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Contamination pathways of polychlorinated biphenyls (PCBs) – From the worker to the family

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ABSTRACT

The improper recycling of old industrial sized capacitors and transformers in Dortmund, Germany, led to a contamination of workers and environment with polychlorinated biphenyls (PCBs). In 2010, a surveillance program was conducted for the former workers, their relatives, and residents next to the recycling company. In the human biomonitoring, elevated values for PCBs were detected in a proportion of the in-house relatives of the workers, including children. The aim of this study was to reconstruct the contamination pathways from the company to the family.

Brought home working clothes of the former employees were identified as a first pathway for a contamination of the residential environment. Those clothes were cleaned at home, due to temporary employment contract. Washing machines and dryers were in consequence highly contaminated with PCBs. This led to a contamination of the relative's clothes and by this to a dermal uptake of PCBs. Another exposure pathway was the inhalation of PCB containing dust. In particular at locations, that were in contact with the working boots, high levels of PCBs were detected in the ambient monitoring. This established another exposure pathway by inhalation of PCB containing dust. The link between the residential influence factors and the PCB plasma concentration (n = 54) is shown by rank correlation.

In conclusion, it should be avoided to take home potentially contaminated working clothes or boots, to prevent an exposure of children and other family members.

1. Introduction

In 2010, local authorities discovered the inappropriate recycling of PCB-containing large-sized capacitors and transformers in a recycling company in Dortmund, Germany. This improper recycling led to a contamination of workers and environment with polychlorinated biphenyls (PCBs). PCBs belong to the group of persistent organic pollutants (POP), also known as the "dirty dozen". The production of PCB is forbidden since 2001, initiated by the Stockholm convention on persistent organic pollutants. PCBs can be divided into lower chlorinated (2–5 chlorine atoms), higher chlorinated (> 5 chlorine atoms) and dioxin-like PCBs (dl-PCB). Increased blood levels of lower chlorinated PCBs are typically a marker of occupational exposure, while higher chlorinated PCBs accumulate in the human body via nutrition (Safe, 1993). The accumulation of higher chlorinated PCBs in comparison to lower chlorinated PCBs in the food chain and human body is explainable by the long half-life time of higher chlorinated PCBs. Lower chlorinated PCBs have a significantly shorter half-life time and hence do not accumulate (Schettgen et al., 2012a; Wimmerova et al., 2011). The German Research Foundation (DFG) has defined biological reference values (BARs) for lower chlorinated PCBs (PCB 28: 0.02 µg/L plasma, PCB 52 and PCB 101: < 0.01 µg/L plasma) and a biological tolerance value for occupational exposure (BAT). The BARs have been evaluated based on the 95th percentile of the plasma levels of the general population. Therefore, an exceedance of the BAR is a clear indication of additional occupational or environmental exposure to these lower chlorinated congeners (Deutsche-Forschungsgemeinschaft, 2018). In contrast, the BAT-value (ΣPCB > 15 µg/l) is a health-based biological limit value and defined as the concentration of a substance, below which the health of a worker is generally not adversely affected by repeated or long-term exposure.

PCBs have several health effects. For example, they can lead to skin abnormalities (chloracne, hyperpigmentation (Akahane et al., 2018; Fischbein et al., 1982; Leijs et al., 2018)) or to abnormalities of the

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immune system (Haase et al., 2016). Newborn children from PCB contaminated mothers show an impaired growth as well as dysmorphic physical findings, described as "Yusho disease" (Guo et al., 1995). Furthermore, PCBs are suspected to cause cancer. The International Agency for Research on Cancer (IARC) observed a consistent association between PCB exposure and malignant melanoma (IARC, 2013; IARC, 2016). The IARC also identified studies, that indicated an association between PCB exposure and breast cancer and other cancer subtypes (IARC, 2013; IARC, 2016).

In Dortmund, Germany, a surveillance program (HELPcB cohort) was initiated in 2010, to assess possible health effects due to exposure towards high levels of PCBs (Kraus et al., 2012). Initially, n = 263 participants were included in this study. Participants were the former workers of the recycling company, their in-house family members as well as residents and workers from nearby facilities. During this study, all participants were interviewed, examined by a physician and were tested for laboratory abnormalities. Furthermore, ultrasound examination of the thyroid and the abdomen as well as neuropsychological and psychological testing were performed. An important pillar of this study was the biomonitoring of PCBs in the participants. Here, we found significantly evaluated values in former PCB-exposed workers, but also in some of their in-house family members including their children (Schettgen et al., 2012b).

