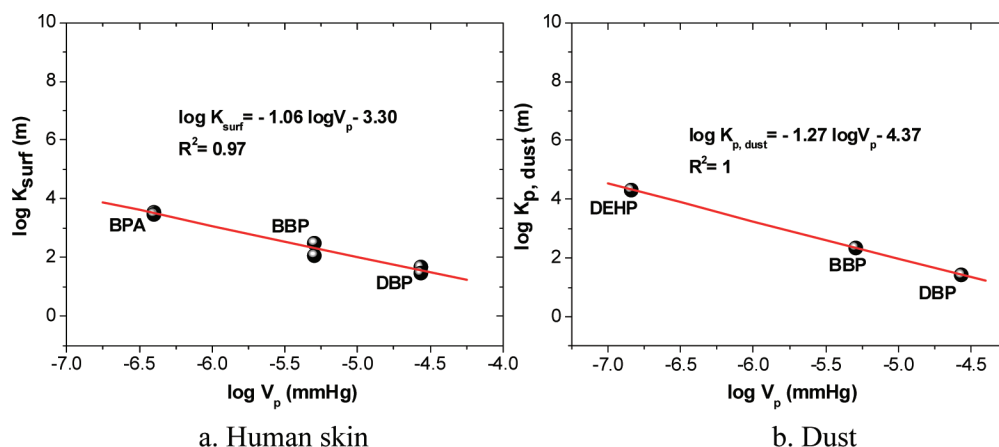


FIGURE 4. Linear regression between $\log(V_p)$ and $\log(K_{surf})$.

TABLE 2. Partition Coefficients for DEHP

surface	partition coefficient, K	isotherm exponent, n
furniture, wall and ceiling ^a	2500 (m)	1.5
carpet ^b	1700 (m)	
glass ^c	3800 ($\mu\text{g}/\text{m}^2$) / ($\mu\text{g}/\text{m}^3$) ⁿ	
skin ^d	9500 (m)	
airborne particles ^e	0.25 (m^3/ug)	
dust ^f	21100 (m^3/g)	

^a Calculated using $\log K_{surf} = -0.779 \log V_p - 1.93$, Figure S6. ^b Calculated using $\log K_{surf} = -0.627 \log V_p - 1.08$, Figure S6. ^c Xu and Little fitted the Freundlich isotherm for glass. ^d Calculated using $\log K_{surf} = -1.06 \log V_p - 3.30$, Figure 4. ^e Calculated using $\log K_{p, particle} = -0.860 \log V_p - 4.67$, eq S3. ^f Regression result of Figure S5.

we developed simple correlations between the equilibrium parameters and the vapor pressure of the target chemicals.

Correlation of Equilibrium Parameters with Vapor Pressure. Correlations were obtained between vapor pressure (V_p) and sorption parameters (K_{surf}) for different interior surfaces, including settled dust. Linear relationships between $\log(V_p)$ and $\log(K_{surf})$ were found (e.g., human skin and dust) as shown in Figure 4. Results for all surfaces are shown in Figure S6. Data for child hand wipe and adult hand wipe were combined to get the relationship between the human skin partition coefficient and vapor pressure. The partition coefficient of BPA for dust did not conform well to this relationship, thus Figure 4b only shows the relationship between phthalate vapor pressures and partition coefficients for dust.

While using only three chemicals does not provide a conclusive relationship, the overall results suggest that it is

possible to relate the equilibrium partition coefficients to vapor pressures. Finally, we used the new correlations to obtain the partition coefficient for DEHP on different interior surfaces, as shown in Table 2. The isotherm used for glass is based on a previous study (13), although the nonlinear nature of the isotherm may have been due to the data fitting procedure. In the SI, the skin/air partition coefficient is checked using a completely different procedure and shown to be acceptable.

