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Inadvertently Generated PCBs in Consumer Products: Concentrations, Fate and Transport, and Preliminary Exposure Assessment

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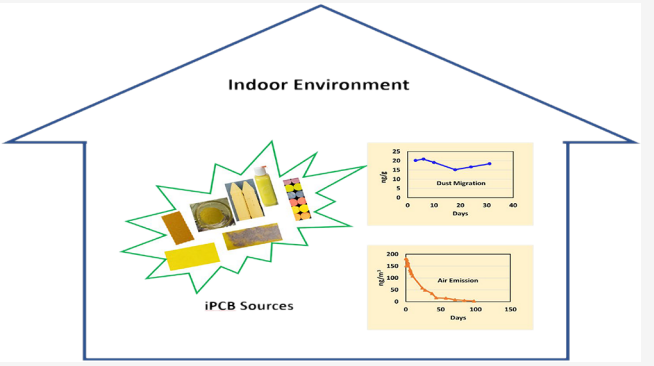
ABSTRACT: Although commercial polychlorinated biphenyl (PCB) production was banned in 1979 under the Toxics Substance Control Act, inadvertent generation of PCBs through a variety of chemical production processes continues to contaminate products and waste streams. In this research, a total of 39 consumer products purchased from local and online retailer stores were analyzed for 209 PCB congeners. Inadvertent PCBs (iPCBs) were detected from seven products, and PCB-11 was the only congener detected in most of the samples, with a maximum concentration exceeding 800 ng/g. Emission of PCB-11 to air was studied from one craft foam sheet product using dynamic microchambers at 40 °C for about 120 days. PCB-11 migration from the product to house dust was also investigated. The IAQX program was then employed to estimate the emissions of PCB-11 from 10 craft foam sheets to indoor air in a 30 m^3 room at 0.5 h^{-1} air change rate for 30 days. The predicted maximum PCB-11 concentration in the room air (156.8 ng/m^3) and the measured concentration in dust (20 ng/g) were applied for the preliminary exposure assessment. The generated data from multipathway investigation in this work should be informative for further risk assessment and management for iPCBs.



KEYWORDS: *inadvertent PCBs, emission, dust migration, source characterization, exposure assessment, PCB-11, indoor air*

INTRODUCTION

Polychlorinated biphenyls (PCBs) are a class of 209 organic compounds, known as congeners, as shown in Figure S1 in the Supporting Information (SI). They are semi volatile chemicals (SVOCs) that are persistent, bioaccumulative, and toxic.[1,2] Although commercial PCB production was banned in 1979 under the United States (US) Toxics Substance Control Act (TSCA), EPA's regulations implementing TSCA for PCBs allow some inadvertent generation of PCBs to occur in excluded manufacturing processes, as defined in title 40 of the Code of Federal Regulations (CFR) section 761.3. Specifically, the PCB regulations allow inadvertently generated PCBs (iPCBs) at defined concentrations, under certain conditions, and with requirements to report to EPA and maintain certain records.[2]

Production of iPCBs may occur during a variety of chemical manufacturing processes that involve carbon, chlorine, and high heat. They have been reported in pigments and dyes used in paints, inks, textiles, paper, cosmetics, leather, and in silicone as well as in vinyl chloride and TiO_2 (titanium dioxide) nanoparticles.[3−7] The most studied process leading to inadvertent PCB (iPCBs) contamination is the production of the diarylide yellow pigment, with the resulting generation of PCB-11.[8−16] Other iPCBs found abundant in pigments and consumer products include, but are not limited to, PCB-5, PCB-8, PCB-12, PCB-13, PCB-15, PCB-28, PCB-35, PCB-36, PCB-40, PCB-52, PCB-56, PCB-77, PCB-206, PCB-207, PCB-208, and PCB-209,[7] which are also detected in legacy PCB sources in the environment.[1,5,17−19] Recent studies by Washington State's Department of Ecology have identified a broad distribution of iPCBs in over 200 pigmented consumer products at concentrations up to parts per million.[16] These iPCBs contaminate not only products but also waste streams, air, water, sediment, and biota.[3,5−7,16,20−23] The relative toxicities of common inadvertent iPCBs congeners are not clear.[24−29] In 2014, EPA nominated 3,3′-dichlorobiphenyl (PCB-11) for evaluation by the National Toxicology Program

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(NTP) based in part on its ubiquity in the environment.[30] As an example, the Spokane River Regional Toxics Task Force (SRRTTF), which leads efforts to reduce toxic compounds in the Spokane River in the US, has reported the presence of iPCBs in some discharges to the river, which is also impaired by legacy PCBs.[31] Water quality standards for PCBs in the Spokane River are extremely low due to fish consumption rates of tribal members. Sources of pollution exceeding the water quality criteria in this tribal lifeway must be reduced as part of meeting treaty obligations ensuring the rights of tribal members to consume the fish from this river.

