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Plasma polychlorinated biphenyls in residents of 91 PCB-contaminated and 108 non-contaminated dwellings—An exposure study

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ABSTRACT

Background: In the1950s–1970s polychlorinated biphenyls (PCBs) were used in several countries as plasticizers in elastic sealants in buildings.
Objective: The primary objective was to study whether residents of PCB-contaminated dwellings had higher plasma levels of PCBs than their neighbours in non-contaminated dwellings. The secondary objective was to study possible associations between concentrations of PCBs in the indoor air and in the plasma of residents.
Methods: Stratified cross-sectional study of residents of a housing estate with four sections, of which only one section had PCB-containing sealants. The determination of 27 PCB congeners in plasma was performed among 134 exposed and 139 non-exposed residents. Air measurements were conducted in 104 flats.
Results: Significant differences in plasma PCBs between exposed versus non-exposed were found for most of the lower chlorinated and many of the higher chlorinated congeners. The median of sum of 27 PCBs was approximately four times higher in exposed compared with non-exposed residents. The elevated PCB concentrations persisted in multivariable analyses controlling for relevant cofactors. We found significant correlations between PCB indoor air concentrations and the PCB concentration in the plasma of the residents for ten of the lower chlorinated congeners.
Conclusion: Our study confirms that indoor air exposure to PCBs from PCB containing sealants may result in a considerable internal PCBs exposure of the residents. For the first time we were able to demonstrate that the internal exposure to low chlorinated PCBs is significantly associated with the indoor air concentration of these congeners.

Introduction

In the 1950s–1970s polychlorinated biphenyls (PCBs) were used in several countries as plasticizers in elastic sealants of public and residential buildings (Priha et al., 2005). Due to toxicological studies (Safe, 1994; Giesy and Kannan, 1998) and the experience gained from accidents at Yusho, Japan and Yu-Cheng, Taiwan (Chen et al., 1994; WHO, 1993), which revealed several severe health hazards caused by PCB exposure, the production and use of PCB were banned in most industrial countries from the end of the 1970s up to the end of the 1980s (ECC, 1985; USC, 1979). Nevertheless, even several decades after construction, the PCB-containing building materials still pollute the indoor air due to their high stability and low volatility (Heinzow et al., 2007; Herrick et al., 2004; MacIntosh et al., 2012; Priha et al., 2005).

Some studies investigated the concentrations of PCBs in indoor air or in indoor dust (Harrad et al., 2010; Heinzow et al., 2007; Kohler et al., 2005), and a few studies measured exposure of residents in contaminated buildings by biomonitoring (Gabrio et al., 2000; Johansson et al., 2003; Knobeloch et al., 2012; Liebl et al., 2004; Schettgen et al., 2012; Schwenk et al., 2002). These studies deal mainly with the PCB exposure of individuals who stayed in the buildings for work or education, showing weak relationship between indoor exposure to PCBs and levels in the blood. One study conducted in residences suggests however, that this correlation might be much stronger in home environments, probably due to longer time of exposure (Johansson et al., 2003). We had the opportunity to investigate the exposure of residents of

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a housing estate in the suburban area of Copenhagen where only some of the housing blocks were contaminated by PCB-containing sealants (Frederiksen et al., 2012). The aims of the study were to analyse the difference in the internal PCB exposure between the residents of PCB-contaminated and non-contaminated flats; secondly to assess the association between the PCB concentration in the indoor air and in the blood plasma of the residents and finally to further analyse cofactors for the internal exposure to PCBs.

Materials and methods

Design

This stratified cross-sectional study was performed on a housing estate, Farum Midtpunkt, constructed in stages between 1970 and 1974, 20 km north of Copenhagen, Denmark. The estate consists of 27 blocks with 1645 flats distributed on four sections, only one of which was PCB contaminated through the use of PCB-containing sealants. The constructions have been described in detail by Frederiksen et al. (2012).

Residents of approximately 200 inhabited contaminated flats and approximately 1300 non-contaminated flats were invited to participate in the investigation by letter. More than 100 contaminated flats were empty prior to renovation. Altogether 138 exposed and 151 non-exposed aged at least18 years accepted participation. Sixteen non-exposed were excluded since they had previously lived in contaminated flats. One of the non-exposed had moved after many years in a contaminated flat to a non-contaminated flat one month before examination, and was included as exposed. The final study population was 139 exposed from 91 flats, and 134 non-exposed from 108 flats.

