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PCB in serum and hand wipes from exposed residents living in contaminated high-rise apartment buildings and a reference group

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ABSTRACT

Exposure to polychlorinated biphenyls (PCB) in buildings constructed with PCB-containing materials can lead to increased body burden of PCB for building users. Exposure to PCB from building related sources was assessed by measuring PCB in serum and hand wipes in two groups of tenants living in the same building estate in apartments constructed with and without PCB. The median serum levels of the sum of 19 PCB congeners was 777 ng/g lw (5–95th percentile: 219–2576 ng/g lw) for the exposed group and 282 ng/g lw (5–95th percentile: 49.8–797 ng/g lw) for the reference group. The congener pattern in serum of the exposed was shifted towards lower chlorinated congeners and was dominated by tri- and tetrachlorinated congeners. The largest difference in serum levels between the groups was observed for PCB-28, with median levels of the exposed being 70 times higher than in the reference group. For very persistent lower chlorinated congeners, like PCB-28, 66 and 74, an increase with residence times was observed as a result of accumulation over time. Less persistent congeners such as PCB-44 and 70 were also elevated in the exposed group -but independent of residence time. The less persistent congeners can therefore be used as markers of recent exposure to original PCB sources. The hand wipes also showed a large exposure contrast, e.g. PCB-28 being more than 60 times higher in the exposed group compared to the reference group (medians 14.4 and 0.23 ng/wipe, respectively) and no overlap between the groups. All measured di- to pentaPCB congeners were significantly higher in hand wipes from the exposed group compared with the reference group. Thus hand wipes seem to be a good, non-invasive screening tool for ongoing indoor exposure to PCB. Furthermore, the measured level of PCB on hand wipes was significantly correlated to PCB in blood for almost all congeners. In conclusion, PCB exposure in contaminated buildings can contribute significantly to the exposure and total body burden of PCB and the lower chlorinated congeners can make up the majority of the total PCB body burden.

1. Introduction

Polychlorinated biphenyls (PCB) were among other things used as plasticizer and dielectric fluid in the 1950s–80s. Even though the production ceased almost 40 years ago, the original materials such as elastic sealants and capacitors in fluorescent lighting ballasts are still present in buildings in use, potentially resulting in severe contamination of the indoor environment and exposure of building users.

PCB comprises 209 different congeners, many of which are highly persistent. PCB was included in the Stockholm Convention as one of the initial “Dirty Dozen”. PCB has been classified as carcinogenic to humans (Group 1A) IARC (2016). Most of the knowledge on toxicity of PCB is based on either technical mixtures or the congeners that are typically present in food, i.e. highly chlorinated, persistent congeners such as PCB-153, 138 and 180. However, in contaminated buildings the majority of the exposure is to lower chlorinated congeners (≤4 Cl) as much of the exposure is mediated through air. Even highly chlorinated mixtures, such as Aroclor 1260, releases a significant fraction of lower

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chlorinated congeners into the gas phase (Carpenter, 2015). The toxicity of lower chlorinated congeners is much less studied than higher chlorinated congeners and mixtures, but more recent studies indicate that lower chlorinated congeners are also carcinogenic, have neurotoxic effects and may cause diabetes, hypertension and cardiovascular disease and act as endocrine disruptors (Aminov et al., 2016; Carpenter, 2015; Gallo et al., 2018). Furthermore, their metabolites may be of equal concern due to their effects on the endocrine system, persistence as well as geno- and neurotoxicity (Antunes-Fernandes et al., 2011; Quinete et al., 2017; Rodriguez et al., 2018; Vasko et al., 2018).

In 2009, the Danish Health Authority recommended two action levels for PCB in indoor air; the levels are 300 ng PCB_{total}/m^3, exceedance is considered to pose a possible health risk and an action plan is needed to bring levels down; and 3000 ng PCB_{total}/m^3, above which immediate action is required (Jensen, 2013). In regard to Danish legislation, PCB_{total} in air is defined as the sum of seven indicator congeners (PCB-28, 52, 101, 118, 138, 153 and 180) multiplied with a factor of five based on German values (Jensen, 2013). The recommendations have led to increased awareness about potential PCB exposure indoors and many social housing organizations and municipalities started screening their buildings. On a national level, it is estimated that 60–95% of Danish buildings constructed in 1950-77 contain some PCB and that 0.7–1.6% of all buildings in Denmark have indoor air levels exceeding the lower recommended action level (Grontmij, 2013).

As a result of the screening among social housing organizations, two large estates were identified as having both contaminated and non-contaminated dwellings due to extensive use of PCB-containing elastic sealants in the sections construction first. The two estates were Farum Midtpunkt (FM) and Brøndby Strand Parkerne (BSP). Common for both of these estates are that the first sections were built with PCB-containing sealants, while the rest were not. The exposure of the residents in FM was extensively studied taking samples of air, sealant and blood of residents in both contaminated and non-contaminated apartments, this is described in detail elsewhere (Frederiksen et al., 2012; Meyer et al., 2013).

In the current study, exposure of the residents in BSP was examined. Compared to FM, a number of lower chlorinated congeners were added to the analyses and more matrices were included in the indoor exposure description (see Andersen et al.) both providing more information on the specific exposure from the indoor environment. The aim of the current paper is to confirm the indoor environment as a substantial contributor to the body burden when living in PCB contaminated buildings. Furthermore, the use of hand wipes as a possible screening tool for ongoing PCB-exposure and its correlation with blood levels were also investigated.

