

Direct and Air-Mediated Transfer of Labeled SVOCs from Indoor Sources to Dust

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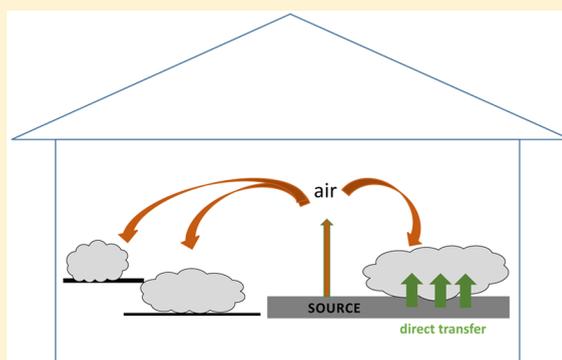
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Supporting Information

ABSTRACT: Two small-scale field studies were conducted to investigate the transfer of substances from products into dust due to direct and air-mediated transfer. The project focused on semivolatile organic compounds (SVOCs), which are frequently found in and re-emitted from dust. For the field studies, four artificial products containing deuterium-labeled SVOCs (eight phthalates and adipates) were installed in residential indoor environments. Two plastic products were installed vertically to investigate substance transfer due to evaporation into air. One plastic product and a carpet were installed horizontally to investigate the direct transfer from source to dust. A pyrethroid was intentionally released by spraying a commercial spray. Dust samples were collected from the floor, elevated surfaces in the room and the surfaces of the horizontally installed products. We observed that the dust concentrations of substances exclusively transferred via air were similar at different collection sites, but the concentrations of chemicals present in horizontal products were up to 3 orders of magnitude higher in dust deposited on the source. We conclude that direct transfer from source into dust substantially increases the final SVOC concentration in dust in contact with the source, regardless of the vapor pressure of investigated SVOCs, and may lead to larger human exposure.



1. INTRODUCTION

In industrialized countries, people spend most of their time indoors^{1,2} where they are continuously exposed to dust, an important secondary source of chemical exposure.^{3,4} Dust contributes substantially to human exposure to various chemicals^{4–9} including plasticizers^{5,8,10} and insecticides.^{11,12} Particularly for low-volatile substances released by using consumer products, dust is an important transfer and storage medium^{10,13} and may cause internal exposure to semivolatile organic compounds (SVOCs) as evidenced by recent biomonitoring studies.^{14–17} Despite the importance that dust may have for human exposure and associated health risks, many open questions remain regarding the transfer processes and efficiency.^{18–20} Improving the mechanistic understanding of SVOC transport into dust helps to identify the most important nondietary sources for human exposure, allowing in turn for more effective risk mitigation.

The estimation of substance concentrations in dust in most models is based on the assumption that the substances are first emitted into the boundary air layer, then transfer to bulk air and finally are absorbed by dust.^{8,21–23} However, another possible transfer pathway is direct substance transfer from product to

dust, where the transfer to bulk air is not taking place. The direct transfer may occur (1) through abrasion or wear of the product,²⁴ (2) mediated by the boundary air layer at the source/air interface,^{19,20} or (3), as hypothesized by Clausen et al.,¹⁸ directly into the dust particles that act as lipophilic sorbents.^{20,24}

In chamber experiments, the direct transfer of di(ethyl-hexyl) phthalate (DEHP),¹⁹ polychlorinated biphenyls (PCBs),³ polybrominated diphenyl ethers (PBDEs)²⁴ and hexabromocyclododecanes (HBCDs)²⁰ to house dust was investigated. In a test house, Bi et al.²⁵ have compared the concentration of benzyl butyl phthalate (BBzP) in dust accumulated on a source of BBzP (floor) and other nonsource surfaces. The authors have shown that transfer of BBzP into dust is much faster when house dust is in direct contact with the source in comparison to the transfer via air. However, to our knowledge, direct transfer has not yet been investigated in real, inhabited apartments.

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Table 1. Products and Substances Used in the Field Studies

Product	Symbol	Abbr.	Field study	Substance name	Abbr.	Emission process
Vertical plastic		PV	1	² H ₄ -di-butyl adipate	DBA	Evaporation
			1, 2	² H ₄ -di-(ethyl-hexyl) phthalate	DEHP	
Vertical plastic with thermal exposure		PT	1	² H ₄ -di-isobutyl adipate	DiBA	Evaporation under thermal stress
			1, 2	² H ₄ -di-(ethyl-hexyl) adipate	DEHA	
Vertical plastic-2 (no thermal exposure)		PT-2	2	² H ₄ -di-isobutyl adipate	DiBA	
Horizontal plastic		PH	1	² H ₄ -di-isobutyl phthalate	DiBP	Evaporation and direct diffusion to dust particles
			1, 2	² H ₄ -di-octyl adipate	DOA	
Horizontal plastic-2		PH-2	2	² H ₄ -di-isobutyl phthalate	DiBP	
Carpet		C	1, 2	² H ₄ -di-butyl phthalate	DBP	Evaporation and abrasion
				² H ₄ -di-octyl phthalate	DOP	
Spray		S	1, 2	Tetramethrin	Tetr	Intentional release

The importance of the direct transfer of SVOCs from a surface to dust in contact with that surface was hypothesized in previous studies.^{18,23,26} However, the mechanism of direct transfer is not well understood, and it is not known for which substances this transfer is relevant.