Inadvertent PCBs are present in human body fluids[5−7,31,32] due to exposure via ingestion, inhalation, dermal contact, and dietary intake.[32,33] While food consumption has been well-known as the most significant exposure route for the general population,[1,17,27,33,34] there is increasing evidence that inhalation of indoor air and ingestion of indoor dust via hand-to-mouth or hand-to-object-to-mouth pathways also contribute to human exposure, especially for children.[7,17,33−35] Nevertheless, few reported concentrations of iPCBs in indoor air and dust are available in the literature,[7] and the role of iPCB-contaminated dust in human exposure has not yet been well characterized.

Existing research tends to focus on either end of the source-to-body burden spectrum, i.e., evaluating iPCB concentrations either from the source such as consumer products and environmental media, or in human biomonitoring samples. An understanding of the fate and transport and exposure pathways is the missing link between these two areas of research. This leads any discussion regarding the fate and transport, exposure, and human health risks to be subject to assumptions and conjecture. To address this knowledge gap, EPA has been working to collect data to quantify the transport of iPCBs from consumer products to the environment. This research generates the first data on migration pathways of iPCBs from consumer products into the environment and potential routes of human exposure. The efforts include (1) identification of iPCBs from 39 consumer products purchased on the current retail market, (2) selection of PCB-11 as the major conger to be studied for fate and transport and exposure assessment, (3) measurement of PCB-11 emissions from consumer products, (4) investigation of PCB-11 migration from the source to settled dust, and (5) preliminary assessment of potential exposure to PCB-11. The generated data enhances our ability to predict iPCB exposure. These results also assist the regional efforts of the SRRTTF and state and local partners who are trying to find upstream solutions to iPCB contamination.

MATERIALS AND METHOD

Chemicals. A complete set of five congener mixtures in isooctane (2.5−7.5 μg/mL) as calibration standards for the determination of all 209 PCB congeners on the gas chromatography/mass spectrometry (GC/MS) system was purchased from AccuStandard, Inc. (New Haven, CT, USA). Individual isotopically labeled compounds in nonane solution (50 μg/mL) were purchased from Wellington Laboratories Inc. (Guelph, Ontario, Canada). ^{13}C-PCB-9, ^{13}C-PCB-52, and ^{13}C-PCB-202 were used as internal standards (IS) for GC/MS analysis, and ^{13}C-PCB-4, ^{13}C-PCB-77, and ^{13}C-PCB-206 were used as extraction recovery check standards (RCS) for the product, dust, and polyurethane foam (PUF) cartridge (pre-cleaned, certified, Supelco, St. Louis, MO, USA) extractions. Initially, 1,4-dichlorobenzene-d_4 (98% purity, Sigma-Aldrich, Inc. St. Louis, MO, USA) was used as an extraction RCS, but it showed low recovery during PUF cartridge sample extraction due to its relatively high volatility and was replaced by ^{13}C-PCB-4. Hexane (ultragrade or equivalent, Fisher, Pittsburgh, PA, USA) and methylene chloride ($MeCl_2$, HPLC grade, >99.9%, Honeywell Burdick & Jackson, Muskegon, MI, USA) were used as extraction solvents. Individual congener standards PCB-11, PCB-52, PCB-28, PCB-101, PCB-47, and PCB-206, 100 μg/mL each in hexane, were purchased from Chem Service, Inc. (West Chester, PA, USA) and used as Internal Audit Program (IAP) standards to evaluate the GC/MS performance in terms of accuracy and precision. Sodium sulfate added in the extraction process was anhydrous grade or equivalent from Fisher, Pittsburgh, PA, USA.

Determination of iPCBs in Consumer Products. The products tested are summarized in Table S1 of the Supporting Information (SI). Thirty-seven of the 39 consumer products were purchased from local or online retailers in the US between 2018 and 2019. The products were selected based on the literature information that iPCBs could be generated during the production of the diarylide yellow pigment and previously reported data, which shows PCB-11 was detected in 135 (62%) out of the 216 samples.[16] The other two products (CP-15 & CP-16) were previously tested and donated by the Washington State Department of Ecology in March 2019. Products included sidewalk chalk paint, modeling dough, crayons, sidewalk chalks, glue sticks, foam sheets, food bags and boxes, finger paint, tempera paint, sunscreens, lotions, dyes, markers, bath tablets and bubbles, construction paper, and pencils. With a few exceptions, items tested were yellow in color (Table S1). For pencils, the paint was scrapped off and only paint chips were tested.