The main goal of this analysis was to reconstruct the exposure pathways from the PCB exposed employees to their spouses, their parents, or their children. We used ambient monitoring techniques to detect the contamination route from the company to the children's room.

2. Material and methods

2.1. Participants, human and ambient monitoring

The participants of the HELPcB cohort included former workers of the recycling company, the in-house family members, employees of surrounding companies, and residents. The recycling site was closed in May 2010 and the study started in July 2010. After informed consent, all participants were interviewed and examined by an occupational health physician. General blood analyses and human biomonitoring were performed at the beginning of the study in 2010, as well as in the following years 2011, 2012, 2013, 2015, and 2017. In this study, initial human biomonitoring data from 2010 were selected. The analysis of the blood plasma samples to determine the concentration of the different PCB congeners was carried out by gas chromatography coupled with mass spectrometry (Schettgen et al., 2012a, 2012b). We analyzed the non-dioxin like indicator congeners (ndl) PCB 28, 52, 101, 138, 153, and 180 and the dioxin like congeners (dl) PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189. The lower detection limit for all congeners was 0.01 µg/L plasma. If the number of measurements above the detection limit was less than 10% of the sample count, these congeners were excluded from further analysis. Congeners excluded from the analyses were PCB 77, 81, 126 and 169.

The ambient monitoring was performed in the residential environment of those workers, who were highly exposed to PCBs according to their field of work and/or where the family members showed elevated blood levels of PCBs and who agreed in this analyses. Ambient monitoring included dust samples, wipe samples, and material samples from the houses, apartments, or vehicles of the selected participants and was carried out by Eurofins Umwelt Nord GmbH in 2010 and 2011. Material samples from washing machines and dryers were analyzed since the workers brought their working clothes home for cleaning purposes. Overall, the households of n = 18 families with n = 36 family members, give a written consent, were selected for this investigation. The analysis of the dust, wipe and material samples to determine the concentration of the different PCB congeners was carried out by gas chromatography coupled with mass spectrometry. The Laboratory of Eurofins Umwelt West GmbH analyzed the non-dioxin like indicator congeners (ndl) PCB 28, 52, 101, 138, 153, and 180. The lower quantification limits for all congeners in dust and material were between 0.05 mg/kg and 0.1 mg/kg depending on the sample quantities used for the analysis. For wipe samples the lower quantification limits for all congeners were between 0.005 $\mu g/m^2$ and 0.028 $\mu g/m^3$ depending on the size of the sampled area.

The surveillance program was approved by the ethics committee of the RWTH Aachen University (EK 176/11).

2.2. Statistical examinations/data management

In a first step, descriptive analyses of the ambient- and biomonitoring data were carried out.

For each household (n = 18), a dust sample and a wipe sample was collected. Indoor air samples were collected in n = 6 households; cloth samples were collected in n = 13 households and material samples, such as sealing and filter from washing machines or dryers were collected in n = 4 households. The cloth samples were washed and/or dried in the washing machine and/or dryer. PCB concentration was determined before and after the washing/drying process. In order to allow a clear evaluation, the sample with the highest measured value was selected for each area and used for the analyses. We used the highest measured value for the correlation analyses since a large proportion of the ambient monitoring data did not show a PCB contamination. The median PCB contamination could therefore underestimate the actual environmental contamination, we could observe in the hot spot areas (e.g. on the carpets). The ambient monitoring results were compared with the PCB plasma level samples of the participants (n = 54).

Relevant outliers in correlation analysis were identified by cook's distances ($cut\ of: \frac{4}{N} = 0.07$) and leverage values ($cut\ off: 3x\frac{(k+1)}{N} = 0.12$). Two participants were identified as significant outliers (PCB 28 > 100 mg/l plasma and PCB 52 > 5 µg/l plasma) and were excluded from the correlation analysis.

Data handling was carried out by using MS Excel, 2016 (Microsoft, 2016). Statistical examinations were carried out with SPSS 25 (IBM, 2017).