In indoor field studies dust samples are often collected from a single surface (e.g., floor).^{3,8,10,27–30} Measured substance concentrations in dust are then used for human exposure assessment, even though it is not clear whether floor dust is a good proxy for the dust fraction that is inhaled ($\leq 10 \mu\text{m}$),^{31,32} or whether the single sample is representative for the whole room (i.e., whether the dust is homogeneously distributed). The direct transfer may even enhance dust inhomogeneity, since larger amounts of substances may be transferred to dust deposited on the surface of the source than elsewhere and lead to greater human exposure. Hence, the sampling location of the settled dust needs to be carefully selected.

The objective of this study was to experimentally investigate the influence of sampling location on substance concentrations in dust. The substance concentrations in floor dust, dust on elevated surfaces (here “lifted dust”) and dust deposited on the surface of the horizontal products then formed the basis to investigate (1) the importance of the direct substance transfer to dust in contact with the source in comparison to air-mediated transfer and (2) dust homogeneity indoors.

2. MATERIALS AND METHODS

Two field studies were conducted: The first study had made use of five inhabited residential apartments for 12 weeks, and the second study involved three apartments selected from the previous five for 8 weeks. Both studies were conducted during the same season of the year (autumn) with a nine-month break in-between, which ensured low background concentrations of the target substances (see [Methods](#) section). In a previous publication, we reported the substance distribution in the room based on the concentrations in air and floor dust samples collected during the first field study.³³ Here, we focus on investigating direct substance transfer from the source to house dust during both field studies by analyzing concentrations in floor dust, lifted dust, and dust in direct contact with the source.

3.1. Materials. Nine SVOCs were selected as target substances for the field studies (see [Table 1](#)). To enable tracking of ubiquitous plasticizers without interfering background concentrations, eight deuterium-labeled phthalates and adipates were synthesized. The labeled plasticizers were incorporated into four artificial products: three plastic products and a carpet. Two labeled plasticizers with different octanol–air partition coefficients (K_{oa}) were incorporated into each product so that all products contained a unique combination of labeled substances. A commercial spray containing the synthetic pyrethroid tetramethrin (0.16 g/100 g) (which is seldom applied indoors) was purchased from Martec AG.

In the first field study, the plastic products were installed in the rooms as follows: one plastic product was laid down on a horizontal surface (PH), one was installed vertically (PV), and one was installed vertically and additionally exposed to heat (PT) using a 60 W electric lamp for 6 h/day. The plastic products PH, PV, and PT consisted of 3, 4, and 2 LDPE sheets ($24 \times 18 \times 0.01 \text{ cm}^3$), respectively. The products were prepared as reported previously.³³ An insecticide spray containing pyrethroid was applied once a week during the first 5 weeks of the field study by spraying for at least five seconds in the middle of the room.

For the first field study, this set of five products (three plastic products, a carpet and a spray) was installed in each of the five apartments in Zurich, Switzerland. In all apartments only one room was selected as the experimental area (normally living rooms, bedroom for apartment 4). Apartment 2 was ventilated mechanically (continuous ventilation), while the others were ventilated by opening windows and doors. Details of the sample installation have been reported previously.³³

During the second field study, three apartments selected from the previous five (apartments 2–4) were investigated. After the first study, all the plastic products were rolled in aluminum foil, placed in resealable plastic bags and stored at $-20 \text{ }^\circ\text{C}$ to reduce losses of the chemicals. The carpets were tightly rolled in aluminum foil and stored in the basement. After a nine-month break, the same products that were used during the first study (no materials added) were installed again for the second field study in the same locations of the respective apartments. Since the most volatile substances

(DiBA, DBA and DiBP) had been completely released from the plastic sheets during the first study (see Table 2), two additional products (PH-2 containing DiBP and PT-2 containing DiBA) were produced and installed to keep both field studies more consistent. However, the source of DBA was not renewed. Further, PT and PT-2 were not exposed thermally, because in the first field study no significant difference had been found between heated and nonheated products.³³

3.2. Methods. Sampling and Sample Analysis. The two field studies were performed between September–December 2013 and September–November 2014, respectively. During the first field study, all apartments were visited every second week to collect floor dust samples. Passive air samples, lifted dust (dust from surfaces elevated above the floor, e.g., book shelves) and dust on the surface of the horizontally installed product were sampled every 4 weeks (see Supporting Information (SI)). During the second field study, both dust and passive air samples were collected every 4 weeks. Lifted dust samples were collected from two (apartments 2 and 4) or three (apartment 3) surfaces in the room. Additionally, dust from the surfaces of both horizontal plastic products (PH and PH-2) was analyzed. Furthermore, the floor area was divided into three parts that were sampled separately: (1) The surface of the experimental carpet was vacuumed with a hand-vacuum cleaner. (2) Dust from a well-defined, small (0.1–0.7 m²) floor area was collected on a filter using the nozzle attached to the vacuum cleaner: the same sampling technique that was also used for the elevated surfaces (hereafter referred to as “filter collection”). In order to assess the distribution pattern, in apartment 3 such floor dust samples were collected from three small, defined areas in the room. (3) The remaining floor area was vacuumed with an ordinary vacuum cleaner. Detailed explanations of the sampling procedure and sample preparation can be found in the SI. Details of the quantitative analytical method using GC/MS have been reported previously.³³