To determine the PCB content in the products, duplicate product samples, weighing approximately 0.5 g each, were prepared and extracted using a sonicator (Ultrasonic Cleaner FS30, Fisher Scientific, USA) with 10 mL of hexane or $MeCl_2$ for 30 min in a 20 mL scintillation vial. For liquid product samples, approximately 100 mg of anhydrous sodium sulfate was added to dry potential water in those samples. Before extraction, 200 μL of each 5 ng/μL RCS was added into the extraction solution. After extraction, 990 μL of the extract was placed in a 1 mL volumetric flask containing 10 μL of each 10 ng/μL IS and then transferred to GC vials for GC/MS analysis. The final concentrations of each RCS and each IS were 50 and 100 ng/mL, respectively.

In addition, products CP-15 and CP-16 were extracted using hexane sonication, $MeCl_2$ sonication, and $MeCl_2$ Soxhlet extraction (EPA Method 1668C)[36] methods to compare extraction solvents and methods. Product variability was investigated using CP-17, a yellow glitter foam sheet. We purchased 10 packages of CP-17 from an online store. Each package contained three yellow sheets along with other color sheets. The packages and yellow sheets were randomly picked for testing both within the same package and between different packages.

Emission of iPCBs in Consumer Products. The yellow glitter foam sheet (CP-17) was selected for duplicate emission tests based on the concentration of iPCBs presented in the material (Figure 1). A 64 mm diameter circle was cut from a sheet of foam and placed in each 114 mL microchamber (M-CTE250, Markes International, Inc., USA). The tests were conducted at 40 °C with 28% relative humidity (RH) and at

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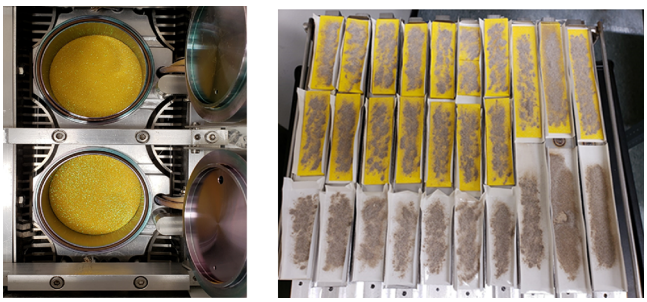

Figure 1. Product yellow glitter foam sheet emission tests in microchambers (left) and the dust migration test in a small chamber (right).

approximately 100 mL/min (54 h^{-1} air change rate) for 120 days. The two microchamber tests (T1 & T2) conformed to American Society of Testing and Materials (ASTM) D7706.[37] Concentrations of iPCBs in the source material were determined prior to chamber tests. PUF cartridge samples were collected from the microchamber with sampling duration up to 16 h each at 102 mL/min.

Migration of iPCBs from Consumer Products to Dust. The migration of iPCBs from products to settled dust test was conducted in a 53 L stainless steel chamber consistent with the ASTM D5116.[38] More details of the dust chamber test settings and operations are described in the literature.[39] The dust used for the migration test was house dust obtained from EPA's previous projects (Table S2). It was thermally conditioned, solvent-extracted, and analyzed by GC/MS to verify that there were no quantifiable PCB concentrations in the dust before use. The small chamber was operated at 23 °C, 1 h^{-1} air change rate, and 48% RH for 32 days. The foam sheets had one side with the glitter affixed (front) and one side without glitter (back). Each side was separately tested to determine if the adhesive applied to adhere the glitter would act as a barrier to minimize the migration of iPCBs to the house dust applied to the foam. The test material for the PCB-free substrate was Gardco RP-1 K release paper (Paul N. Gardner Company, Pompano Beach, FL, USA).

In the test, 0.15 g of dust was loaded onto front and back sides of the foam, so that two strips comprised one test set. 0.15 g of dust was also loaded onto 7 release paper strips (2.86 cm by 10.16 cm) (Figure 1). Additionally, three strips of the front side foam were loaded with multiple amounts of dust (0.2, 0.3, 0.4 g) to investigate the effect of dust loading on migration from the source to dust. Dust samples were collected from the strips at six intervals with one duplicate. After collection, the dust was solvent extracted. The iPCB content in the dust was determined on a weight per weight basis (e.g., ng iPCB/g dust). PCB air concentrations inside the test chamber were monitored prior to and throughout the migration test by collecting samples using PUF cartridges followed by solvent extraction.

Analytical Methods. The foam and release paper samples removed from the test chambers were extracted using the same extraction method used for the consumer products described above. The dust and PUF cartridge samples were extracted slightly differently and described in the SI. The analytical method used for this project was a modification of EPA Method 1668C.[36] The analytical instrument used for quantitative analysis of PCB congeners was an Agilent 6890/5973 GC/MS with a 7683 Auto Sampler. It was calibrated with 209 PCB congeners in the range of 2.5 to 300 ng/mL at five concentration levels in triplicate injections. The operational conditions of the GC/MS are provided in Table S3.