3. Results

Biomonitoring results from 2010 are shown in Table 1. The employees show the highest mean and median concentrations for PCB 28, 52, 101, 153, 138, 180, and 118 in comparison to their relatives including their children. For PCB 28, a maximum blood level concentration of 177.95 µg/l has been detected in one worker. The mean sum of indicator congener concentration was exceeding the BAT value, though (ΣPCB mean = 18 µg/L). Both, all relatives and the subgroup of children showed elevated values in comparison to a non-occupational PCB exposed cohort. The mean as well as the median for PCB 28, 52, and 101 exceed the BAR of 0.02 µg/L for PCB 28, as well as the BAR of 0.01 µg/L for PCB 52 and PCB 101. The results for the congeners PCB 81, 77, 123, 114, 105, 126, 167, 156, 157, 169, and 189 can be found in the supplement section. Under consideration of the congener pattern, relatives showed elevated values in particular for lower chlorinated biphenyls and less for higher chlorinated biphenyls. This finding is in contrast to the PCB congener pattern of the workers, where also higher chlorinated biphenyls were elevated.

The results for ambient monitoring are presented in Table 2. In all 18 households, dust and wipe samples have been collected. 59.5% of the dust samples exceeded the reference value of 1.65 mg PCB/kg dust and were therefore considered as PCB contaminated (Müssig-Zufika et al., 2008). The highest PCB concentration could be measured in carpets, in sofas, in floor dust, and in the footwell of the employee's cars. The highest PCB value was detected in the dust of the carpet at the

Table 1
PCB biomonitoring results in workers and relatives from 2010 (IQR: inter quartile range).

Variable	age	PCB28 (BAR < 0.02 µg/L)	PCB52 (BAR < 0.01 µg/L)	PCB101 (BAR < 0.01 µg/L)	PCB153	PCB138	PCB180	Sum ind. PCB	PCB118
All (n = 54)									
Mean (SD)	27.3 (18.6)	6.9 (27.9)	0.53 (1.4)	0.63 (1.2)	3.8 (6.4)	4.2 (7.2)	1.9 (3.2)	18 (41.8)	2.6 (5.1)
IQR	32.2	2.6	0.22	0.47	4.50	4.38	2.8	16.7	2.9
Median	28.4	0.44	0.05	0.13	0.98	1.03	0.43	3.98	0.63
Range	0–63	0.44–178	0.01–8.2	0.01–5.4)	0.07–29.6	0.07–33.2	0.03–14.5	0.2–236	0.04–28.6
Employee (n = 18)									
Mean	36.8 (11.3)	19.5 (46.6)	1.5 (2.2)	1.7 (1.7)	9.9 (8.4)	10.91 (9.52)	4.8 (4.1)	48.2 (63.2)	6.9 (7.2)
IQR	17.1	4.59	1.6	2.5	12.9	13.4	5.9	29.8	6.8
Median	32.6	3.81	0.72	1.2	7.5	8.5	3.5	25	4.7
Range	24.1–63.8	0.33–178	0.01–8.2	0.01–5.4	0.82–29.6	0.83–33.2	0.42–14.5	2.8–236	0.53–28.6
All relatives (n = 36)									
Mean	22.5 (19.8)	0.65 (1.5)	0.06 (0.1)	0.12 (0.13)	0.83 (0.77)	0.82 (0.74)	0.42 (0.46)	2.90 (2.81)	0.48 (0.47)
IQR	33	0.38	0.05	0.12	0.98	0.82	0.5	3.43	0.53
Median	13.5	0.24	0.03	0.1	0.53	0.51	0.3	1.83	0.3
Range	0–61	0.02–8.9	0.01–0.58	0.01–0.66	0.07–3.03	0.07–2.69	0.03–2.02	0.2–12.9	0.04–2.01
Children (n = 20)									
Mean	7.1 (6.1)	0.3 (0.19)	0.04 (0.03)	0.1 (0.1)	0.66 (0.43)	0.7 (0.47)	0.26 (0.17)	2.1 (1.5)	0.43 (0.29)
IQR	9.6	0.12	0.03	0.13	0.8	0.9	0.32	2.2	0.55
Median	5.3	0.46	0.04	0.08	0.62	0.68	0.25	1.8	0.42
Range	0–19.8	0.02–2.1	0.01–0.15	0.01–0.29	0.07–2.4	0.07–2.6	0.03–0.96	0.2–6.5	0.04–1.6

entrance of one worker's apartment with 530 mg/kg. In contrast, 12.3% of all wipe samples exceeded a surface PCB load of 5 µg/m^3. Elevated values could be detected on the floor and in particular on storage places for the working clothes. The highest PCB value was detected on the floor in front of the washing machine with 2000 µg/m^2.