For quality control, background, blank and triplicate samples were analyzed (see SI). Before the first field study, background dust and active air samples confirmed that the background concentrations of tetramethrin were below the limits of detection (LOD) of 5.7 ng/m³ for air and 0.24 µg/g for floor dust.³³ Background samples collected before the second field study showed that the target substances had been almost completely removed from the environment during the nine-month break between both studies (see SI). The blank samples showed no sample contamination. The triplicate samples were analyzed to investigate the measurement uncertainty and showed an average relative standard deviation of 20%.³³ For all substances and matrices, the LODs were equal to the limit of quantification in our study. For dust samples, LODs are sample-specific since they were calculated on a mass-basis and adjusted to each sample depending on its size.³³ Measured values that were below the corresponding LOD (i.e., could not be quantified) were treated as nondetects. In figures, nondetects are presented as LODs, while in calculations half of corresponding LODs were used. More sophisticated methods of treating nondetects in the calculations were not suitable because of the small sample number.³³

Mass Balance. The total amount of each substance in air during the whole period of the field study was calculated based on measured air concentrations, volume of the room and air exchange rate.³³ Air exchange rates were measured based on the carbon dioxide generated by the occupants as a tracer gas and

were reported previously.³³ For dust, the total amount is split between floor dust, lifted dust and dust on the surface of the product. These three dust types were sampled separately. For floor dust and dust on the product, the entire available surfaces were sampled. Therefore, for the mass balance it could be assumed that floor dust had been sampled quantitatively. For the lifted dust, however, only a fraction of the total surface had been sampled. For calculating the total amount of substance on elevated horizontal surfaces in the room, it was therefore assumed that the total surface area of these surfaces equals the floor area with a dust loading of 0.3 g/m².³⁴ The theoretical amounts of substances sorbed onto textiles, polyurethane foam contained in a sofa and other sorptive surfaces were calculated from the corresponding surface-air partition coefficients and the average gas phase concentrations (C_{air}), which were calculated from the total air concentrations (C_{tot}) measured during the field study using an approach suggested by Weschler and Nazaroff:^{22,23}

$$C_{\text{air}} = \frac{C_{\text{tot}}}{1 + \frac{\text{TSP}}{\rho_{\text{part}}} \times f_{\text{om_part}} \times K_{\text{oa}}}$$

TSP is “total suspended particles”, ρ_{part} is the density of airborne particles, and $f_{\text{om_part}}$ is the fraction of organic matter associated with airborne particles (see SI).

The surface-air partition coefficients were approximated by K_{oa} values.^{22,25} Foam-air and textile-air partition coefficients were extrapolated from values reported in the literature (see SI).^{35,36} More details on the mass balance calculation are given in the SI.

4. RESULTS

Concentrations of target substances in plastic products were measured both before and after the field studies (see Table 2). For analysis, usually 12 discs (with a nominal diameter of 4.5 mm) were cut from different locations in each of the products to ensure representative samples. Since the doping technique of the carpet did not allow homogeneous doping, the total amount of substance applied on the carpet was assumed to equal the initial amount (before the first field study) in the carpet.³³ For the second field study, the same carpets were used. Since the amounts measured before and after the second study were not consistent (sometimes amounts before were less than afterward), no initial value could be calculated. Instead, the average amounts before and after the second field study are provided in Table 2. This approach has no direct influence on our results since we are only reporting trends, and the initial values serve to give an indication of roughly which amount of substance was available for transfer to air and dust. The amount of tetramethrin released by spraying was assessed by weighing the spray can before and after application.

Table 2 presents average amounts of the deuterium-labeled substances before and after both field studies. Relative standard deviations of the initial amounts (before the first study) show the variability between the plastic products produced in different batches (PT products were produced in a single batch). Relative standard deviations of the amounts after both field studies and before the second study represent the variability between products in all apartments investigated in the study: five and three apartments during the first and second study, respectively.