Quality Assurance and Quality Control. Solvent blank, extraction method blank, and field blank samples were prepared and analyzed. Recovery check standards were spiked in each of the samples prior to extraction. Acceptance criteria for the extraction and analysis were that the RCS had to be within 100 ± 25% recovery and the relative percent difference of the duplicate samples be within ±25%. The data presented were not adjusted for recovery of RCS. The data that did not meet the criteria were either not reported or reported with a note.

RESULTS AND DISCUSSION

Extraction Protocols. We compared solvent extraction methods using Soxhlet and sonication with $MeCl_2$ and hexane. The results are presented in Table S4. It was found that the results from the three extraction methods (hexane sonication, $MeCl_2$ sonication, and $MeCl_2$ Soxhlet) are comparable, with $MeCl_2$ Soxhlet extraction resulting in larger concentration variations in most cases. Given that the Soxhlet extraction method used much more $MeCl_2$ (150−300 mL depending on the size of the Soxhlet and boiling flask) and took much longer for extraction and sample preparation (about 38 h) than the sonication methods and that the sonication methods met our data quality goals, we concluded that the $MeCl_2$ sonication method for consumer product extractions was the most efficient practice for this project.

Table 1. iPCB Average Concentrations in Consumer Products Tested

	concentration (ng/g) ± %RSD (n = 2)							
product IDs	PCB-11	PCB-95	PCB-121	PCB-85	PCB-181	PCB-149	PCB-153	PCB-138
crayon-yellow	71.5 ± 1.08	ND[a]	ND	ND	ND	ND	ND	ND
crayon-green	43.3 ± 1.34[b]	ND	ND	ND	ND	ND	ND	ND
sidewalk chalk	167.5 ± 9.71	ND	ND	ND	ND	ND	ND	ND
foam sheet	122.1 ± 0.67	ND	ND	ND	ND	ND	ND	ND
fiberboard box (wafers)	ND	66.7 ± 4.46[b]	101.1 ± 5.08	137.2 ± 3.78	85.3 ± 4.53[b]	63.9 ± 8.69[b]	67.5 ± 2.89[b]	122.3 ± 4.03
glitter foam sheet[c]	345.7 ± 2.61	ND	ND	ND	ND	ND	ND	ND
sidewalk chalk Paint	18.5 ± 8.39[b]	ND	ND	ND	ND	ND	ND	ND
Glitter Foam Sheet[d]	696.7 ± 23.39	ND	ND	ND	ND	ND	ND	ND

[a]Not detected. [b]Concentration above the instrument detection limit but below the lowest calibration concentration. [c]From Washington State. [d]Purchased online.

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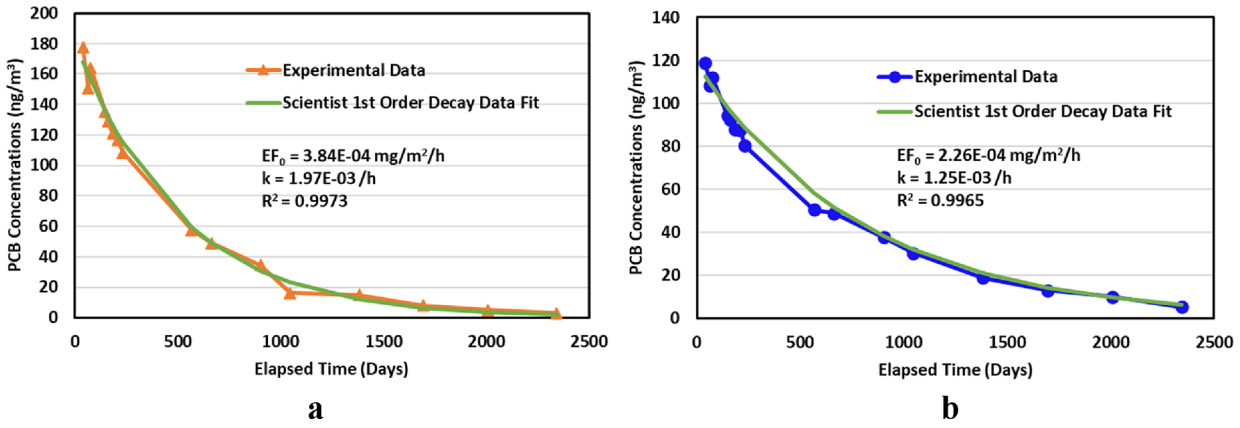

Figure 2. Least square fit of first order decay emission data from microchamber emission test T1 (a) and test T2 (b).

iPCB Concentrations in Consumer Products. We detected PCBs from seven out of 39 products, with concentrations ranging from below the lowest calibration concentration to 697 ng/g (Table 1). Six out of the seven products contained PCB-11 only. In one of the seven products, fiberboard packaging for wafer cookies, PCB-95, PCB-121, PCB-85, PCB118, PCB-149, PCB-153, and PCB-138 (Table 1) were detected with a concentration range from below the lowest calibration concentration (Table S5) to 137 ng/g, but PCB-11 was not detected. Our results are slightly lower than the iPCB concentrations in over 200 pigmented consumer products reported by Washington State's Department of Ecology.[16]