Questionnaire

Completion of a questionnaire and blood sampling was performed in March 2011 on premises without PCB-contamination with the help of trained medical students. The questionnaire included questions on age, gender, years of living in the present flat, and years of living in other flats on the estate. Questions were also asked on estimated number of hours spent away from the flat on weekdays and during weekends, whether they slept with an open window in summer and winter, occupational exposure to PCB-containing building materials, smoking habits, and dietary habits with focus on the consumption of meat, fish, eggs and dairy products. Answer categories for the latter were “more than daily”, “5–7 times a week”, “3–4 times a week”, “once or twice a week”, “more seldom” or “never”. Female participants were queried about the number of children and the number of months each child had been breastfeed. No questions on health status were asked.

Plasma analyses

EDTA coated 9 ml tubes were used for blood sampling. After centrifugation at 800 × *g* for 10 min the plasma was transferred to 6 ml glass vials with an aluminium foil screw cap (Wheaton, Millville, NJ, USA), which were tested to be PCB free. After 1–2 days in the refrigerator, the plasma samples were frozen to −20 °C and kept at that temperature until analysis was performed in summer 2011.

The determination of 27 PCB congeners in plasma (congener 28, 52, 66, 74, 77, 81, 99, 101, 105, 114, 118, 123, 126, 138, 153, 156, 157, 167, 169, 170, 178, 180, 182, 183, 187, 189, and190) was performed using the isotope dilution GC–MS method. For all congeners analysed the relevant native standards were purchased from Ehrenstofer (Augsburg, Germany). ^{13}C-labelled compounds of 17 components were purchased from Cambridge Isotope Laboratories (CIL, Andover, USA) and were used for internal standardization of their native compounds as well as for compounds of similar chromatographic retention time and of the same grade of chlorination. The extraction of the analytes and the clean-up of the extracts were performed as described by Liebl et al. (2004). The PCBs were analysed by GC–MS system consisting of an Agilent 7890A GC and an Agilent 5975C MS (Santa Clara, USA) with inert ion source using electron ionization and selected ion monitoring. Separation of the PCBs was performed on a 60 m ZB-XLB fused silica column (0.25 mm ID, 0.25 μm film, Phenomenex, Aschaffenburg, Germany). For quantification, standard solutions in the range of 0.02–5.0 μg/l were applied in each series. For quality control bovine serum was spiked with a mixture of all PCBs and an aliquot of this solution was analysed in each series. The concentrations were 0.25 μg/l for PCBs 28, 52, 101, 138, 153 and 180 and approximately 0.05 μg/l for all other PCB congeners. The results of the quality control material were used to assure the comparability of the results from different series. The coefficients of variation (CV) for the precision between series (N = 25) were in the range of 1.53–9.91% for all PCB congeners with the exception of PCB 189, for which CV was 13.7%. The limit of quantification (LOQ) was 0.01 μg/l serum for each congener. The accuracy of the analyses was controlled by the successful participation in the proficiency tests of the German External Quality Assessment Scheme (G-EQUAS).

Air samples

Indoor air samples were collected from March to April 2011. In total, air from 83 contaminated and 21 non-contaminated flats was sampled. Twenty-four congeners were analysed including the 12 dioxin-like PCBs (DL-PCBs) and the 6 common indicator PCBs. Details on the sampling procedure and laboratory analyses can be found elsewhere (Frederiksen et al., 2012).

Statistics

The strategy for the statistical analyses was first to perform bivariate analysis of differences in the plasma levels of all congeners between exposed and non-exposed. Secondly the relation between PCB levels in the indoor air and the plasma was illustrated using scatter plots and Spearman correlations tests. Analyses of association between the air measurements and the plasma PCB were only done bivariately, since the data only were present for part of the study population. Multivariable analyses were performed to assess the relative importance of PCB exposure and other cofactors of PCBs in plasma. Associations between PCB exposure, the number of years in the flat and the plasma PCB concentrations were finally illustrated with stratified scatterplots with regression lines included.