Mass Transfer Coefficient. The value of h_m , the mass-transfer coefficient for the boundary layer adjacent to the various surfaces, was estimated using correlation equations (21), which express h_m as a function of Reynolds number and Schmidt number. Huang et al. (22) measured air velocities in a typical house in the United States. They found velocities with a range of 0.01–0.16 m/s and showed that values near the floor are higher than those in the center of the room. In

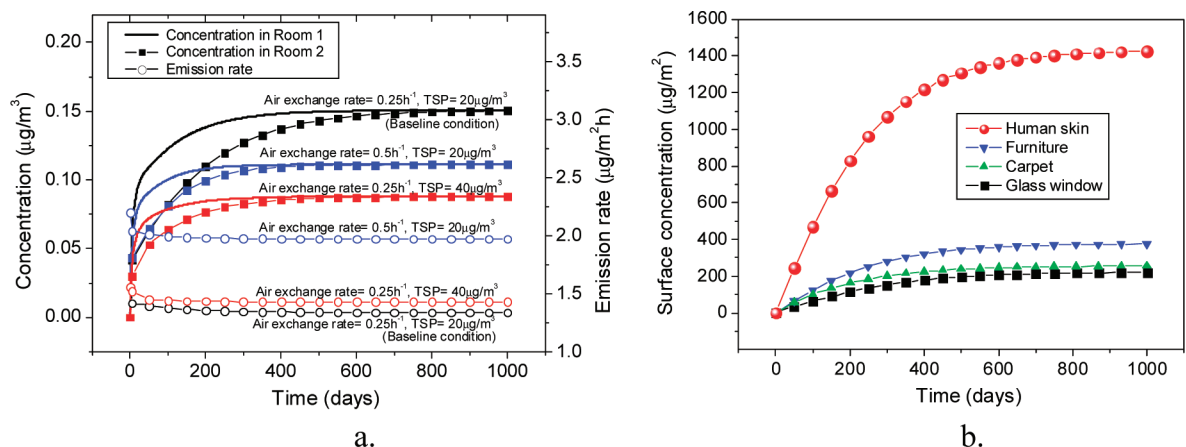


FIGURE 5. (a) Effect of air exchange rate and TSP concentration on DEHP concentration and emission rate. (b) Predicted DEHP concentration on interior surfaces.

TABLE 3. Concentration of Phthalates in Indoor Air and Household Dust Samples

DEHP	references	n	mean	max	our study
gas phase concn ($\mu\text{g}/\text{m}^3$)	(25)	40	0.48	1.6	0.15
	(26)	125	0.14		
	(7)	102	0.07	1.0	
	(4)	59	0.19	0.4	
dust phase concn ($\mu\text{g}/\text{g}$)	(25)	12	950	3100	3000
	(27)	600	1200	3500	
	(7)	101	340	7700	
	(4) and (28)	30	776	1542	

the CLIMPAQ chamber (5), which roughly approximates conditions in a real room, the velocity at the test piece surface was estimated to be 0.15 m/s and this value is used to estimate h_m . Odum et al. (23) measured mass transfer of PAHs and others SVOCs to and from combustion aerosols at 25 °C, and their result is used here as h_{mp} , the mass-transfer coefficient for particles.

We now have estimates of all the partition coefficients for DEHP between indoor air and interior surfaces (hardwood floor, carpet, human skin, and particles), as well as the associated mass transfer coefficients. We are therefore able to use the two-room model to estimate the emission rate and evolving gas-phase DEHP concentration following the installation of vinyl flooring in room 1.

Because no other data for DEHP concentrations on real interior surfaces are available, the CTEPP study provides the only available data that we can use to estimate DEHP adsorption isotherms. Even though the values of the partition coefficient for DEHP on interior surfaces can only be considered rough estimates, we showed in a sensitivity analysis (24) that they do not have a strong influence on the steady state indoor air DEHP concentration, which is the basis for our exposure analysis (although they do influence the time it takes to reach steady state). We therefore believe that our emissions and transport model represents a reasonable first step.