2. Materials and methods

2.1. Population and recruitment

BSP is a social housing area located approximately 15 km southwest of Copenhagen, Denmark. It consists of 12 identical high-rise apartment buildings (15 floors; 4–5 apartments/floor) and lower apartment buildings surrounding them. The high-rises were constructed from the late 1960s to early 1970s. During the construction, PCB-sealants were used in the first five high-rises, but not in the last seven. Thus, approximately 300 apartments were highly contaminated with PCB, while more than 400 high-rise apartments were not; further details about the high-rises can be found elsewhere (Andersen et al.; Lyng et al., 2016; Morrison et al., 2018). Prior to the study, a consulting company had mapped the contamination and found indoor air levels up to 5000 ng/m^3 PCB_{total} (median: 1260 ng/m^3) in the first five high-rises and $<$ 300 ng/m^3 (median: 35 ng/m^3) in the last seven (Golder Asssociates, 2017). More detailed analyses have shown, that the indoor air is dominated by lower chlorinated PCB congeners in a very consistent pattern; the most abundant measured were PCB-18, 28, 31 and 52, furthermore, analyses of sealant material have indicated that most likely it was Clophen A-40 or Aroclor 1248 that was used (Andersen et al.).

For this study, participants in the “exposed group” were recruited among tenants of the five high-rises constructed with PCB-sealants, while the “reference group” was recruited from the remaining seven high-rises as well as the surrounding lower houses. Funding allowed recruitment of up to 100 participants, thus we were aiming at 80 exposed and 20 reference participants. Participants were recruited through invitation letters distributed to all tenants in the five contaminated high-rises as well as three of the reference high-rises. In addition, posters were hung in all high-rises as well as in common facilities throughout the estate and locals helped spread the word. Three information meetings were held at different dates and locations of the estate. The inclusion criteria for participants were: $\geq$18 years of age; residing at least 6 months in the current apartment. All participants signed a written, informed consent. The study was approved by the Regional Ethics Committee (H-16041946) and reported to the Data Protection Agency through University of Copenhagen (SUND-2017-03). All participants were informed about their individual results upon request.

2.2. Sampling

The blood samples were drawn in two temporary clinics set up in local, non-contaminated common facilities; in a few cases where the participants were not able to come to the common facilities the samples were taken in their home. Approximately 50 ml (5 × 10 ml) of blood was drawn into Vacutainer tubes (BD Vacutainer, CAT ref. 367896), and after at least 30 min they were spun at 2500 G for 15 min. Serum was collected and stored on ice before and during transport to University of Copenhagen where they were stored at −20 °C. At the session for taking the blood sample, the participants also filled in a detailed questionnaire. The most relevant questionnaire topics for this paper were age, gender and address history within the estate.

Appointments for two home visits for indoor measurements were also made. Hand wipes were taken at one of the home visit using folded isopropanol wipes (Alkoholswab: 70% isopropanol, 30 × 60 mm, Mediq Danmark A/S), wiping from wrist to each fingertip, then the wipe was folded inside-out and the wiping procedure was repeated on the same palm. Both left and right palm were wiped (using two wipes) and the wipes were put together in a single 10 ml amber glass vial (PTFE/butyl rubber septum) in which they were later extracted. In addition, back-of-hand samples were also collected following the same sampling procedure; except for a small subset of back-of-hand samples, only palm samples were analyzed. The research assistants were instructed to change gloves between participants. The participants were asked not to wash their hands 30 min prior to sampling, and the approximate time since last hand wash was recorded. All vials from the same address were stored in a single Rilsan bag at −20 °C until analysis.

All samples were collected between October and December 2017. After completing the sample collection, serum samples were packed on dry ice and all samples were shipped to the Norwegian Institute of Public Health, where they were again stored at −20 °C until analysis. Along with the blood and hand wipes samples, matched measurements of PCB in air, dust and surface wipes in the participants’ apartments were performed. The results from these measurements are reported in detail elsewhere (Andersen et al.).

2.3. Serum and hand wipe analyses

In Denmark and most other European countries seven indicator PCB congeners (PCB-28, 52, 101, 118, 138, 153 and 180) are most often reported. However, the lower chlorinated congeners, which are more abundant in the indoor air of buildings with PCB-containing materials, are not well represented among the indicators. As an economical compromise between number of samples and congeners it was chosen to add as many lower chlorinated congeners as possible to the existing method. Priority was given to congeners previously reported in full congener analyses of e.g. air from schools (Ampleman et al., 2015) and blood of workers in contaminated buildings (Kraft et al., 2017). Knowledge of congeners present in the original technical mixtures with low to medium chlorination (e.g. Frame, 1997; Schulz et al., 1989) as well as recent findings of from biomonitoring studies was taken into consideration. Kraft et al. (2017) found all DL-PCB congener to constitute < 1% of the total PCB burden and Meyer et al. (2013), who analyzed five DL-PCBs, only detected PCB-169 in 1.4% of the samples from highly exposed residents. Based on this, the following congeners were given priority and selected for analysis of serum and hand wipes: PCB-8, 18, 28, 31, 44, 52, 66, 70, 74, 99, 101, 105, 118, 138, 153, 170, 180, and 187; in addition 194 was measured in serum.

The serum samples were extracted and analyzed as described in Caspersen et al. (2016), except that GC-MS/MS was used for detection and that an HT8 column (60m, 0.25 mm x 0.25id) was used for the GC separation. ^{13}C- labeled internal standards were used for quantification. ^{13}C-Isotope labeled internal standards of the identical PCB congeners were used for all except PCB-31 (^{13}C PCB-28), PCB-44, 66, 70, and 74 (^{13}C PCB-52), PCB-99 (^{13}C PCB-101) and PCB-187 (^{13}C PCB-170). To ensure high quality of the determinations throughout the project, procedural blanks (water of LC-MS/MS grade, n = 22) and in-house control serum samples (commercially available human serum, n = 4) were analyzed along with the serum samples.