The amounts of substances installed in the apartments were comparable during both field studies, with the amount of less

Table 2. Log K_{oa} Values and Average Amounts of the Labeled Target Substances in the Products

Substance	DiBA	DBA	DiBP	DBP	DEHA	DOA	DEHP	DOP	Tetr ^b	
log K_{oa} ^a	7.9	8.0	8.8	8.9	10.8	10.9	11.7	11.9	12.2	
Product										
1 st field study	Amount in the product before the experiment, mg	15	30	26	80 ^c	190	310	280	800 ^c	160
	Relative standard deviation before the experiment, %	n.a.	11	8.0	n.a.	n.a.	5.9	15	n.a.	-
	Concentration in the product before the experiment, kg/m ³	1.8	1.8	2.0	0.059 ^c	22	24	16	0.59 ^c	-
	Amount in the products after the experiment, mg	0.0016	0.12	0.59	n.a.	120	240	190	n.a.	0
	Relative standard deviation after the experiment, %	65	50	118	n.a.	4	14	3	n.a.	28
	Concentration in the products after the experiment, kg/m ³	$< 2.9 \times 10^{-4}$	6.7×10^{-3}	4.5×10^{-2}	n.a.	14	18	11	n.a.	-
2 nd field study	Amount in the product before the experiment, mg	19	-	40	50 ^d	120	230	155	500 ^d	94
	Relative standard deviation before the experiment, %	3.0	-	6.0	n.a.	1.4	12	5.5	n.a.	-
	Concentration in the product before the experiment, kg/m ³	2.2	-	3.1	0.038 ^d	14	18	9.0	0.38 ^d	-
	Amount in the products after the experiment, mg	0.020	-	2.6	n.a.	29	37	59	n.a.	0
	Relative standard deviation after the experiment, %	73	-	23	n.a.	5	11	14	n.a.	36
	Concentration in the products after the experiment, kg/m ³	2.3×10^{-3}	-	0.20	n.a.	3.3	2.9	3.4	n.a.	-

^aValues from EpiSuite Version 4. ^bOn average 160 mg (1st field study) and 94 mg (2nd field study) of tetramethrin were released from a commercial spray containing 0.16% w/w of tetramethrin. ^cAssumed values based on substance amount doped to the carpet (weight 1.545 kg, density 1140 kg/m³). ^dAmounts before and after the study were averaged; n.a.: not applicable (see text).

volatile substances a factor of 5–10 higher in comparison to the more volatile substances. The durations of the field studies (12 and 8 weeks) were sufficient for more volatile plasticizers (DiBA, DBA, and DiBP) to be almost completely depleted from their sources (residual 0.01–6.5%). Loss of less volatile substances was smaller and varied between 23–37%³³ and 34–62% of the initial amount in the product (before the first field study) during the first and the second study, respectively. However, the absolute released amounts of the less volatile substances were higher in comparison to the more volatile substances.

Concentration Patterns in Floor Dust. During the first field study, floor dust samples were collected by vacuuming the total floor area including parts covered by carpets. Then, the floor dust samples were sieved using a 2 mm sieve. Both dust fractions were analyzed separately: the results of this analysis have been reported previously.³³ Briefly, it was observed that the difference between the two fractions was small; therefore, during the second field study no sieving was applied. Moreover, during the first field study the phthalates present in the carpet (DBP and DOP) were detected in all floor dust samples and in much higher concentrations in comparison to other substances.

Therefore, to investigate whether the carpet substances are distributed in these high concentrations homogeneously in the rooms (i.e., whether there would be a difference between dust on the surface of the carpet and dust on the remaining floor area), during the second field study floor dust was divided into three samples: dust from the carpet (vacuuming), dust from a small defined area on the floor (filter collection) and dust from the remaining floor area (vacuuming) (see Figure 1a). Since in apartment 3 dust samples were collected from three small, defined areas in the room, in Figure 1a these were averaged for better comparison to the other apartments. If the substance was not detected in dust collected from at least two out of three surfaces, the average concentration in the figure is marked as nondetect. In the figures, the concentrations are normalized to the amount of substance (source strength) at the beginning of the corresponding field study.

DBP was detected in dust sampled directly from the surface of the carpet, but apparently it had not been transferred to the dust accumulating on the remaining floor area (see Figure 1a). Moreover, DBP concentrations in carpet dust were on average a factor of 48 higher compared to concentrations of other more volatile substances (DiBA, DiBP). The DOP concentrations in