Additionally, a total of 13 yellow glitter foam sheets purchased online (CP-17) in the same package (three sheets in each of three packages) or in different packages (second sheet in each of the other four packages) were extracted for the product variability investigation. The results are shown in Figure S2. The average PCB-11 concentration of all sheets was 662 ng/g with the % relative standard deviation (%RSD) being ±17.6%.

iPCB Emissions from Products. Even though yellow glitter foam sheets (CP-17) were used for the emission tests because of its highest iPCB (PCB-11) concentration detected among all the tested products, the average concentration was less than 700 ng/g. Taking into account that PCBs are SVOCs, we decided to conduct the microchamber emission tests at 40 °C, which also simulates some possible classroom conditions, such as lack of air conditioning during the summertime, storage on shelving in front of windows, or storage on heaters during the wintertime. The results are presented in Figure 2. The concentrations of PCB-11 peaked within the first 2 days to about 120−180 ng/m^3 for the duplicate tests and then started to decay. After ∼20 days, the decay rate slowed down. At 120 days, PCB-11 reached concentrations lower than our practical quantification limit (lowest calibration). The total mass of PCB-11 in one foam sheet was calculated based on the foam sheet extraction data (5.7 μg/sheet from T1 and 5.6 μg/sheet from T2). We also calculated the total mass using the microchamber emission data by eq 21 in ASTM D5116 with the value being 5.3 μg/sheet from T1 and 4.9 μg/sheet from T2. The difference between these two calculations was less than 14%. Furthermore, the emission data collected was fitted by the least square method to the first-order decay model using the SCIENTIST program (Micromath Scientific Software) to obtain the initial emission factor (EF_0) and the decay rate constant (k) based on ASTM D5116. The data from the model fitting and experimental measurement are shown in Figure 2. The IAQX program (Simulation Tool Kit for Indoor Air Quality and Inhalation Exposure) developed by EPA[40] was then used to estimate the gas phase PCB-11 concentrations in a 30 m^3 room at a 0.5 h^{-1} air change rate with 10 foam sheets in the room for 30 days. The model parameters are listed in Table S6, and the comparison of simulation results by different EF_0 and k values is provided in Figure 3. The model predicts

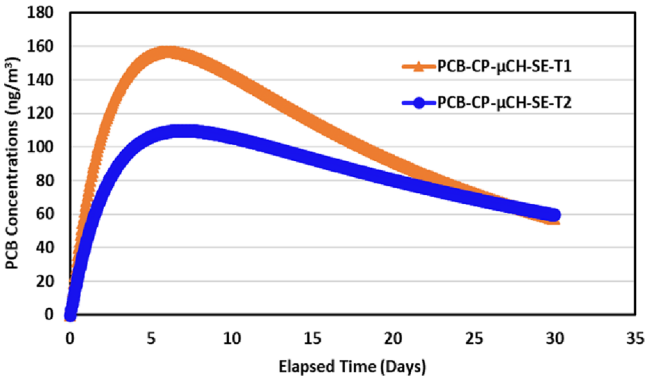

Figure 3. Gas-phase PCB-11 concentrations predicted in a 30 m^3 room with 10 foam sheets, 0.5 h^{-1} air change rate for over 30 days.

that after 30 days, the PCB-11 concentration in the room air is approximately 60 ng/m^3, which is much higher than its reported concentrations ranging from 0.018 to 1.819 ng/m^3 in indoor air in the literature.[7] One possible reason is that in the IAQ model simulation, the emission factor and first-order decay rate constant were obtained at 40 °C from a coated stainless-steel chamber. In addition, our simulation scenario did not take into account any sink effects, such as sorption of PCB-11 on airborne particles, dust, floors, carpets, and other indoor surfaces. However, the emission data confirm that the presence of iPCBs in consumer products can be an open PCB source to indoor air. Given that PCB-11 is relatively volatile among congeners, its existence in the indoor air cannot be ignored.

iPCB Migration from Source to Dust. The house dust used for the test was demonstrated to be free of PCBs at the beginning of this research. Under the test conditions, there were three mass transfer processes taking place, namely, emissions of PCB-11 from the foam product covered with