The instrumental analyses of the hand wipe samples were performed using the same method as for the serum samples, while the extraction procedure was somewhat different. In short, the amber glass vials, each containing two hand wipes, were opened and internal standards along with 5 mL of dichloromethane (DCM) was added to each sample, which was put in an ultrasonic bath for 10 min. The DCM was transferred to a reagent tube and another 5 mL of DCM was added to the amber glass vial and left in an ultrasonic bath for another 10 min. DCM from the second extraction was combined with the first extract, evaporated to 1 mL using a Rapid Vap ® (Labconco Cooperation, 8811 Prospect Avenue, Kansas City, Missouri, USA) at 34 °C and 700 mbar. The concentrated extract was then added 3 mL of heptane, cleaned-up on a sulfuric-silica column and concentrated under nitrogen, in the same way as for the serum samples. Unused wipes where used as procedural blanks (n = 6), these were left under a gentle stream of nitrogen at 40 °C for 15 min for evaporation of remaining isopropanol in the wipes before adding internal standard.

Concentrations of cholesterol and triglycerides in serum were determined by the Fürst Medical Analysis Laboratory using the ADVIA® Chemistry XPT System. All serum levels were lipid adjusted using the measured cholesterol and triglyceride concentrations and the conversion method described by Covaci et al. (2006).

Low levels of PCB-8, 18, 28, 31, 44, 52, 101, 138 and 153 were consistently observed, while PCB-66, 70, and 99 were occasionally observed in the blanks. The source is believed to be the windows of the laboratory building, despite much effort it was not possible to eliminate the background contamination completely. Therefore blank correction was applied for these congeners by subtraction of the average blank concentration (separate for serum and wipes) and adjusting LOQ to the highest blank level for each congener. For congeners with no quantifiable blank levels, LOQ was set to the instrument LOQ of 1–10 pg/sample; details of blank levels and LOQ can be found in Table S1.

2.4. Statistics

Generally, non-parametric tests were used in the statistical analyses i.e. not assuming normal or log-normal distribution. This was chosen being simple and robust models, which are less sensitive to extreme values though with the cost of non-parametric models generally having less power than parametric. For correlation analyses Spearman's Rank correlations were made with a significance level of 5%. Groups were compared using Mann-Whitney *U* test also with a significance level of 5%. Values below limit of quantification (LOQ) were replaced with half the value of LOQ in all statistical analyses and no correction for potential confounders were applied in these analyses. All statistical analyses were performed using GraphPad Prism v. 8.0.2.

Table 1
Demographics of the exposed and reference groups. 'Gender' and 'smoking': n (%); 'age', 'years in BSP', 'BMI', 'est. palm surface area': mean [5th - 95th percentile], p-value from *t*-test.

	Exposed group (n = 71)	Reference group (n = 23)	p
Gender			
Men	24 (34%)	8 (35%)	–
Women	47 (66%)	15 (65%)	
Age [y]	62.4 [28.8–85.2]	61.4 [33.2–87.4]	0.79
Years in BSP	16.1 [1.7–38.8]	12.6 [1.0–39.2]	0.19
BMI [kg m^{-2}]	27.5 [18.1–41.0]	28.9 [20.0–39.9]	0.38
Est. palm surface area [cm^2]	472 [371–583]	462 [347–633]	0.62
Smoking			
Current	15 (21%)	3 (13%)	–
Never	30 (42%)	10 (43%)	

3. Results

A total of 94 participants were recruited: 71 in the exposed and 23 in the reference group; details on the demographics of the groups are given in Table 1. Three participants from the exposed group no longer had their official address in a contaminated apartment as they had moved recently; two of these were still visiting and sleeping there several times per week and were fully included in the study. For the latter no hand wipe sample was taken; in addition two hand wipe samples were missing or mis-sampled i.e. three hand wipe samples are missing in the exposed group. Generally, the two groups were very similar on the recorded demographic parameters (Table 1). The exposed group was marginally older and had lived there slightly longer, but in general it was an aging population with a median age above 60y; in both groups and there were a majority of women. More than 90% had lived in BSP for more than 5 years and 13% more than 30 years; the median residence time was 11 years. Two persons from the reference group had previously lived in the contaminated section (< 5y) but had moved out more than 10 years before the time of blood sampling. Finally, the two groups were also similar regarding educational and household income levels.

3.1. Blood

Levels of PCB in serum of the exposed and reference group are shown in Fig. 1. Further details on detection frequency, percentiles etc. can be found in Table S2, for comparative reasons the results for all congeners based on wet weight are given in Table S3. The congener pattern in serum from the reference group followed the regular pattern of PCB-153, 180 and 138 being the most abundant of the analyzed congeners. The congener pattern in the exposed group was clearly shifted towards lower chlorinated congeners and dominated by PCB-28 followed by similar levels of PCB- 66, 74, 153 and 180. Statistically significant differences between the groups were observed for all tri- to pentachlorinated congeners except for PCB-18 and 101. The largest relative difference between the two groups was observed for PCB-28 as the median level of the exposed group was approximately 70 times higher than the reference group. For the sum of all 19 measured PCB congeners, the exposed group had a median level 2.8 times higher than the reference group and for most of the exposed participants lower