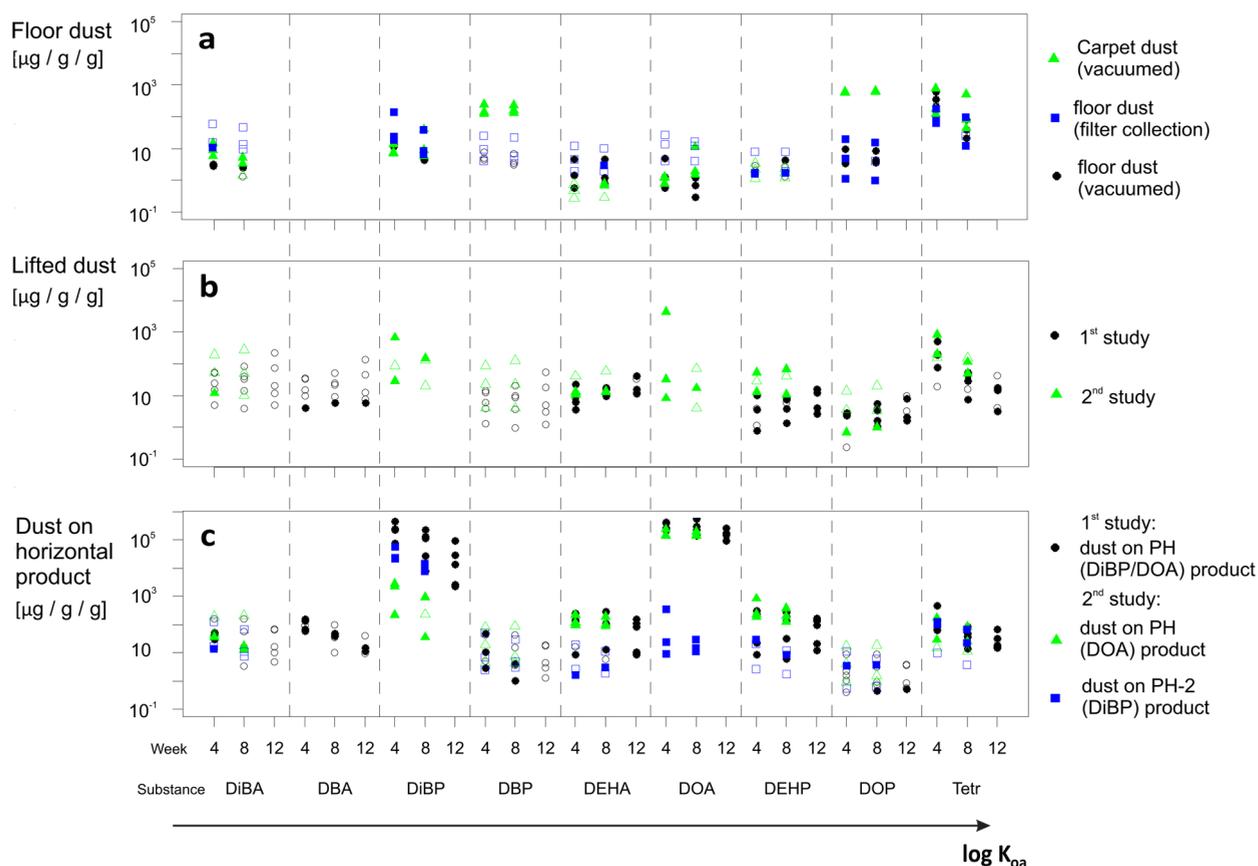


Figure 1. Measured normalized concentrations ($\mu\text{g/g}$ dust/ g substance) of target substances in (a) three types of floor dust from all apartments during the second field study, (b) lifted dust of all apartments during both field studies and (c) dust on the surface of the horizontally installed products of all apartments during both field studies. Nondetects were substituted by the detection limits normalized to the initial strength of the sources and are represented as “empty” symbols.

carpet dust were also more than 2 orders of magnitude higher in comparison to other less volatile substances contained in plastic products and on average a factor of 130 higher in comparison to the DOP concentration in floor dust. DOP was detected in dust across the entire floor area.

In agreement with the first field study,³³ concentrations of more volatile substances (with smaller K_{oa}) in floor dust were higher in comparison to the less volatile plasticizers.

Concentration Patterns in Lifted Dust. During the first field study, lifted dust samples were collected from one elevated horizontal surface in each apartment. During the second field study, among other elevated surfaces, this same elevated surface was sampled (see Figure 1b).

The concentrations of DiBP and DOA from the first field study are not presented in Figure 1b. In the first field study, lifted dust samples were collected with the same nozzle that was used for the collection of dust deposited on the source (cleaned on site; see section “Dust in contact with the source”). The concentrations of DiBP and DOA during the first field study were much higher in comparison to the second field study (see SI). Thus, we assume that in the first field study the lifted dust samples had been cross-contaminated with substances contained in the horizontal product.

The results show that in the first field study the only measurable concentrations of the more volatile substances were for DBA, while in the second field study only DiBP (and one value for DiBA) were detected (see Figure 1b, DBA was not

investigated in the second field study). The less volatile substances were detected in most of the lifted dust samples.

During both field studies, in apartment 2 none of the target chemicals had been detected in lifted dust (see SI). The main reason for this diverging substance distribution may be that the inlet of fresh air of the continuous mechanical ventilation system was situated right above the sampling surface.

Dust in Contact with the Source. During the first field study dust samples were collected from the surface of the horizontally installed product (PH) containing DiBP and DOA. In the second field study, dust samples were collected from both horizontally installed products: PH containing only DOA and PH-2 containing only DiBP. Figure 1c shows concentrations of all target substances in dust on the horizontal products. It can be seen that dust accumulating directly on the surface of the horizontally installed products contains up to 4 orders of magnitude higher concentrations of the substances that are contained inside the horizontal product (DiBP and/or DOA) in comparison to the other substances (see Figure 1c). The concentrations of DiBP in dust collected from the surface of the PH product during the second field study are slightly elevated, possibly due to a small amount of DiBP remaining in the products after the first study.