The statistical analyses were performed in SPSS v. 6.1.2. Chi-square, Likelihood Ratio was used for 2 × 2 tables (i.e. exposed/non-exposed versus gender), and Mann–Whitney U in analyses of continuous variables (i.e. exposure status versus age). The multivariable analyses were performed with Multiple Linear Regression. Standardized regression residual histograms were produced for visual assessment of normal distribution of residuals. Factors of potential significance for example age, gender, body mass index, breastfeeding, dietary and smoking habits were tested in bivariate analyses with the outcome, as were exposure factors like years and daily hours in dwelling and indoor air exposure status. Factors that were significantly associated with the outcome in these analyses were included in the multivariable model. Backward elimination was performed manually for variables in the models with $p > 0.1$, though PCB indoor air exposure status was included in all models. As significance level 0.05 was chosen.

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Table 1
Background data on 139 participants exposed to PCB in their dwellings and on 134 non-exposed participants.

		Non-exposed (n = 134) Median (min.–max.) or %	Exposed (n = 139) Median (min.–max.) or %	p Mann–Whitney U
Gender	% women	62.7%	50.4%	0.04*
Age	Years	48 (18–89)	52 (19–90)	0.04
BMI	Bodyweight/height^2	25.4 (17.1–40.9) (n = 129)	24.7 (16.4–47.5) (n = 138)	0.77
Years in dwelling		6.5 (0.1–38)	12.0 (2.0–38.0)	0.005
Hours away from home on a week day		8 (0–15)	8 (0–21)	0.56
Cumulative months of breastfeeding		9 (0–86) (n = 84)	10 (0–41) (n = 70)	0.83
Smoking	% daily smokers	26.1%	35.3%	0.10*

* Chi-square, Likelihood Ratio.

Ethics

The participants were only exposed to their normal environment, and the only discomfort of the study was the blood sampling procedure. The project has gained acceptance from the Ethics Committee of the Copenhagen Region (protocol number H-4-2011-001). Written informed consent was obtained from all participants.

Results

Specifications of the study population are given in Table 1. The exposed group had a higher proportion of males, had lived longer in their dwellings, and were slightly older than non-exposed participants. No differences were found for body mass index (BMI), number of hours away from home on week days, months of breastfeeding, or smoking habits.

The PCB-concentrations in plasma are presented in Table 2. Significant differences between exposed versus non-exposed were found for most of the lower chlorinated congeners (PCB 28–PCB 123). Smaller but still statistically significant differences were found for many of the highly chlorinated congeners. Besides the differences in measured plasma concentrations, measurements under LOQ were more frequent among the non-exposed.

Table 3 presents the sums of different aggregations of PCBs in plasma. The medians of the sum of 6 indicator PCBs, the sum of 27 PCBs and the sum of non-DL congeners were between 3.4 and 4.1 times higher among the exposed than the non-exposed. The median of the sum of DL congeners among exposed was 1.9 times higher than non-exposed, which was still statistically significant. When separating by number of chlorine atoms, the median of the sum of congeners with 3–4 atoms (tri/tetra PCBs) was 51.6 times higher among the exposed compared with non-exposed, falling to factor 4.2 for the sum of congeners with 5 chlorine atoms (penta PCBs), and further to factor 1.2 for the highly chlorinated congeners with 6–7 chlorine atoms (hexa/hepta PCBs).

A summary of the measured PCB concentrations in the air of the apartments is presented in Table 4. Especially for the lower chlorinated congeners with relatively high vapour pressure there is a

Table 2
Concentration of PCB in plasma (μg/l) according to exposure status, for 27 congeners. Statistics: Mann–Whitney U. <LOQ set to ½ × LOQ in calculations (0.005 μg/l).