Only one indoor air sample exceeded the guidance value of 300 ng/m^3 (Umweltbundesamt, 2007). We analyzed the distribution pattern of the PCBs and compared it to the PCB congener pattern of the company. Here, the distribution of the PCB congeners was completely different in comparison to the congener distribution at the recycling company. At the recycling company as well as in the dust samples, the PCB congeners 101, 138, 153, and 180 were found. The indoor air sample was dominated by PCB 28 and 52. Therefore, this exposure is likely due to indoor PCB exposure from the building substance (e.g. sealants), while the PCB source within the building is unknown.

Material samples were taken from the washing machines and the dryers in three households. Here, the sealing of the washing machine showed abnormal values in all three households. The plastic components of the washing machine, as well as the sealing and the plastic components of the dryer contained high levels of PCBs. Those levels ranged from 98,5–5340 mg/kg. In 13 households, PCB free cloths were washed and dried with the devices of the participants. In 16 from 21 cloths, PCBs were detected after washing and drying. A maximum of 1 mg/kg PCB was transferred from the machine into the cloth after drying in one household. Lints inside the washing machine contained up to 8.5 mg/kg PCBs. In addition, worker's private clothes were analyzed regarding their PCB concentration. The highest PCB concentration was detected in the sweatpants and the t-shirt of one employee with 169 mg/kg and 15.5 mg/kg PCBs.

Correlation analyses found an association between the PCB dust sample hot spots and the internal PCB 28 and PCB 52 exposure (Fig. 1 A and C). After eliminating the outliers, the association remained weak for PCB 52 with an rho of 0.3 ($p = 0.03$) and for PCB 28 with rho = 0.32 ($p = 0.02$). Correlations between other samples and the internal PCB exposure did not reveal any significant effect (Supplemental rank correlation matrices).

4. Discussion

In this study, we observed a contamination of employees as well as their in-house relatives with PCB, caused mainly by improper recycling of industrial capacitors and transformers. In particular, the children of the recycling workers showed elevated blood levels for PCBs. Although the BAR values are developed as an indicator for occupational exposure and are not valid for the general population, the plasma PCB concentration distribution indicates an additional exposure exceeding the background pollution.

The aim of these analyses was to reconstruct the pathway from the recycling company to the worker's homes. At the recycling company, a large proportion of temporary workers have been employed. Due to these temporary contracts, the workers did not receive cleaning services for their working clothes. Therefore, the employees took their working clothes home, washed, and dried it in their personal washing machines and dryers. In the ambient monitoring results, we found elevated PCB

Table 2
Ambient monitoring results (elevated values in brackets). Elevated values indicate the proportion of elevated samples from all taken samples.

Samples	n house-hold	Elevated values n (%)	Mean (SD)	IQR	Median	Minimum	Maximum
Dust PCB[a] mg/kg (1,65 mg/kg)	18	44 (59.5) n = 74	52.2 (123.1)	63.5	11	0.75	530
Wipe PCB[a] µg/m^2 (5 µg/m^2)	18	12 (10.3) n = 117	164.1 (479.7)	172.5	11.3	0.52	2000
Air PCB[a] ng/m^3 (300 ng/m^3)	6	1 (9.1) n = 11	99.8 (108.3)	228	51.3	41.7	318
Cloth[b] PCB[a] WM/D mg/kg	13	16 (76.2) n = 21	1.15 (2.41)	3.42	0.15	0.01	8.5
Clothes PCB[a] mg/kg (10 mg/kg)	3	4 (66.7) n = 6	66.1 (89.1)	43.3	17.4	12	169
Sealing WM ΣPCB x 5 mg/kg (50 mg/kg)	3	3 (100) n = 3	2419.5 (2671.7)	(5142.5)	1820	98.5	5340
Plastic WM ΣPCB x 5 mg/kg (50 mg/kg)	2	1 (50) n = 2	511.5 (704.99)	(997)	511.5	13	1010
Sealing Dryer ΣPCB x 5 mg/kg (50 mg/kg)	2	1 (50) n = 2	249 (7.07)	(125.5)	249	244	254
Plastic Dryer ΣPCB x 5 mg/kg (50 mg/kg)	2	1 (50) n = 2	58.5 (34.5)	(53)	58.5	32	85
Filter Dryer ΣPCB x 5 (50 mg/kg)	2	0 (0) n = 2	0.76 (1.1)	(1.5)	0.76	0.01	1.5