Dust Homogeneity Indoors. During the second field study lifted dust samples were collected from two (apartments 2 and 4) and three (apartment 3) surfaces in the room. In apartment 3 (see Figure 2), concentrations in dust on all three surfaces were very similar. For all substances and both sampling

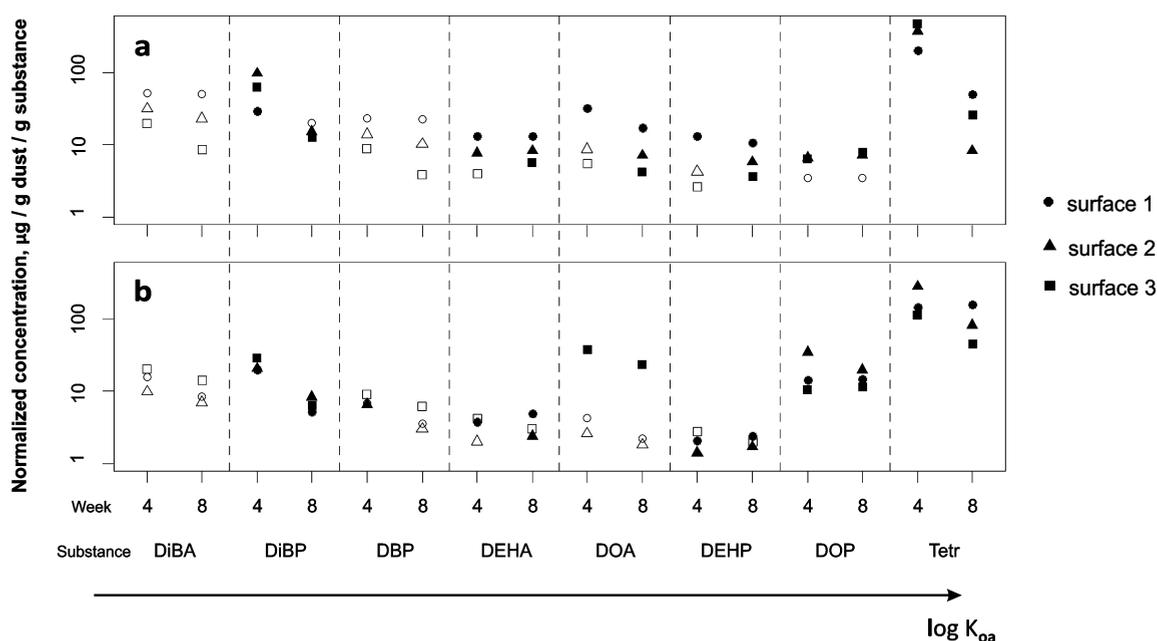


Figure 2. Normalized concentrations ($\mu\text{g/g dust/g substance}$) of target substances measured in apartment 3 in (a) lifted dust collected from three elevated surfaces and (b) floor dust collected from three small, defined surfaces on the floor using the same sampling technique (filter collection). Nondetects were substituted with the detection limits normalized to the initial strength of the sources and represented as “empty” symbols.

events (after 4 and 8 weeks) concentrations differed on average only by a factor of 3.1 (calculated when replacing nondetects with half LODs). Accordingly, in apartments 2 and 4, concentrations in dust on two surfaces in the rooms varied by factors of 2.9 and 2.2, respectively.

In apartment 3, floor dust samples were also collected from three small, defined areas on the floor using the same sampling technique as for lifted dust (see SI). Concentrations of all target substances in floor dust samples collected from these three sampling locations during both sampling events varied on average by a factor of 3.6. Neither for the floor nor for the lifted dust was a clear relationship observed between substance concentration and distance from the source.

The comparison of averaged concentrations in lifted dust and floor dust (filter collection) for each substance at both time points shows that floor and lifted dust in the room differ by factors of 4.1, 2.6, and 9.1 in apartments 2, 3, and 4, respectively.

After collecting dust on filters, the same sampling surfaces were additionally wiped to collect any remaining amount of dust. The concentrations of chemicals in the wipe samples were comparable to the concentrations in dust collected on the filters (see SI).

Mass Balance. The mass balance was calculated to identify the main sinks of the substances present in plastic products and the spray (for the carpet substances DBA and DOP the mass balance could not be calculated due to highly uncertain amounts released during the field studies). It was estimated that during both field studies only up to 3% (DOA in the first field study) of the released amount was detected in the indoor air and settled dust (see SI). Hence, other sinks have to account for the missing mass. It was determined that up to 15% and 8% of the released amounts of DEHP could be sorbed to textiles, including clothes of the inhabitants during the first and the second field study, respectively. Two people in the room could take up less than 1% by dust ingestion and air inhalation. In contrast to these smaller sinks, foam in soft furniture may

represent a much larger sink: the amounts of DEHP that could theoretically be accumulated by furniture could account for more than 80% and 220% of the amounts released from the products during the first and the second field studies, respectively (see SI).