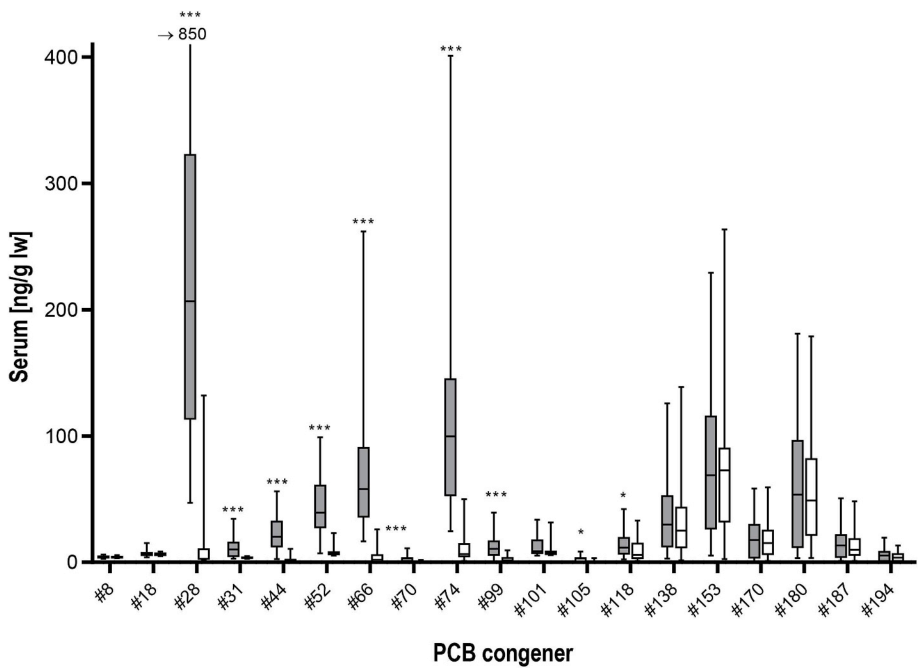

Fig. 1. Serum levels [ng/g lw] in the exposed (grey, n = 71) and reference groups (white, n = 23). Boxes with interquartile range and whiskers of 5th and 95th percentiles. Level of p-values in Mann-Whitney U-tests are indicated by asterisks: * < 0.05; ** < 0.001, *** < 0.0001. < LOQ were set to ½LOQ.

chlorinated congeners made up the majority of the estimated total body burden (Table S2).

A large fraction of the participants (43%) shared their apartment with at least one other participant; among these were 14 couples (11 exposed/3 reference). Statistically significant Spearman's rank correlations of paired samples from couples were obtained for various congeners including PCB-28, 44 and 153 when including the reference couples in the analyses. However, if only analyzing exposed couples, the significance only remained for higher chlorinated congeners including PCB-153 (see Fig. S1), but the dataset was very small and it should be interpreted with caution.

For PCB-28 and 44 and lower chlorinated congeners in general, elevated levels were seen for the exposed group even after only a few years living in a contaminated apartment (Fig. 2 and Fig. S2). Significantly higher levels of PCB-28, 31, 44, 52, 66, and 74 were observed in the exposed participants with less than 5 years of residency relative to the reference group (data not shown). Fig. 2 also shows examples of three different patterns of the association between congener levels and years in dwelling. PCB-28 increased with increasing residence time for the exposed participants indicating accumulation over the years (Spearman's rank r = 0.46, p < 0.0001) whereas the level in the reference group remained stable and independent of residence time. On the other hand, PCB-44 concentrations in the exposed group were not associated with length of residency (Spearman's rank r = 0.02, p = 0.86). For highly chlorinated congeners such as PCB-153, an identical increase with years in dwelling was observed for the two study groups.

3.2. Hand wipes

Levels of PCB on hand wipes from the exposed and reference group are shown in Fig. 3 and further details on detection frequency, percentiles etc. can be found in Table S4. For hand wipes the differences between the exposed and reference group were even more pronounced than in serum. The levels in the reference group were typically one to two orders of magnitude lower and statistically significant differences between the groups were observed for all analyzed di-to penta-PCB congeners. Hand wipes from the exposed group were dominated by PCB-18, 28, 31, 44, and 52, whereas in the reference group the concentrations were much lower and more similar across congeners including highly chlorinated ones.

Both palm and back-of-hand samples were analyzed from six participants. Generally, fairly good correlations were observed between the two sampling areas, but there was no clear trend of levels being higher on one compared to the other (Fig. S3). However, since the sample size was very small, these results should be interpreted with caution.

Hand wipe samples from participants living together in the same apartment are shown in Fig. 4. In some cases there is good agreement

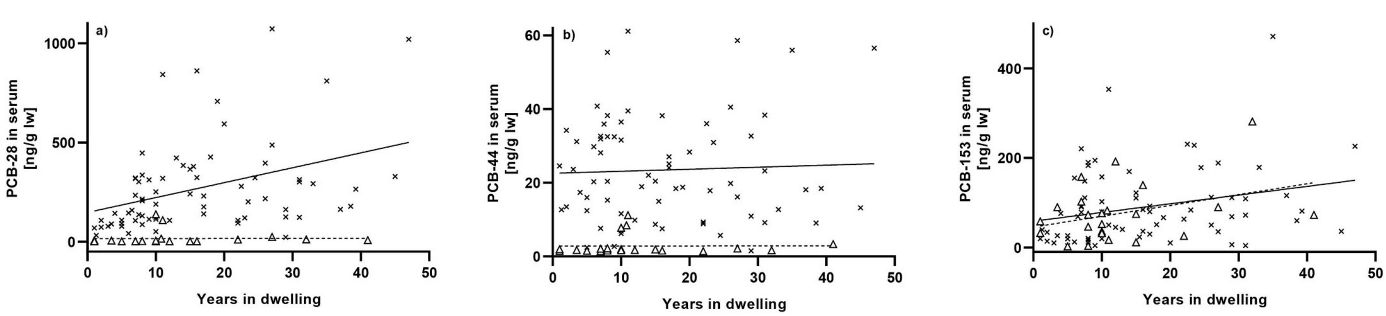

Fig. 2. Levels of PCB in serum as a function of years in dwelling (in case of multiple contaminated dwellings, the time has been summed). Exposed: crosses w. solid line (linear regression); References: triangles w. dotted line (linear regression). Remaining congeners are shown in Fig. S2.