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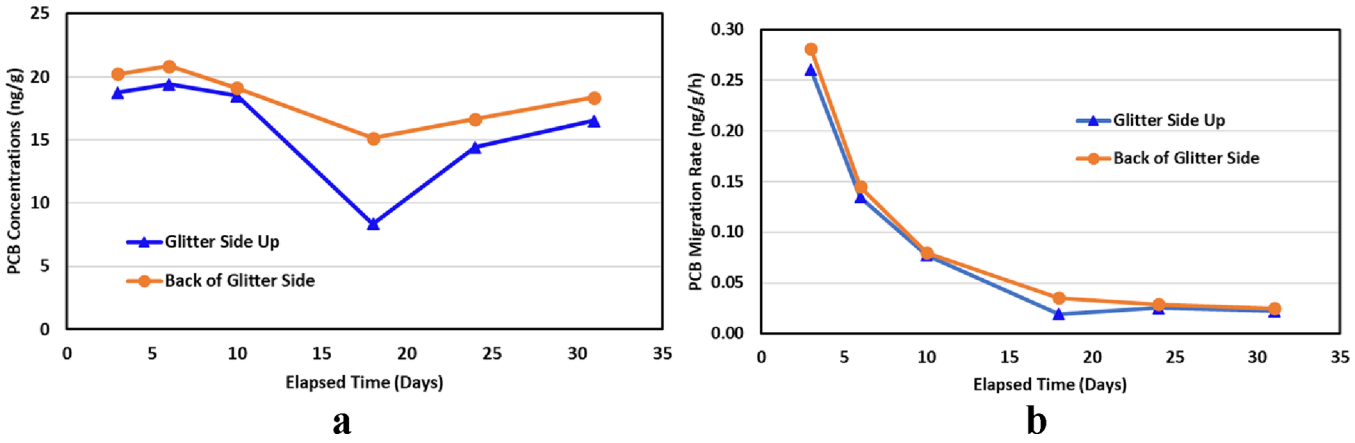

Figure 4. Migration concentration of PCB-11 from the CP-17 foam sheet to the house dust (a) and the migration rate (b).

"clean" dust, PCB-11 migration from the foam source to dust via direct contact, and sorption of PCB-11 from air into "clean" dust on the PCB-free release paper. The results are shown in Figure S3. Foam pieces were removed during sampling leading to the gradual decrease of the air emission concentrations. PCB-11 was not detectable in the dust on PCB-free release paper until in the last sample collected at 774 h, in which the concentration was above the instrument detection limit but below our lowest calibration concentration, indicating negligible sorption into dust from airborne PCB-11. The plausible reason is that PCB-11 concentrations emitted from the dust-covered foam sheet to the chamber air was low (<10 ng/m^3) and thus the absorption of PCB-11 from the air to "clean" dust was low as well.

The migration concentrations of PCB-11 from both the front and the back of the foam product pieces are about 15−20 ng/g as shown in Figure 4a. By comparing Figures 4a and S3, we observe that PCB-11 migration into the dust via dust-material partition is highly effective. The time-averaged migration rate (ng/g/h) was calculated by the migration concentration divided by the exposure time and presented in Figure 4b. It is illustrated that PCB-11 migrated from the foam sheet source at a much faster rate at the beginning to 17 days and then remained stable until the end of the test. Dust loading samples at four levels between 0.15 and 0.4 g per material strip were all taken out within 5 min at the end of the test. The results are presented in Figure S4. The migration concentrations increased with the amount of house dust, peaked at 0.3 g, and then decreased. Overall, the results from the dust migration test via direct contact demonstrate that dust plays an important role in the fate and transport of iPCBs both in creating a pathway for PCBs to migrate out of the product and in reducing the potential for PCBs to directly emit into the air.

The amount of iPCBs transferred from the source materials to the dust depends on dust-source partitioning, diffusivities, and distributions of iPCBs in dust and source materials. The dust-source partition coefficient (K_{dm}, dimensionless) at equilibrium can be calculated by the ratio of equilibrium concentrations in dust vs in the source. Since the dust migration test lasted for over 744 h, we made use of the last data point at the end of the test and estimated the dust-source partition coefficient (K_{dm}), using eq 1.

$$K_{dm} = C_d/C_m \quad (1)$$

where C_d is the PCB-11 concentration in dust in equilibrium with the source material, μg/g, and C_m is the PCB-11 concentration in the source material in equilibrium with dust, μg/g. The estimated value of K_{dm} is 0.021 for the front glitter side and 0.023 for the back side of the foam sheet.

Preliminary Exposure Estimation. Human exposure to iPCBs in consumer products is through air and dust inhalation, dust ingestion, and dermal contact via air absorption, dust contact, and direct contact with the product in the indoor environment. The daily human intake levels from exposure to PCB-11 was estimated based on the emission and dust migration measurements from CP-17 using standard exposure factors available from EPA's Exposure Factors Handbook (EFH)[41] and standard route-specific algorithms for (1) inhalation of air, (2) dermal absorption from air, (3) ingestion of dust that absorbed PCBs from the air, (4) ingestion of dust contaminated by migration via direct contact with the source material, and (5) dermal absorption of contaminated dust. The contaminated dust herein is defined as dust on the surface of glitter foam sheets into which PCBs migrate directly through contact. Three age groups were assessed separately: children 3 to <6 years, children 6 to <11 years, and adults (16 to <21 years) when data is available.