5. DISCUSSION

Concentration Patterns in Dust. The concentrations of DiBP and DEHA in dust measured in our study are in agreement with values published in literature,^{5,8} indicating that these substances may not be abundantly used in consumer products (our products contained much less plasticizer than used in consumer products).^{37,38} However, dust concentrations of DEHP measured in other studies are 2 orders of magnitude higher since the amount of DEHP released indoors may be orders of magnitude higher in comparison to the amount of deuterium-labeled DEHP that we have used in our study.

Our finding that the more volatile substances (DiBA, DBA, DiBP, DBP) were seldom detected in lifted dust samples, while the less volatiles were detected in most of the dust samples is in agreement with other studies^{8,22} indicating that more volatile substances (with lower K_{oa}) are predominantly found in air (see SI). However, more volatile substances were also detected in a substantial part of the floor dust samples (concentrations in floor dust during the first field study were reported previously³³). An explanation could be that the vacuum cleaner acted as an active air sampler.³³ This would mean that the floor dust concentrations of more volatile substances during the first field study (data too scarce in the second field study) may be overestimated by up to 10% (assuming a sampling duration of 10 min and maximal vacuum cleaner airflow of 2.8 m^3/min). Lifted dust was also collected using the nozzle attached to the vacuum cleaner, however, the sampling duration was substantially shorter.

Despite its relatively high volatility, DBP was not detected in air (see SI). Assuming DBP is transferred to dust via air, this would explain why it was not transferred to the lifted dust or to

the floor dust (vacuumed). DOP was detected sporadically in air samples and only during the first field study (see SI), however, it was detected in all floor dust samples. Since the same carpets were used for both field studies, the nondetection of DOP in air during the second field study may indicate a weakening of the source. However, the fact that DOP was detected in all floor dust samples during both field studies may be explained by DOP being distributed in the room while attached to dust, for example, by inhabitants stepping on the carpet and then moving around in the room. DBP was possibly also transferred in the same manner, however, with its concentration below the limit of detection. Since the initial amount of DOP installed in the room was 10 times higher than the amount of DBP, the measured concentration of DBP in dust (before normalization) was much lower than the DOP concentration (see SI).

The concentrations of DBP and DOP in dust deposited on the carpet were a few orders of magnitude higher in comparison to those of substances from the plastic products. Such elevated concentrations are mainly caused by the direct transfer process from the carpet to dust in contact with the source.³³ However, the specific transfer process (e.g., abrasion or direct contact) could not be identified.

Even when the surface area of the horizontal source is small (like the 0.5 m² carpet in this study) in comparison to the total floor area, it may contribute considerably to total concentrations in dust due to direct transfer. Therefore, it is important for studies on substance concentrations in dust to report whether the surface of the source was included or not in the sampling.

Dust in Contact with the Source. We have observed that dust accumulating directly on the surface of the horizontally installed products (see Figure 1c) is enriched by up to 4 orders of magnitude in substances that are present in these products (DiBP and/or DOA). Substances in vertically installed products (e.g., DEHA, DEHP) were transferred to dust accumulating on the horizontal products via air due to evaporation, while DOA was in contact with the dust deposited on the surface of the source and was most likely transferred directly. This indicates that direct transfer is a leading and efficient pathway for substance transfer from consumer products to dust accumulating directly on the surface of the source. Our results correspond with the findings of a small chamber experiment analyzing house dust with and without direct contact to a plasticized PVC polymer.¹⁹ The results of the chamber experiment showed an elevated mass transfer (dependent on the initial concentration in the source) of DEHP in case of direct contact between dust and the emission source. More recently, test chamber studies demonstrated a rapid and substantial direct transfer of PBDEs from a TV casing sample²⁴ and HBCD-treated fabrics²⁰ to dust.

In our study, we have observed that more volatile compounds are also directly transferred to dust. As discussed previously,^{8,22,33} substances with lower K_{oa} values (i.e., more volatile) tend to be emitted into air but are seldom detected in dust. However, DiBP from the horizontally installed plastic product (PH) was detected in high concentrations in all dust samples collected from the surface of the PH. This finding indicates that direct transfer is an important transfer pathway for all SVOCs regardless of their vapor pressure. Other studies support this: The results of the chamber experiment³ showed that PCB migration into dust is much faster (and, e.g., two orders of magnitude higher for congener 118) when house dust

is in direct contact with a source in comparison to only dust/air partitioning. Moreover, dust/source partitioning was not significantly affected by the volatility of the congener.³ Similar results were observed in a test house where the concentration of BBzP in dust deposited on the floor (source material) was about 20–30 times higher in comparison to nonfloor dust, while little difference was observed for DEHP (not present in the flooring).²⁵

Although the transfer of substances from the products to dust in contact with the product is very efficient, the subsequent substance transfer into the dust on other surfaces (due to evaporation from dust or transfer of dust particles itself) is very limited. Therefore, the importance of this direct transfer in determining dust concentrations increases with the size of the horizontal product.