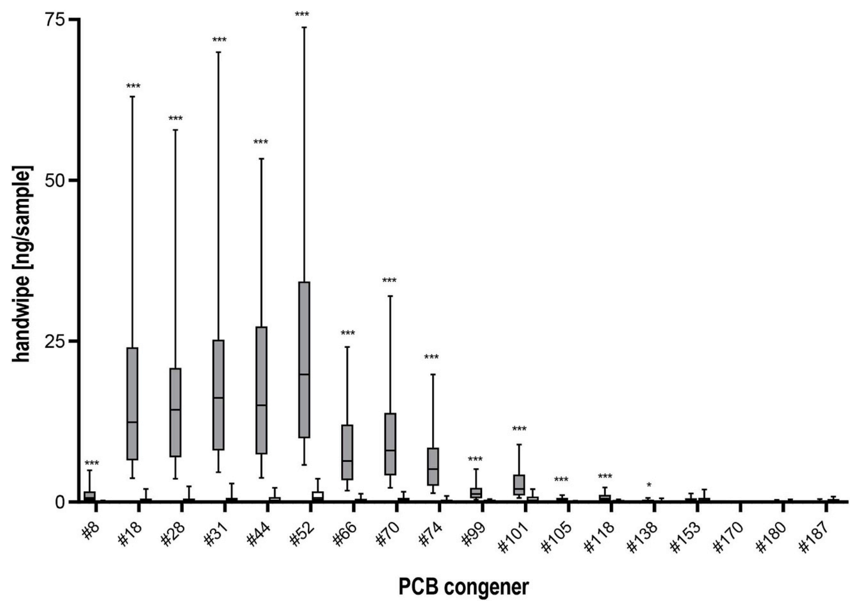

Fig. 3. Hand wipe (palm) levels [ng/sample] in the exposed (grey, n = 68) and reference (white, n = 23) groups. Boxes with interquartile range and whiskers of 5th and 95th percentiles. Level of p-values in Mann-Whitney U-tests are indicated by asterisks: * < 0.05; ** < 0.001, *** < 0.0001. < LOQ were set to ½LOQ.

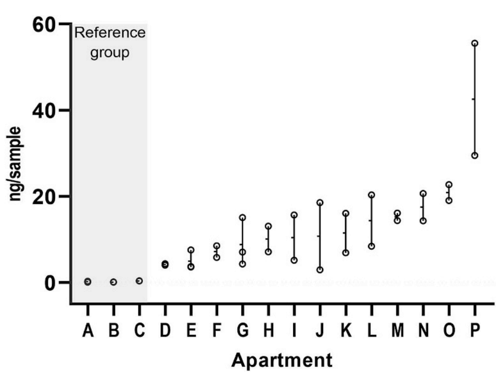

Fig. 4. PCB-28 on hand wipes from participants living together, apartment mean is indicated by horizontal tick. Generally there where two participants per apartment, except in apartment E and G where there were three (individuals are indicated by circles). Mean of references is indicated by the dotted line.

between samples, whereas for others the variation is quite large; there was no apparent relationship with the time since last hand wash.

3.3. Serum vs. hand wipes

Highly significant, though not very strong, correlations were found between the individual congeners on hand wipes and in serum ($p < 0.001$ for most congeners, $r = 0.40–0.55$). When including the reference group, PCB-101 was the only congener with a non-significant Spearman's rank correlation; in the exposed group alone, PCB-101 and 105 were the only congeners not significantly correlated in the two matrices. As an example Fig. 5 shows the correlation for PCB-28.

4. Discussion

4.1. Blood

The reference group had serum median levels and range fairly similar to the reference group of the previous Danish study on exposure in buildings by Meyer et al. (2013), though the 95th percentile in the current study was slightly elevated for lower chlorinated congeners (PCB-28, 52, 66 and 74) due to an unknown exposure of two participants in the reference group. The level of the 95th percentile was similar to that of the German provisional reference value (RV_{95}, 60-69y), but this is only based on three congeners i.e. PCB 138, 153, and 180 (Apel et al., 2017). More information is available on congener levels from NHANES, though the latest dataset based on individual samples is from 2003/04. Generally the levels of the reference group in the current study (Table S2) was similar to or perhaps slightly higher than the 2003/04 "20+ years" of NHANES (CDC, 2019) for both the 50th percentile (e.g. PCB-74: 5.00 and PCB-153: 24.2 ng/g lw) and 95th percentile (PCB-28: 11.1; PCB-74: 50.0 and PCB-153: 101 ng/g lw). This may be due to reasons mentioned above for the lower chlorinated, while the higher chlorinated could be due to our reference group being rather old (mean age: 61y). But overall, though the reference group of the current study is rather small, the blood levels are similar to what has previously been found in groups of similar age with background exposure to PCB.