The route specific algorithm for the air inhalation intakes (Intake$_{inh}$, μg/kg/day) is as follows:

$$\text{Intake}_{inh} = (\text{Conc}_{air} \times \text{InhRate} \times \text{ET} \times \text{ABS})/(\text{BW}) \quad (2)$$

Specifically, the maximum IAQX model prediction of PCB-11 concentration in a 30 m^3 room at 0.5 h^{-1} air changes per hour with 10 sheets of glitter foam in Figure 3 (156.8 ng/m^3, equivalently 0.1568 μg/m^3) was used as the air concentration (Conc$_{air}$, μg/m^3). Pulmonary absorption from inhaled air (ABS, %) was assumed to be 70%. The inhalation rate (InhRate, m^3/day) and the body weight (BW, kg) for different receptors were adopted from EPA's EFH[39] and provided in Table S7. The exposure time of 8 h/day,[42] by default, was also used to represent the time spent in a school or childcare setting.

The route-specific algorithm for the dermal absorption from air (Intake$_{derm}$, μg/kg/day) is as follows:

$$\text{Intake}_{derm} = (\text{Conc}_{air} \times \text{TransDermPerm} \times \text{BSA} \times \text{ET})/(\text{BW}) \quad (3)$$

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The dermal absorption from air relied on transdermal permeability (0.13 m/h) and was calculated according to methods of eq 24 in the literature[43] for the indoor air transdermal permeability coefficient, and a default 6 m/h was used for the mass-transfer coefficient between bulk air and skin surface.[43] The molecular weight (MW, 223.1), air−water partition coefficient (log K_{aw}, −2.134), octanol−air partition coefficient (log K_{oa}, 7.419), and octanol−water partition coefficient (log K_{ow}, 5.285) were obtained from EPA's CompTox Chemistry Dashboard.[44] The exposure time was assumed to be 8 h/day. Age-appropriate body surface areas (BSA, m^2) were from EPA's EFH[41] (Table S7).

The route specific algorithm for the ingestion of dust that absorbed PCBs in the air ($Intake_{inj}$, μg/kg/day) is as follows:

$$Intake_{ing} = (Conc_{dust} \times IngRate \times ABS)/(BW) \quad (4)$$

In this equation, two values of dust concentration ($Conc_{dust}$, ng/g) were used for the calculation. One value (56 ng/g) was estimated from the air concentration ($C_{dust\text{-}air}$, μg/m^3) in the following relationship from Dodson et al.,[45] assuming all PCB in air to be in the gas phase:

$$Conc_{dust}\ (\mu g/g) = (C_{dust-air} \times fom_{dust} \times K_{oa})/\rho_{dust} \quad (5)$$

where

$C_{dust\text{-}air}$ is the measured air concentration during the dust migration test, 10 ng/m^3 (0.01 μg/m^3, Figure S3), fom_{dust} is the volume fraction of organic matter associated with settled dust, assumed to be 0.2, and ρ_{dust} is the density of dust (g/m^3), measured to be 9.38×10^5 g/m^3 (Table S2).

Verification of this relationship was assessed by the data reported by Andersen et al.[18] The other value of $Conc_{dust}$ used for eq 4 was the directly measured PCB-11 concentration in the dust that was on the release paper during the migration test. It was 4.5 ng/g, a concentration below our lowest calibration standard but above the instrument detection limit. The IngRate (mg/day) was from Table S7 of Mitro et al.,[46] and gastrointestinal absorption (ABS) was 75% based on the value from Schlummer et al.[47]

Ingestion of contaminated dust through hand-to-mouth transfer ($Intake_{dust\_ing}$, μg/kg/day) was calculated using the algorithm as follows:

$$Intake_{dust\_ing} = (Mass_{dust\_contam} \times SA \times HtoM_{freq} \times HtoM_{eff} \times Frac \times ET \times Conc_{dust\_contam} \times ABS)/(BW) \quad (6)$$

In eq 6, the mass of dust adhering to skin per unit area ($Mass_{dust\_contam}$, mg/cm^2), skin surface area contacting dust (SA, cm^2), hand-to-mouth contact frequency ($HtoM_{freq}$, 1/h), and BW were taken from EPA's EFH.[41] The hand-to-mouth transfer efficiency ($HtoM_{eff}$, 0.2) and the fraction of hand mouthed per contact event (Frac, 0.13 1/contact) were from the literature.[48] The $Conc_{dust\_contam}$ (ng/mg) is the maximum PCB-11 concentrations in the dust measured though our test. Assumptions include that surface adherence was attained instantaneously and was fully replenished between each mouthing event through contact with glitter foam sheets, gastrointestinal absorption (ABS) of 75%[47] and 1 h/day of exposure time.[48]

Dermal absorption of contaminated dust ($Intake_{dust\_derm}$, μg/kg/day) was estimated using the following algorithm:

$$Intake_{dust\_derm} = (Mass_{dust\_contam} \times Conc_{dust\_contam} \times SA \times ABS \times ET)/(BW) \quad (7)$$

In eq 7, in addition to the measured $Conc_{dust\_contam}$, the absorption fraction (ABS) for dermal absorption was assumed to be 6% in keeping with EPA Risk Assessment Guidelines for Superfund.[49] An estimated exposure duration of 1 h was used.