The sampling procedure of dust on the product may have caused abrasion of the plastic surface or transfer of the carpet fibers, which may have increased the substance concentration in dust. This hypothesis is also supported by other authors.^{18,20} However, due to the gentle handling of the source material (and the absence of any visible fibers in the sampled dust) it is not likely that abrasion accounts for the large concentration difference observed.²⁰ In addition, the surface area of the nozzle touching the surface of the product during sampling was very small (see SI), which reduces the possible impact of abrasion. Moreover, during the first field study, the concentrations of DiBP and DOA in dust on the products were higher than the initial concentrations in the products (see SI), which supports the finding that the direct contact pathway is highly efficient and that direct transfer may need to be considered for calculating concentrations in dust.

Dust Homogeneity Indoors. It is expected that substance distribution indoors differs between countries,^{28,39–41} regions,⁴⁰ and seasons⁴² as substance transfer into dust depends on the organic matter fraction of the dust²³ and the air exchange rate.⁴³ Since all the apartments investigated in our study are located in the same region and samples were collected during the same season of the year, the substance distribution in the investigated apartments was comparable and varied by less than a factor of 20 during both field studies³³ (see SI). Some variation within and between the rooms, however, can be expected: Sampling surfaces in our study were relatively small (0.1–0.7 m²) and the air circulation in a room may differ at different heights or locations (e.g., corners) in the room.

During the second field study, we have also observed that the substance concentration in lifted dust varied only by a factor of 2–3 and for floor dust by a factor of 3.6 (in apartment 3). Most field studies have collected dust samples from a large (or even the whole) surface area of the floor. Due to the limited variation we observed, this surface area may be representative for the room, but only if the floor is not the source of the investigated substance.

The fact that two different sampling techniques (filter collection and wiping) yielded comparable substance concentrations in dust indicates that both sampling methods are suitable for dust sample collection.⁴⁴ The choice of the sampling technique is governed by the experimental conditions: Since the filters are relatively small, only small amounts of dust can be collected. Wipes on the other hand are more suitable for collecting larger samples. However, the analysis of wipes is more complicated due to interference from chemicals present in the wipes and requires more preparation before sampling.

Mass Balance. The fact that only up to 3% of the applied substances could be measured in indoor air and settled dust shows the importance of various sorptive surfaces as sinks for SVOCs. Based on the case of DEHP, it was estimated that the foam of soft furniture represents the strongest sink in the room (be reminded that polyurethane foam (PUF) is used as sorptive material for passive air sampling), much stronger than textiles or other sorptive surfaces. For textiles, this results from our extrapolation of partition coefficients: the textile-air partition coefficient for DEHP is 1 order of magnitude lower in comparison to the foam-air partition coefficient. For sorptive surfaces, the sorbed amount of DEHP is significantly lower than for PUF because sorptive surfaces are normally imagined as organic films. Therefore, their thickness is generally assumed to be only 10 nm, while for textiles and foam we derived a value of 1 mm on the basis of the study duration (see SI). These estimates, however, are very rough due to the uncertainty in diffusion coefficients.

Another explanation for the low substance recovery could be the degradation of target substances indoors. However, the photochemical degradation rates due to reaction with hydroxyl radicals (estimated by EPI Suite Version 4, see SI) are smaller than the air exchange rates (0.10–0.49 1/h)³³ indicating that degradation accounts for much smaller losses in comparison to the room ventilation. Only for tetramethrin (degradation rate 0.69 1/h) did degradation account for similar losses in comparison to ventilation. It is also unlikely that the deuterium (used for labeling of the target plasticizers) was replaced by a hydrogen atom and therefore disabled detection by MS. Deuterium labeling in the ring, which was used for the phthalates, has been previously reported as stable.^{45,46} Since the same trend of accumulation in different indoor media was observed for adipates and phthalates, it is assumed that the deuterium labeling for adipates was also stable under room conditions.

The evaluation of the data from both field studies confirms two important processes for the substance transfer from products to dust: transfer via the gas phase and direct transfer. Our study indicates that in cases with no direct contact between dust and the source, SVOCs distribute homogeneously throughout the entire room. However, when dust deposits on the source (horizontal products in this study) direct transfer occurs that significantly increases the concentration of the contained SVOCs in the deposited dust. Human exposure assessments should therefore consider whether a larger fraction of dust is in direct contact with the source of the investigated substance, since this may lead to increased human exposure. Depending on size and location, horizontal surfaces might contribute differently to dust concentrations than vertical surfaces. Therefore, in our future work we will investigate for which consumer products (e.g., size, location) direct transfer may be relevant. Future observational measurement studies may also want to consider the dust sampling location more carefully by noting whether dust is or is not in direct contact with the source. Future research could also focus on the quantitative investigation of the abrasion during sampling and how this contributes to the substance transfer from the product to indoor dust. Estimations of the diffusion coefficients in various media, for example, PUF and textiles are additionally needed for better estimations of the mass balance.

■ ASSOCIATED CONTENT

📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: [10.1021/acs.est.6b06051](https://doi.org/10.1021/acs.est.6b06051).

Sampling procedure, sample preparation and quality control descriptions. Variation between apartments. Measured concentrations in air, floor dust, lifted dust, wipes, dust on the product, comparison of concentrations, and mass balance estimations (PDF)

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Notes

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