[a] Sum of PCB by DIN/LAGA, WM = washing machine, D = dryer.
[b] After washing drying.

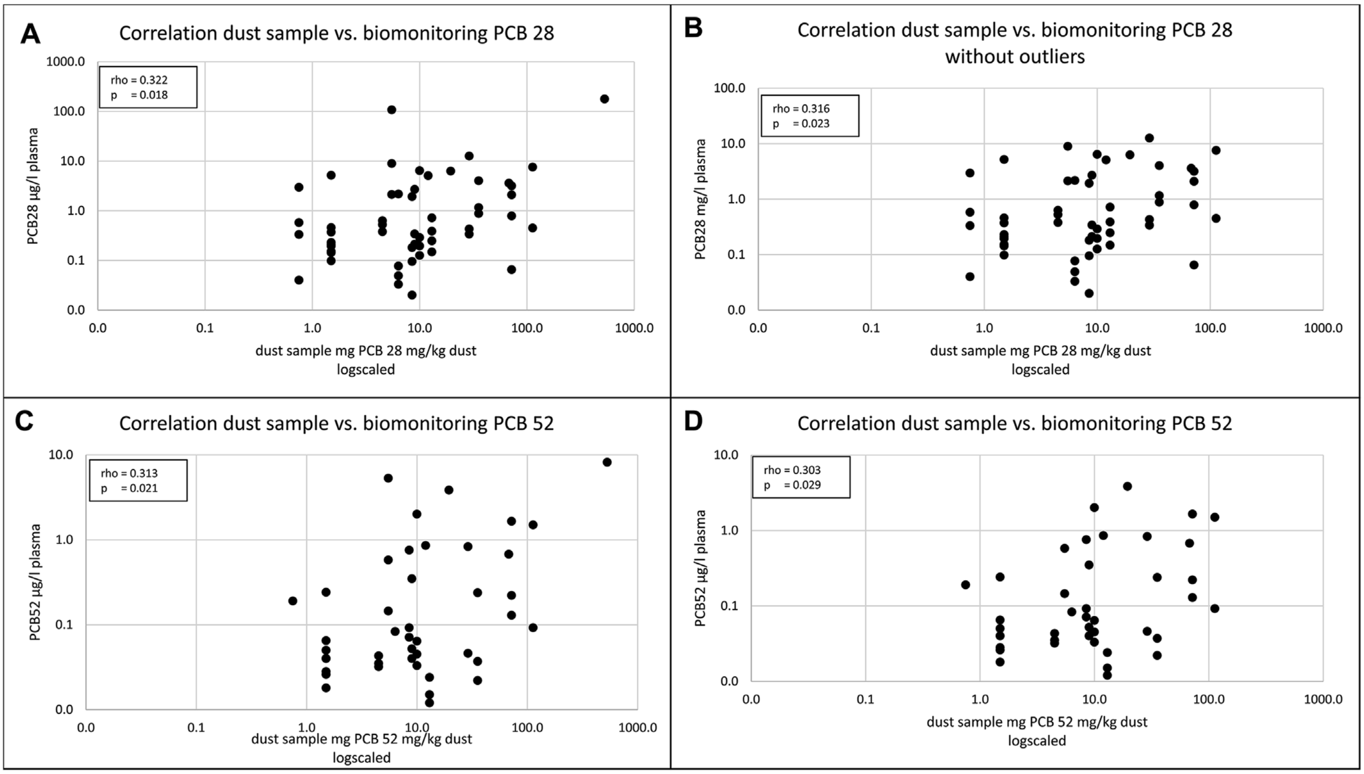

Fig. 1. Spearman correlation between ΣPCB in dust samples and PCB 28 (A)/PCB 52 (C) blood concentration with outliers, and corrected without outliers for PCB 28 (B) and PCB 52 (D).