The calculated results are summarized in Table 2.

Table 2. Estimated Intake of PCB-11 (ng/kg/day)

	age Groups		
exposure pathways	children 3 to <6 years	children 6 to <11 years	adults 16 to <21 years
inhalation from air	19.9	13.8	8.3
dermal absorption from air	6.7	5.5	4.2
ingestion of dust absorbed gas PCB	0.1[a]/0.01[b]	0.04[a]/0.003[b]	0.02[a]/0.001[b]
ingestion of contaminated dust	0.006	0.002	NA[c]
dermal absorption of contaminated dust	1.1×10^{-7}	8.8×10^{-8}	NA[c]
total	26.7[a]/26.6[b]	19.3[a]/19.3[b]	12.5[a,d]/12.5[b,d]

[a]Calculated based on eq 5. [b]Calculated based on measured data. [c]Not calculated because of no data available for adults' $Mass_{dust\_contam}$. [d]Calculation did not include ingestion or dermal absorption of contaminated dust.

Historically, consumption of contaminated food has been the major route of PCB exposure among the general population.[1,17,27] Our results from Table 2 suggest that inhalation and dermal absorption from air are important intake pathways for the volatile iPCBs such as PCB-11. Although measured indoor air concentrations of iPCBs and experimental data on their potential exposure routes are scarce, we compared our results with that from the limited literature. Wang et al. estimated human intake of PCB-11 from inhalation, dust ingestion, and dermal contact with dust based on their paired indoor air and floor dust samples collected from 28 Australian residential houses and offices.[50] Their reported values of inhalation intake (26 and 31 pg/kg/day for adults and toddlers) are much lower than our results, but their data showed inhalation has the highest intake among the three exposure routes they studied. Conversely, our estimated dust ingestion intakes are much lower than the median 0.16 and 0.36 ng/kg/day, reported by Anh et al. for the workers and children, respectively, at the end-of-life vehicle processing sites in Vietnam,[50] which is anticipated when comparing a residential to an occupational scenario. In addition, Weitekamp et al.[17] estimated background PCB exposure through different routes and their relative contribution for selected age groups based on data collected from comprehensive literature review. Although they identified dietary intake as the major exposure pathway, our data are consistent with their conclusions that indoor air inhalation exposure tends to favor volatile, lower chlorinated congeners and that children may experience higher total PCB exposures. It is worth noting that our exposure estimates were based on emission testing of one consumer product that was found to have the highest concentration of PCB-11 and that had a large surface area (with respect to volume) in a room size of 30 m^3. Moreover, our simulation scenario did not include any sink

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effects and other potential iPCB sources. Including sink effects likely would greatly reduce the air concentration as SVOCs prefer to partition to surfaces. Conversely, our estimates were only based on one product releasing iPCBs into the room and only 10 sheets from that product. We might expect the increased exposure to iPCBs caused by cumulative impacts of emissions from multiple products in a single room.

Implication. Our research indicates that some iPCBs, e.g., PCB-11, in consumer products are present at a maximum concentration exceeding 800 ng/g. The multipathway exposure assessment is informative for discussions about potential migration from products into the environment. Whether the solution lies in preferred purchasing programs, green chemistry, effluent controls, regulatory changes, or elsewhere, understanding the fate, transport, and exposure pathways is a critical step in designing the ultimate solution. In addition, the data generated from this study will be valuable to contextualize the toxicity data for PCB-11 generated by the NTP, once it is released. This research will be foundational for additional future research to better understand the concentrations, fate, and transport of iPCBs in yellow pigmented consumer products and their cumulative risk assessment. Our next step is to conduct more product testing and generate more data for other migration pathways of iPCBs from consumer products into the environment and potential routes of human exposure. Further characterizing the variability and reducing uncertainties in the data will increase confidence in the estimations of iPCB exposures both indoors and via the environment.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c02517.

Additional details on the chemical structure of PCBs, experimental methods, calibration and instrument detection limits, and exposure estimation, and more research results in Table S4 and Figure S2 to S4 (PDF).

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Notes

The views expressed in this article are those of the authors and do not necessarily represent the views or policies of the U.S. EPA. Mention of trade names or commercial products does not constitute endorsement or recommendation for use by the U.S. EPA. The authors declare no competing financial interest. The authors declare no competing financial interest.

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