On the other hand, there are a limited number of studies on body burden of PCB resulting from indoor exposure particularly in homes. The results from a previous Danish study of blood (plasma) from residents of contaminated dwellings in FM (Meyer et al., 2013) were remarkably similar to what was observed in serum in the current study both in terms of level, variation and congener pattern (Fig. 6). FM and BSP where constructed in the same period, but FM had exposed indoor sealants with several different PCB-mixtures being used (Frederiksen et al., 2012), while in BSP apparently the same commercial mixture was used throughout (Andersen et al.); in both estates the variation in air concentration levels were large. As in the study by Meyer et al. (2013), we observed that PCB-28 was by far the most abundant congener measured in the blood of exposed residents, in the current study it makes up on average 30% of the sum of PCB measured (Σ_{19}). The largest exposure contrast between exposed and reference groups was observed for PCB-28, with median values being more than 70 times higher among the exposed in the current study and x100 in the study by Meyer et al. (2013). In biomonitoring studies and surveillance programs e.g. in Germany, PCB-138, 153 and 180 have often been used as the sole indicators of PCB-exposure (the sum multiplied by two) (Kraft et al., 2017). In the exposed group of the current study, the Σ_{19} on average exceeds the estimated total when using only three indicator congeners (PCB-138, 153 and 180) by 300%; when using six indicators (i.e. adding PCB-28, 52 and 101) as suggested by Kraft et al. (2017) Σ_{19} is about 80% of the estimated total. Our results stresses the importance of

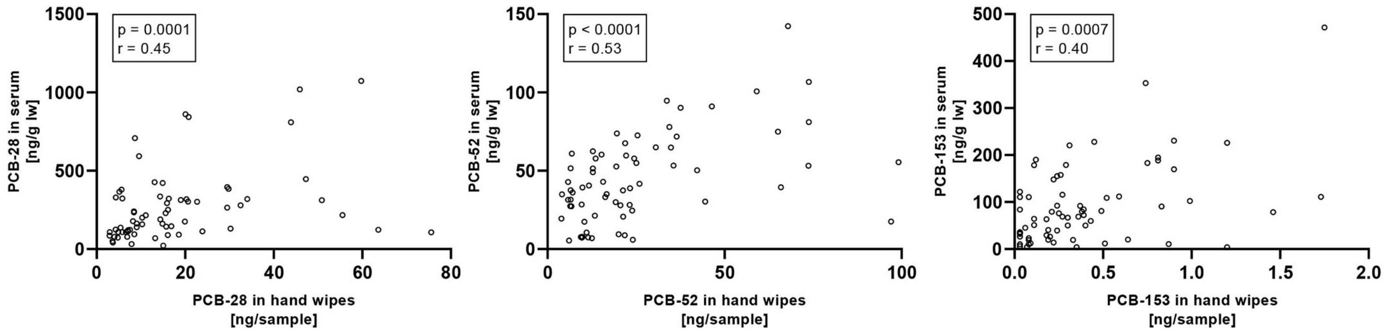

Fig. 5. PCB-28, 52 and 153 in serum vs. hand wipes from the exposed group. Spearman's rank correlation r and p (n = 68).

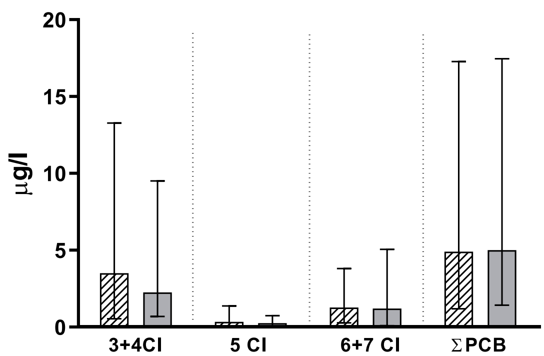

Fig. 6. Levels per homologue group in blood samples from exposed groups of BSP (grey bars; this study, serum) and FM (hatched bars; from Meyer et al. (2013), plasma). Median and 5-95th percentile [μg/l], density of 1.024 g/ml serum was assumed. Included congeners in BSP-bar was based on congeners in common with Meyer et al. i.e. 3 + 4Cl: PCB-28, 52, 66, and 74; 5Cl: PCB-99, 101, 105, and 118; 6 + 7Cl: PCB-138, 153, 170, 180, and 187. ΣPCB all 19 and 27 congeners analyzed in BSP and FM, respectively.

adding PCB-28 and other lower chlorinated congeners in biomonitoring in order to capture potential indoor exposure, as recently suggested by Kraft et al. (2017).

Others have looked at occupational exposure of e.g. teachers and offices workers in buildings contaminated with PCB. Though air levels vary greatly between studies, the levels of lower chlorinated congeners following occupational indoor exposure were generally one order of magnitude lower than observed in the present study as well as other studies of home exposure (Table 2). More time is spent at home, which may explain most of the difference of blood levels following indoor exposure at work versus at home, but other factors may contribute to further increase the difference. One example could be contaminated clothes, Morrison et al. (2018) estimated that wearing contaminated clothes when leaving a contaminated environment e.g. the home cause dermal exposure to continue and increase its relative contribution dramatically. Oppositely, wearing clothes from a non-contaminated home to e.g. a contaminated workplace may have some protective effect for dermal exposure, as has been observed for phthalates (Morrison et al., 2016). Finally, there are other exposure pathways such as dust ingestion and dermal absorption after contact with contaminated surfaces.