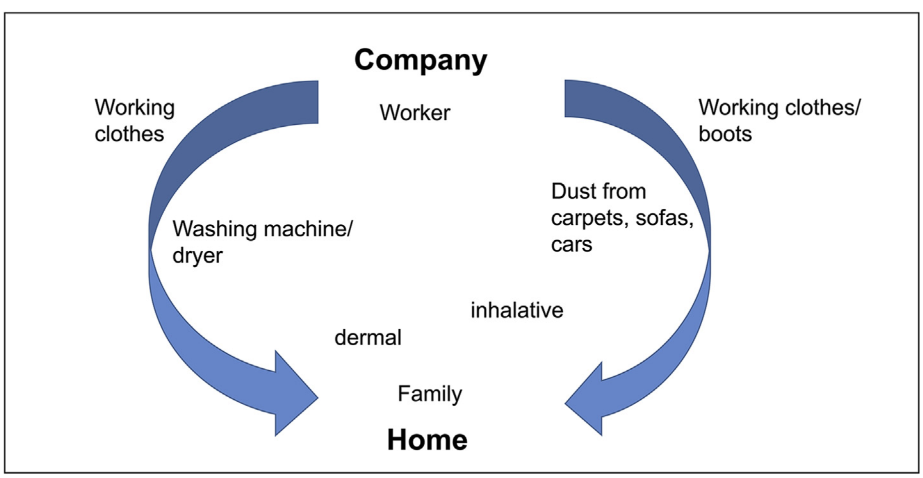

Fig. 2. Exposure routes for PCB from the company to the households of the workers.

values at the places, where the used clothes were stored, and in particular in and around the washing machine/dryer. After washing PCB-free cloths for ambient monitoring analyses, a contamination route from the washing machine or dryer components to the cloth was detectable. In the sealing of the washing machines, PCB concentrations greater than 5 g/kg were detected. In two households, the PCB congener pattern in the sealing of the washing machines and/or dryers was analogous to the PCB congener pattern in the company. Values exceeding 5 g/kg are a marker for the strong contamination of the devices. Even after chemical cleaning of one of the workers personal T-Shirt, abnormal PCB values were still measurable. We can therefore clearly define a contamination pathway from the employees working clothes to the washing machine and from the washing machine to the clothes of the other family members. The contaminated clothes of the family members lead to a dermal exposure towards PCB and therefore an elevation of PCB blood levels, based on their lipophilic character (Fig. 2).

Working boots and again working clothes play a crucial role in a second contamination (Fig. 2): From dust sample analyses, we identified carpets and sofas as highly PCB containing components in the households. In addition, we found elevated PCB values in the car, and here in particular in the footwell. In one family, the sofa, where the worker was used to sit on with his working clothes after finishing his job, was moved to the room of his child. In this household, the highest PCB dust values were therefore detectable in the children's room on the sofa. In another household, the locker for the working boots was the place with the highest PCB concentration. The working clothes and boots contaminated the carpets, the sofas and the cars. The exposure pathway from carpet, sofa or car is probably given via dermal contact but mainly also via inhalation of the dust. In contrast, wipe samples collected on smooth surfaces did contain abnormal PCB values only in a few cases. Indoor air samples were only abnormal in one case.

Interestingly, in relatives of recycling workers, mainly increased levels for lower chlorinated PCBs were seen. In contrast, recycling workers showed also elevated levels for higher chlorinated PCBs. This finding is in accordance with previous studies, where the uptake of lower chlorinated PCBs were based on inhalative and dermal exposure (Dennerlein et al., 2013; Schettgen et al., 2012a). Dennerlein et al. showed, that PCB28, 52, or 101 were more likely to diffuse through the human skin in comparison to higher chlorinated PCBs, such as PCB 180 (Dennerlein et al., 2013). This uptake behavior explains the high levels for PCB 28, 52, and 101, while higher chlorinated PCBs were not elevated. Schettgen et al. estimated an internal exposure of 5–10% for lower chlorinated PCB in relatives in comparison to that of the workers (Schettgen et al., 2012b).