Results and Discussion

For baseline conditions (Table 1), the indoor air DEHP concentration at steady state is $0.15 \mu\text{g}/\text{m}^3$. As shown in Table 3, this value is similar to that measured within homes in both the United States and Europe, although it should be emphasized that vinyl flooring is the only source of DEHP considered here. Room 1 reaches steady state within about one year, while the adjacent room reaches steady state about

three months later. Airborne particles increase the rate at which DEHP is transported between rooms by a factor of 5 relative to gas-phase transport. The boundary layer surrounding the airborne particles is much thinner than the boundary layer adjacent to the other indoor surfaces and the suspended particles reach equilibrium with the gas-phase much more rapidly than the larger surfaces. Suspended particles are therefore very effective at transporting DEHP from one room to another, because DEHP also desorbs very rapidly from the particles.

In Figure 5a, the impact of air exchange rate and total suspended particle concentration on the DEHP emission rate and the DEHP concentration in rooms 1 and 2 is examined. Increasing air exchange rate will increase the DEHP emission rate from the vinyl flooring significantly while an increase in the TSP concentration causes a substantial decrease in the gas-phase concentration in both rooms, but increases the emission rate in Room 1. An increase in the air exchange rate was assumed to double the velocity of the air above the vinyl flooring (from 0.15 to 0.30 m/s) and this higher value was used to calculate the h_m associated with the flooring.

Figure 5b shows the predicted DEHP concentration change with time on various interior surfaces in room 2. The predicted DEHP concentration on human skin is 5–7 times higher than on the other surfaces due to the high skin/air partition coefficient for DEHP. The skin/air partition coefficient was obtained from hand-wipe samples in the CTEPP study. It is generally believed that these hand-wipe samples are measuring chemicals transferred from indoor surfaces onto the hands directly. However, the fact that the skin/air isotherms determined for both adult and child are almost identical for DBP, BBP, and BPA (see, for example, Figure 4a), suggests that SVOCs may be transferring directly from the air to the skin, or that if large amounts are picked up by direct dermal transfer, that some desorbs to re-establish

equilibrium with the air. Indeed, in a subsequent paper (24), we show that there is a strong correlation between the concentrations of DBP, BBP, and BPA on skin and those in the gas phase, but almost no correlation with those on interior surfaces. This further suggests that certain SVOCs may reach the skin through the gas phase, and not via dermal transfer as is commonly suspected.

Estimating Exposure to DEHP from Vinyl Flooring.

Based on the model results, we are interested in evaluating exposure to vapor phase DEHP in air, particle bound DEHP in air, and DEHP in settled dust. The exposure pathways of interest are inhalation of vapor, inhalation of particles, dermal absorption of DEHP deposited on the skin, and oral ingestion via household dust.

The detailed exposure calculations are shown in the SI. For dermal exposure, the overall skin permeability coefficient, P , is controlled by permeation through the skin ($P_{\text{skin/air}}$) as well as by permeation through the air boundary layer adjacent to the skin (P_{air}), or:

$$P = \frac{1}{\frac{1}{P_{\text{skin/air}}} + \frac{1}{P_{\text{air}}}} \quad (10)$$

where $P_{\text{skin/air}}$ (cm/hr) is vapor to skin permeability, and P_{air} (cm/hr) is permeability of the boundary layer. For low volatility compounds, convective mass transfer through the air boundary layer adjacent to the skin may become the rate limiting factor, and this is the case for DEHP. As detailed in the SI, the estimated value of P is 580 cm/hr.

As shown in Table S8, the reference dose (RfD) for DEHP is 20 $\mu\text{g/kg/d}$ according to the U.S. EPA. Airborne particles contribute 80% of the inhalation exposure, although the highest value of total inhalation exposure is less than 0.6 $\mu\text{g/kg/d}$, which is much lower than the RfD. For infants, exposure through oral intake via dust is 1.6 times higher than the RfD, although the estimate for dust intake rate of 10.3 mg/kg/d (29) may be high. Exposure via these two pathways is similar to other study results (6, 30, 31). Dermal absorption of DEHP deposited on skin is greater than that taken up through inhalation. For DEHP, the primary route of exposure is oral ingestion of dust. Overall, children experience 2–10 times higher exposure risk than adults based on all exposure pathways.

Acknowledgments

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Supporting Information Available

Further details on the regression results for the CTEPP data, modification of surface partition coefficients, and detailed exposure calculations. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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