Even though the serum levels observed in the current study are among the highest reported for non-occupational exposure, the levels are still markedly lower than historical occupational exposure of e.g. capacitor workers (e.g. Seegal et al., 2011). Compared to more recent studies on occupational exposure, the levels of lower chlorinated congeners are also lower in the current study than observed among workers of a transformer recycling plant (Quinete et al., 2017), but higher than for renovation workers removing old sealants (Kontsas et al., 2004; Wingfors et al., 2006). More worryingly, the levels were in the same range as the levels of two laboratory technicians “sniffing” transformer oil with tragic consequences as described in a case report by Carpenter (2015). However, it is difficult to assess the health risk associated with this exposure as toxicological data specifically on lower chlorinated congeners are still scarce, but this is an area receiving increasing attention. AFSSA has suggested “critical concentration levels in humans” for total PCB of 700 ng/g lw for women < 45y and 1800 ng/g lw for all others (AFSSA, 2010) considering non-cancer effects. In the current study, 7% of the exposed group is exceeding 1800 ng/g lw, while 38% of women < 45y (n = 8) in the exposed group exceeds 700 ng/g lw. No one in the reference group are exceeding the values.

Fig. 2a shows that PCB-28 levels in blood are independent of residence time in the reference group, thus indicating that blood levels of PCB-28 is not age dependent as was also observed for tri- and tetraPCBs by (Meyer et al., 2013) and indicated by the results reported in NHANES (CDC, 2019). Based on the longest reported half-life (5.5 y) for PCB-28 and a first-order decay (Ritter et al., 2011; Seegal et al., 2011), steady state is expected to be reached in less than 25 years ($t_{95\%}$ = 24y) for PCB-28 assuming constant intake and k (rate constant). Due to the large variations in exposure and resulting blood levels between individuals, as seen in Fig. 2a, it is difficult to determine whether serum levels keep increasing past the 25 years or if it levels off. However, the intake may not be constant as participants become older and may change behavior (e.g. staying more at home) or there could be differences in ADME (absorption, metabolism, distribution, excretion) with age. For example age-related tissue clearance of PCB, which has been observed in rats (Birnbaum, 1983) and indicated in humans (Gao et al., 2019). Also the actual exposure in the apartments have most likely increased over time as the PCB is released from the primary source and redistributed to adjacent materials as well as air, dust and other surfaces.

Accumulation patterns similar to that of PCB-28 were observed for other highly persistent congeners such as PCB-66 and 74 (Fig. S2), but unlike PCB-28 a plateau could not be expected for e.g. PCB-74, which has a $t_{½}$ of 15.9 years (Seegal et al., 2011) resulting in a $t_{95\%}$ of 69 years. The patterns for PCB-44 and 70 (Figs. 2b and S1, respectively) are quite different from PCB-28 as accumulation was not observed with increasing residence time. It has not been possible to find good estimates of $t_{½}$ in humans for these congeners in the literature, but due to their chlorine substitution pattern (2,5) they are expected to be more prone to metabolism than the very persistent PCB-28, 66 and 74, which have a mono-ortho and a para-para substitution (2,4,4’) (Hansen, 1999). Wingfors et al. (2006) suggested PCB-44 and 70 as marker congeners for recent occupational exposure. The data from the current study support these congeners as markers of not only occupational but any recent exposure to original PCB sources, even though there are a few participants in the reference group with slightly elevated levels of PCB-44 too.

The highly chlorinated and very stable congeners (e.g. PCB-153 and 180) increase evenly among exposed and reference group with residence time, however the increase more likely reflects the age dependency, and was also observed by Meyer et al. (2013), and is in line

Table 2
Median levels of selected PCB-congeners in blood following indoor exposure in contaminated buildings.

	Blood media	Exposure Environment	PCB-28		PCB-52		PCB-66		PCB-74		comment
	s/p/wb		µg/l	ng/g lw	µg/l	ng/g lw	µg/l	ng/g lw	µg/l	ng/g lw	
*This study**	s	Home	1.35 (0.35–5.90)	207 (47.2–850)	0.29 (< LOQ-0.68)	39.4 (< LOQ - 98.9)	0.38 (0.096–1.81)	57.9 (16.6–262)	0.62 (0.15–2.94)	99.6 (24.6–401)	Median (5–95th percentile), n = 71. Indoor and outdoor sealants
Meyer et al. (2013)	p	Home	1.371 (0.216–5.279)	–	0.216 (0.066–1.218)	–	0.562 (0.103–2.502)	–	1.101 (0.108–5.139)	–	Median (5–95th percentile), n = 139 Indoor sealants
Johansson et al. (2003)	wb	Home	–	88.9	–	–	–	9.50	–	18.48	Median, n = 21. Outdoor sealants
Gabrio et al. (2000)	wb	Work	0.098	–	–	–	–	–	–	–	Mean, n = 151 contaminated schools
Schwenk et al. (2002)	wb	Work	0.24	–	0.07	–	–	–	–	–	Mean, n = 18 contaminated schools
Peper et al. (2005)	wb	Work	0.28	–	–	–	–	–	–	–	Mean, n = 16 contaminated school
Herrick et al. (2011)*	s	Work	0.0227	–	0.0055	–	0.0163	–	0.0574	–	Median, n = 18 contaminated schools
Schettgen et al. (2012)	p	Work	0.087 (0.352)	–	0.024 (0.091)	–	–	–	–	–	Median (95th percentile), n = 209
Pedersen et al. (2016)	p	Work	0.110 (0.023–0.415)	–	0.016 (< LOQ – 0.045)	–	0.080 (0.013–0.292)	–	0.156 (0.021–0.398)	–	Median (range),n = 15, contaminated office building
Kraft et al. (2018)	wb	Work	–	12 (2.2–39)	–	–	–	–	–	–	Median (range),n = 35

s/p/wb: serum/plasma/whole blood; *assumes a density of serum of 1.024 g/ml (Sniegoski and Moody, 1979).

with the use of these two congeners as markers of dietary exposure.