PCB-congener	Non-exposed (n = 134)		Exposed (n = 139)		p
	Median (5–95 percentiles) (μg/l)	% >LOQ	Median (5–95 percentiles) (μg/l)	% >LOQ	
PCB 28[b]	0.014 (<LOQ–0.132)	65.7	1.371 (0.216–5.279)	100	<0.0001
PCB 52[b]	<LOQ (<LOQ–0.018)	12.7	0.216 (0.066–1.218)	99.3	<0.0001
PCB 66[b]	<LOQ (<LOQ–0.057)	34.3	0.562 (0.103–2.502)	100.0	<0.0001
PCB 74[b]	0.031 (<LOQ–0.162)	94.0	1.101 (0.108–5.139)	100.0	<0.0001
PCB 77[a,b]	<LOQ (<LOQ–<LOQ)	0.0	<LOQ (<LOQ–<LOQ)	0.0	–
PCB 81[a,b]	<LOQ (<LOQ–<LOQ)	0.0	<LOQ (<LOQ–<LOQ)	0.0	–
PCB 99[c]	0.024 (<LOQ–0.070)	81.3	0.145 (0.029–0.664)	99.3	<0.0001
PCB 101[c]	<LOQ (<LOQ–<LOQ) <LOQ)	3.7	0.034 (<LOQ–0.145)	95.0	<0.0001
PCB 105[a,c]	<LOQ (<LOQ–0.020)	28.4	0.030 (<LOQ–0.188)	87.8	<0.0001
PCB 114[a,c]	<LOQ (<LOQ–0.020)	0.7	<LOQ (<LOQ–0.031)	37.4	<0.0001
PCB 118[a,c]	0.033 (<LOQ–0.121)	91.0	0.101 (0.021–0.398)	100	<0.0001
PCB 123[a,c]	<LOQ (<LOQ–<LOQ)	0.7	<LOQ (<LOQ–0.034)	38.8	<0.0001
PCB 126[a,c]	<LOQ (<LOQ–<LOQ)	0.0	<LOQ (<LOQ–<LOQ)	0.0	–
PCB 138[d]	0.134 (0.019–0.462)	100	0.157 (0.039–0.557)	100	0.03
PCB 153[d]	0.346 (0.041–1.143)	100	0.392 (0.089–1.295)	100	0.02
PCB 156[a,d]	0.035 (<LOQ–0.121)	79.1	0.046 (<LOQ–0.137)	88.5	0.03
PCB 157[a,d]	<LOQ (<LOQ–0.022)	33.6	<LOQ (<LOQ–0.028)	45.3	0.04
PCB 167[a,d]	<LOQ (<LOQ–0.033)	47.0	<LOQ (<LOQ–0.044)	47.5	0.40
PCB 169[a,d]	<LOQ (<LOQ–<LOQ)	0.0	<LOQ (<LOQ–<LOQ)	1.4	–
PCB 170[d]	0.111 (<LOQ–0.351)	94.0	0.121 (0.020–0.369)	99.3	0.03
PCB 178[d]	0.018 (<LOQ–0.063)	68.7	0.027 (<LOQ–0.068)	79.9	0.02
PCB 180[d]	0.262 (0.020–0.858)	100	0.341 (0.048–0.943)	100	0.01
PCB 182[d]	<LOQ (<LOQ–<LOQ)	0.0	<LOQ (<LOQ–<LOQ)	0.0	–
PCB 183[d]	0.022 (<LOQ–0.073)	74.6	0.023 (<LOQ–0.084)	85.6	0.12
PCB 187[d]	0.067 (<LOQ–0.198)	88.8	0.080 (0.016–0.254)	99.3	0.008
PCB 189[a,d]	<LOQ (<LOQ–0.019)	26.9	<LOQ (<LOQ–0.021)	38.1	0.06
PCB 190[d]	0.016 (<LOQ–0.050)	64.2	0.019 (<LOQ–0.053)	69.8	0.15

LOQ, limit of quantification.
[a] Dioxin-like PCB.
[b] Tri-tetra chlorinated.
[c] Penta chlorinated.
[d] Hexa–hepta chlorinated.

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Table 3
Concentration of PCB in plasma (μg/l) in differently aggregated groups according to exposure status. Statistics: Mann–Whitney *U* test.

PCB-aggregations	Non-exposed ($n = 134$)		Exposed ($n = 139$)		p
	Median (5–95 percentiles) (μg/l)	Mean (μg/l)	Median (5–95 percentiles) (μg/l)	Mean (μg/l)	
Sum 6 indicator[a]	0.805 (0.118–2.508)	0.954	2.715 (0.754–8.571)	3.454	<0.0001
Sum 27	1.256 (0.260–3.750)	1.520	4.902 (1.186–17.279)	6.816	<0.0001
Sum DL[b]	0.122 (0.065–0.357)	0.154	0.237 (0.088–0.760)	0.310	<0.0001
Sum_NDL[c]	1.138 (0.196–3.402)	1.366	4.700 (1.086–16.466)	6.505	<0.0001
Sum of tri/tetra chlorinated PCBs[d]	0.068 (0.036–0.388)	0.160	3.507 (0.549–13.28)	4.782	<0.0001
Sum of penta chlorinated PCBs[e]	0.080 (0.035–0.219)	0.103	0.337 (0.080–1.374)	0.467	<0.0001
Sum of hexa/hepta chlorinated PCBs[f]	1.052 (0.134–3.365)	1.253	1.268 (0.275–3.808)	1.562	0.02