The transmission of hazardous substances from the occupational environment to the indoor environment of the cohabitant family has been described before (Knishkowy and Baker, 1986). There are several cases, where "occupational diseases" occurred in in-house relatives of workers. Knishkowsky et al. described these diseases as "para-occupational illness" (Knishkowy and Baker, 1986). For TCDD (2,3,7,8-Tetrachlorodibenzodioxin) the occurrence of chloracne in family members of workers, who had contact to that substance, have been observed (May 1973). Jensen described chloracne in the son of a worker after close contact with his father's 2,4,5-trichlorophenol-contaminated working clothes (Jensen, 1972). For PCBs, Baker et al. reported elevated PCB levels in family members of workers from an electrical capacitor manufacturing plant (Baker et al., 1980). An exposure pathway has not been investigated in that case. The CDC described lead contaminations in children from persons who are exposed to lead dust during work (Centers for Disease and Prevention, 2009). In these cases, the lead dust was transferred into the vehicles and in the children's safety seats. These children showed elevated blood lead levels (Centers for Disease and Prevention, 2009). Baker et al. described cases of lead poisoning from lead containing dust already in 1977 (Baker et al., 1977).

Elevated values for persistent organic pollutants in ambient monitoring studies have been shown before. Deziel et al. reported about the association of elevated PCDD/F house dust values and the proximity to industrial facilities as well as freight routes and major roads (Deziel et al., 2012). In contrast, Franzblau et al. could not find a source for the dust contamination with PCDD/F and dl-PCB in five Michigan counties. The authors could only suggest, that the source might be within the house (Franzblau et al., 2009). Whitehead et al. investigated determinants of PCB in dust from several households in California (Whitehead et al., 2013). In particular, in buildings that were built before 1980, higher PCB levels could be observed. In the study of Whitehead et al., the occupation of the residents as a determinant for PCB contamination was assessed, as well. The authors found a significant association between an occupation with PCB exposure and the odds to detect elevated in-house PCB levels (Whitehead et al., 2013).

Elevated PCB concentrations in humans can have several causes. It has been described before, that nutritional related exposure towards PCBs lead to elevated levels of higher chlorinated PCBs and dioxin-like PCBs with increasing age, due to an accumulation in the human body (Safe, 1993). Therefore, a nutritional related PCB body burden shows predominantly a pattern of higher chlorinated non-dioxin like PCBs (Faroon and Ruiz, 2016; Quinn and Wania, 2012). The reason for the accumulation of higher chlorinated PCBs in the human body can be explained by the long half-life time of higher chlorinated PCBs. Lower chlorinated PCBs show only a short biological half-life time and can therefore not accumulate in the food chain or human body (Schettgen et al., 2012a; Wimmerova et al., 2011). Therefore, a mixed PCB congener pattern involving lower chlorinated PCBs, such as PCB 28, 52 and 101, is more indicative of non-food-induced exposure.

In our analyses, the pathway from the company to the household has a strong evidence. Although nutritionals and environmental aspects may play a role, in our analyses, this role might have a minor impact. The profile of PCB congeners measured in different media/matrices (dust, wipe samples, textiles, material, plasma) indicates due to similarity of profile a pathway from the company to the household and to human exposure. We found in particular elevated values for lower chlorinated biphenyls (PCB 28, 52, 101). Lower chlorinated biphenyls are a marker for occupational exposure and not an indicator for nutritional related PCB uptake (Safe, 1993). Since the median PCB 28, 52, and 101 levels in the children of the employees exceeded the BAR, the relation to the recycling company and therefore, the exposure route from the worker to the family can be clearly stated.

Our study has several limitations. First, we only had a small sample size of 54 participants. The small sample size falls in particular into weight, since we made subgroup analyses with even smaller sample sizes, for example in employees (n = 18) or in children (n = 20). Furthermore, not for all households, full environmental monitoring analyses were available. For example, we only had cloth samples in n = 13 homes or sealing samples in n = 3 households. Therefore, correlation analyses were only restricted feasible and analyzable.

In conclusion, it should be avoided to take home clothes or boots, used in a contaminated occupational environment, to prevent contamination of the bystanders environment and to avoid exposure of children and other family members.Acknowledgements

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Conflicts of interest

All authors declare no conflict of interest.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijheh.2019.08.004.

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