Even with relatively short exposure time (1-2y) it seems from Fig. 2a/b and S1 that blood levels are elevated and thus an increase in serum levels occur relatively fast for the lower chlorinated congeners. This is similar to the findings by Meyer et al. (2013) though the minimum residence time was 2 years in that study. Due to the low number of samples from people living in the contaminated section for less than 5 years, it was not possible to show statistically that there are differences even after shorter exposure time.

4.2. Handwipes

Hand wipes samples are likely an intermediate between external exposure (e.g. air, dermal contact) and internal exposure (redistributed to skin lipids). In support of the external exposure Slayton et al. (1998) have shown that significant amounts of PCB can be picked up by wipes from PCB-contaminated material when mimicking skin contact. Weschler and Nazaroff (2012) have estimated that air to skin transfer is significant for PCB congeners such as PCB-28 and 52 and that dermal uptake from approximately equals that of inhalation. The contribution to hand wipes from the body's PCB-reservoir is not well described, but essentially PCB will distribute to the skin lipids and the isopropanol wipe may also extract the upper layer of the skin. In the current study both palm and back-of-hand samples were taken in order to try to separate the external exposure on palms (air + material contact) and back-of-hand (air only); unfortunately it was not possible to analyze all back-of-hand samples within the project. Unlike what has been observed for flame retardants and phthalates (e.g. Allen et al., 2013; Gong et al., 2014), there was no clear trend in the current study of higher levels on palms compared to back-of-hands, which may be due to the limited size of the current data set. Once on the skin, the dermal uptake of PCB would be in favor of the lower chlorinated congeners, which are taken up faster and to a greater extent than the higher chlorinated congeners (Garner and Matthews, 1998). The literature of PCB on hand wipes is scarce but the currently observed levels on palms are somewhat lower than observed on hand wipes from e-waste recycling workers in China (Qiao et al., 2019).

The hand wipe values for participants living together varied quite a lot. In some cases there was good agreement while for others the variation was larger, and the deviations could not be explained by time since last hand wash. One participant had washed hands approximately 15 min prior to sampling, thus a second hand wipe sample was taken 5 days later where it had been approximately 2 h since last hand wash. Despite that the first sample was to be disregarded both samples were shipped for analysis. The difference between the two samples were on average 4% for the individual congeners (all $< 30\%$). Though it is based on a single sample, combined with the data for participants sharing apartments it indicates stability of hand wipes as an exposure measure. The contrast between hand wipes from the reference and exposed group is more distinct than for serum and the hand wipes seem to give a fairly good indication of exposure level. Therefore hand wipes could possibly be used as a quick, non-invasive screening tool at least for ongoing indoor exposure to PCB, despite that it possibly represents a combination of internal and external exposure.

4.3. Serum vs. hand wipes

Significant correlations between serum and hand wipes were observed in the exposed group for almost all congeners, however the correlation coefficients were rather low. The Spearman correlations is a rather crude analysis without adjusting for age, residence time and contamination levels; nevertheless, this supports the use of hand wipes as a screening tool for ongoing indoor exposure. Significant correlations were observed even for persistent congeners such as PCB-28 and 153 though hand wipes were not expected to cover the accumulation in the body well. The correlation for PCB-28 ($p < 0.001$) could be partly explained by the indoor environment being the primary source; while the mechanism for the correlation for PCB-153 ($p < 0.001$) is unclear; perhaps the contribution from the internal exposure was not insignificant after all. Thus it remains unclear whether the hand wipes represent internal exposure or rather is a proxy for the ubiquitous exposure in contaminated buildings.

4.4. Strengths and weaknesses

One of the strengths of the study was that a large number of congeners particularly relevant for indoor exposure were analyzed and that so many different matrices were collected for further data analyses of relative importance of different exposure pathways. Furthermore, the construction of identical high-rises with and without PCB provided a very clear division and a large exposure contrast between the two exposure groups while almost all other parameters remained the same.

Not all 209 congeners and potential metabolites such as hydroxy-PCBs were analyzed; this could have provided even more information on exposure patterns and pathways and potentially identified more markers of recent exposure, though the main congeners are believed to be covered in the current selection. Finally, the reference group was rather small, this was chosen as the groups were very clearly divided and as there was expected to be a lot of non-detects in the non-human samples in the reference group. Furthermore, the population was too small to make reliable analyses of subgroups and the small group size also made the descriptive data (particularly 95th percentile) very sensitive to single reference participants with exposure to lower chlorinated PCBs elsewhere.

5. Conclusion

This study has confirmed that living in a PCB-contaminated building contributes significantly to the total body burden of PCB and that the indoor environment may even be the main source of PCB for people living in contaminated buildings. The congener pattern in both serum and hand wipes was clearly shifted towards lower chlorinated congeners for the exposed. The accumulation of lower chlorinated congeners were quite diverse, e.g. PCB-28 was observed to increase with residence time for decades while e.g. PCB-44 were elevated among the exposed group but without correlation with residence time, thus PCB-44 seem to be a good marker of recent exposure to original PCB-sources. PCB-levels on hand wipes were significantly elevated for the exposed group for all di-to pentaPCB measured, which suggests that hand wipes can be used as a screening tool for ongoing indoor exposure to PCB. Furthermore PCB on the hand wipes was significantly correlated with PCB in serum for most of the individual congeners.

Post script: Since the time of sampling four of the five high-rises have been vacated, in the last one sources have been covered and the building remain in use while demolition is being prepared.

Declaration of competing interest

The authors declare that they have no conflicts of interest.

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Appendix A. Supplementary data

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