[a] Sum of the 6 indicator congeners: PCB 28, 52, 101, 138, 153, 180.
[b] Sum of 12 dioxin-like PCB: PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189.
[c] Sum of 15 non-dioxin-like PCB: PCB 28, 52, 66, 74, 99, 101, 138, 153, 170, 178, 180, 182, 183, 187, 190.
[d] Sum of tri-tetra chlorinated: PCB 28, 52, 66, 74, 77, 81.
[e] Sum of penta chlorinated: PCB 99, 101, 105, 114, 118, 123, 126.
[f] Sum of hexa–hepta chlorinated: PCB 138, 153, 156, 157, 167, 169, 170, 178, 180, 182, 183, 187, 189, 190.

Table 4
Air concentrations of PCB in dwellings (ng/m^3). 6 indicator congeners, PCB 118 and sums of PCB congeners. Statistics: Mann–Whitney *U*. <LOQ set to zero in calculations.

PCB-congener	Non-contaminated ($n = 21$)		Contaminated ($n = 83$)		p
	Median (min.–max.) (ng/m^3)	Mean (ng/m^3)	Median (min.–max.) (ng/m^3)	Mean (ng/m^3)	
PCB 28	<LOQ (<LOQ–19.8)	1.31	61.4 (14.9–296)	80.8	<0.001
PCB 52	<LOQ (<LOQ–28.3)	2.13	94.6 (16.5–426)	112	<0.001
PCB 101	<LOQ (<LOQ–2.64)	0.13	8.90 (1.78–47.1)	12.9	<0.001
PCB 118[d]	<LOQ (<LOQ–<LOQ)	<LOQ	1.18 (<LOQ–9.09)	1.75	<0.001
PCB 138	<LOQ (<LOQ–<LOQ)	<LOQ	<LOQ (<LOQ–<LOQ)	<LOQ	–
PCB 153	<LOQ (<LOQ–<LOQ)	<LOQ	<LOQ (<LOQ–1.48)	0.03	0.48
PCB 180	<LOQ (<LOQ–<LOQ)	<LOQ	<LOQ (<LOQ–<LOQ)	<LOQ	–
Sum 24	0.11 (<LOQ–64.5)	4.80	236 (43.3–1060)	285	<0.001
Sum_DL[a]	<LOQ (<LOQ–0.38)	0.02	2.19 (0.18–16.5)	3.21	<0.001
Sum_NDL[b]	0.11 (<LOQ–64.1)	4.78	234 (43.1–1043)	282	<0.001
Total PCB[c]	<LOQ (<LOQ–254)	17.8	859 (168–3843)	1030	<0.001

LOQ, limit of quantification.
[a] Sum of 12 dioxin-like PCB: PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189.
[b] Sum of 12 non-dioxin-like PCB: PCB 28, 52, 66, 74, 99, 101, 138, 153, 170, 180, 183, 187.
[c] Total PCB calculated as sum of 6 indicator congeners × 5 (PCB 28, 52, 101, 138, 153 and 180).
[d] Dioxin-like PCB.

clear pattern of significantly higher concentrations in the contaminated apartments. For numbers 138 and 180 the levels were under the detection limits in all apartments. Also for the sum variables the differences between contaminated and non-contaminated air samples were orders of magnitude. More details of the air measurements have been presented recently (Frederiksen et al., 2012).

We found significant correlations between PCB air concentration in the flats and the PCB concentration in the plasma of their residents for PCBs 28, 52, 66, 74, 99, 101, 105, 114, 118 and 123 (each $p < 0.0001$). However, we found decreasing correlation coefficients with increasing chlorination. Fig. 1 shows the significant correlation between PCB 28 in air and plasma. No significant correlation was found for PCBs 153, 183 and 187. The remaining congeners were under the limit of quantification either in air or in plasma.

Three models of multiple linear regressions are presented in Table 5: sum of tri/tetra chlorinated congeners (model A), sum of penta chlorinated congeners (model B), and the sum of hexa/hepta chlorinated congeners (model C).

For model A the three strongest risk factors for high plasma concentrations were related to the indoor air exposure: participants living in exposed flats had 4.1 times higher plasma concentrations than non-exposed. For each 5 year interval of living in contaminated flats, the concentration increased with 63%. An inverse association was found for hours away from the dwelling on week days (43% decrease per 2 h interval).

All the above factors were also significant in model B, but with lower regression coefficients. In addition age and gender stayed in the model showing a 3% higher concentration per 10-year interval of age, and 6% higher level for male participants. Living in a PCB-contaminated flat was associated with a 30% higher concentration in plasma.

In model C, none of the flat exposure variables were associated with the outcome. Positive associations were found for age (52% increase per 10 years). Breastfeeding was associated with decreased plasma PCB concentrations, 1% per month.

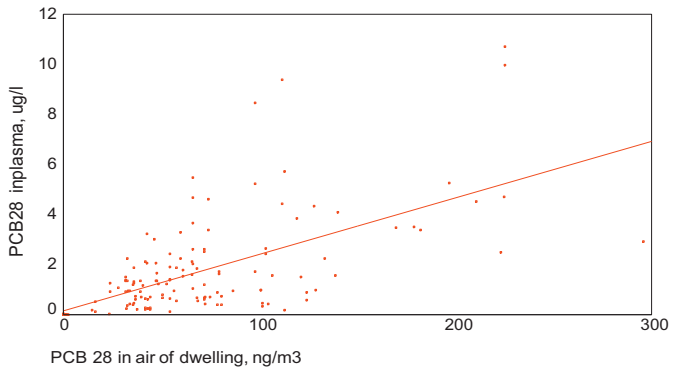

Fig. 1. Scatter plot of air and plasma concentrations of PCB 28. $N = 154$; Spearman correlation coefficient = 0.69; $p < 0.001$. The formula for the fit line based on Robust Linear Regression: $Y_i = 0.156 + 0.023X_i + e_i$.

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Table 5
Multiple linear regressions models. Dependent variables are sums of PCBs in plasma, according to number of chlorine atoms. Regressions coefficients and 95% confidence intervals (CI) and *p*-values are presented.

Co-variables	Dependent variables	
	Regression coefficient (95% CI)	*p*
Model A: sum of tri/tetra chlorinated congeners (# 28–81)		
Exposed to PCBs in dwelling	4.13 (3.46–4.81)	<0.0001
Years in dwelling (per 5 year interval)[a]	0.63 (0.44–0.83)	<0.0001
Hours away from dwelling per week-day (per 2 h interval)	−0.43 (−0.64 to −0.22)	0.0001
Frequency of cheese consumption (per interval out of six)	−0.37 (−0.61 to −0.13)	0.003
Frequency of consumption of fish as warm meal (per interval out of six)	0.52 (0.10–0.94)	0.02
Model B: sum of penta chlorinated congeners (# 99–126)		
Exposed to PCBs in dwelling	0.30 (0.25–0.36)	<0.0001
Years in dwelling (per 5 year interval)[a]	0.06 (0.04–0.08)	<0.0001
Frequency of cheese consumption (per interval out of six)	−0.03 (−0.05 to −0.01)	0.003
Hours away from dwelling per week-day (per 2 h interval)	−0.03 (−0.05 to −0.008)	0.006
Frequency of consumption of fish as warm meal (per interval out of six)	0.05 (0.009–0.08)	0.01
Age (per 10 years interval)[b]	0.03 (0.001–0.05)	0.04
Gender (male)	0.06 (0.0002–0.12)	0.049
Model C: sum of hexa/hepta chlorinated congeners (# 138–190)		
Exposed to PCBs in dwelling	0.08 (−0.11 to 0.26)	0.41
Age (per 10 years interval)[b]	0.52 (0.45–0.58)	<0.0001
Months of breastfeeding (per month)	−0.01 (−0.02 to −0.008)	0.001
Frequency of consumption of fish as cold dish (per interval out of six)	0.10 (0.03–0.18)	0.008

[a] For exposed: sum of years of living in exposed dwellings in Farum Midtpunkt.
[b] The first interval covers the age span 18–29 years.

Associations between the plasma concentration of low, medium and highly chlorinated PCBs and the years living in contaminated and uncontaminated flats respectively, are shown in the three graphs of Figs. 2 and 3. Influence of time in the dwelling was clearly different for exposed and non-exposed with respect to the low and medium chlorinated PCBs. For the sum of hexa/hepta PCBs, the regression lines increased with number of years in an almost identical manner for both exposed and unexposed.

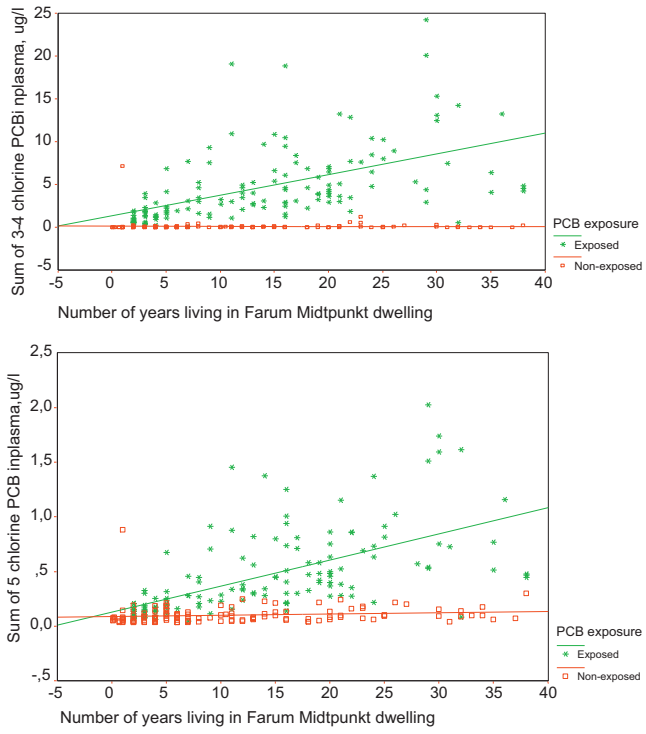

Fig. 2. Relations of sums of low (tri/tetra) and medium (penta) chlorinated PCBs in plasma, versus number of years living in exposed or non-exposed flats. Please note the different units on the *Y*-axes.

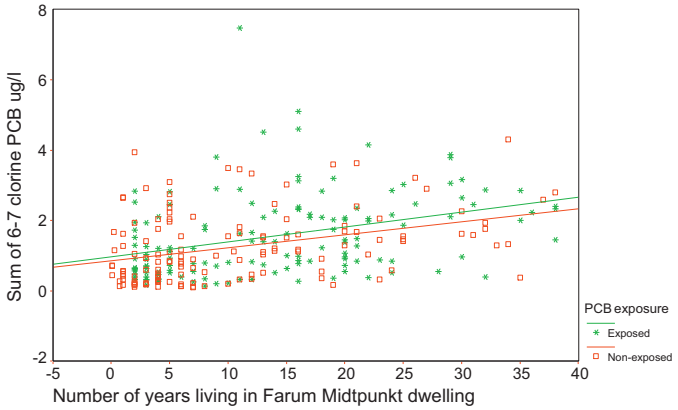

Fig. 3. Relations of sums of highly (hexa/hepta) chlorinated PCBs in plasma, versus number of years living in exposed or non-exposed flats.

Discussion

General findings

As to the primary aim of this study, we found a clear pattern of significantly higher PCB plasma concentrations in the residents of PCB contaminated flats. This was especially the case for the lower chlorinated congeners. The median sum of tri/tetra chlorinated congeners was 52 times higher among the exposed compared to the non-exposed. This effect was higher than reported in the literature. Johansson et al. (2003) found a ratio of 17 for the exposed versus non-exposed (sum of PCBs 28, 66 and 74), and for PCB 28 the ratio was 30 (measured as median ng/g fat in blood). Separating on DL and NDL PCBs, they also found smaller differences, ratio 2 for NDL versus our ratio 4, and ratio 1.2 for DL PCBs versus our ratio 2. However, proper comparative analysis of these two studies is not feasible, since no data are available on the PCB exposure in air of the buildings or on the mean residential time in the study of Johansson et al. (2003).

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The clear associations between exposure status and plasma concentrations of PCBs found in the present study remained significant after controlling for potential confounders in the multivariable analyses. As illustrated in Table 5, the major factors of exposure for high chlorinated congeners shifted from indoor air to exposure via the diet. Moreover age became more important for the internal exposure to higher chlorinated congeners.

Months of breastfeeding was inversely associated with plasma concentration, indicating that the female participants had reduced their body content of PCB by passing it on to their babies, as shown by Abraham et al. (1998) among others. This effect is probably also one of the reasons for the gender difference found for sum of medium chlorinated PCBs 99–126, where men had higher levels than women. Other reasons for the gender differences may be the different volumes of distribution in the body due to different body fat content, and differences in intake of caloric foodstuffs.

The age effect is likely also to be connected with the dietary intake of PCB. Looking at the non-exposed in the three graphs of Figs. 2 and 3, the two horizontal curves for sums of low and medium chlorinated PCBs illustrate no association with year in dwelling (and to some extent age and thereby cumulative PCB intake via the diet). Quite the opposite is seen in the third graph (highly chlorinated PCBs) where both exposed and non-exposed show an almost identical increase of plasma concentration by duration of occupancy.

The association between the blood or plasma concentration of highly chlorinated PCB congeners and age is well known and was associated in several studies with the accumulation of the dietary intake of these PCBs (Becker et al., 2002; Bates et al., 2004; Schettgen et al., 2011; Vaclavik et al., 2006).

Comparable studies of plasma PCB background levels

We found only two studies on the PCB exposure of the Danish population when excluding Greenland and the Faroe Islands, which had been investigated due to the traditional diet with seal and whale (e.g. Deutch et al., 2004; Fängström et al., 2002). In late 1970s Høyer et al. (2000) measured PCB concentrations in serum from 429 women with the mean age of 54.6 years. Median concentration of the sum of PCBs 118 + 138 + 153 + 180 was 1101.5 ng/g lipid. With a rough estimate of a lipid fraction in serum of 0.6% that equals 6.6 μg/l serum. Five years later the median concentration in the same population had decreased to 5.9 μg/l. For comparison, the median concentration of the same PCB sum in the 84 non-exposed women in the present study (mean age 49.1 years) was 0.81 μg/l while it was 0.84 μg/l in the 70 exposed women (mean age 52.6 years). Halldorsson et al. (2008) measured plasma PCBs in 100 nulliparous women aged 25–35 years from the Danish National Birth Cohort in 1998–2002. Median concentration of the sum of the same four PCB congeners plus PCB 105 and 156 was 1.15 μg/l. This illustrates that the body burden of PCB in women 35 years ago as presented by Høyer et al. was much higher, which was also described in studies of other human tissues (Jensen, 1987; Vaclavik et al., 2006).

In the Netherlands Koopman-Esseboom et al. (1994) collected plasma samples from 108 pregnant women in Rotterdam and 209 in the more rural Groningen in the period 1990–1992. The sum of means of PCB 118 + 138 + 153 + 180 was 2.18 ng/g plasma and 2.17 ng/g plasma respectively.

In Germany, the Human Biomonitoring Commission has published reference values as an indication of the upper margin of the background level of PCBs in blood, based on studies from 1998 to 2002 (Schulz et al., 2007). The reference value for PCB 138 in the age span 50–59 years is 1.7 μg/l compared with the 95% percentile of the exposed in our study of 0.56 μg/l. The corresponding values for PCBs 153 and 180 were 2.8 μg/l versus 1.30 μg/l, and 2.1 μg/l versus 0.94. In 2011 the German Commission for the Investigation of Health Hazards of Chemical compounds in the Work Area estimated upper reference values for the background levels of low-chlorinated PCB for persons of working age who are not occupationally exposed to PCBs. These reference values are 0.02 μg/l plasma for PCB 28, and <0.01 μg/l plasma for the PCB 52 and 101 (Göen et al., 2012). In a recent study Schettgen et al. (2011) presented a study of plasma concentrations of PCBs in 105 people from the general German population. Compared with the non-exposed of our study, the plasma concentrations were almost identical for the DL PCBs. The same was true of the three lowest chlorinated of the indicator PCBs 28, 52 and 101, whereas the German values were 2.6–4.6-fold higher for PCBs 138, 153 and 180. All in all, the PCB concentrations of the non-exposed participants in our study are similar to the plasma concentrations in the general German population.

Indoor exposure and PCB blood levels

Earlier studies with assessment of the PCBs in the blood in residents do not present comparable air measurements between contaminated and non-contaminated dwellings. Fitzgerald et al. (2011) investigated PCBs in indoor air, where the PCB source was a neighbouring capacitor plant that was active from 1945 to 1977. Even though indoor air concentrations were relatively low (mean sum of 12 PCBs was only 14 ng/m^3), they found significant associations between the concentration in the serum of the residents and indoor air levels for congeners 28 and 105. Orloff and colleagues (2003) studied 18 homes with 78 residents near a chemical plant that formerly produced PCBs (Alabama, USA). They found no correlations between PCBs in house dust and blood concentrations.

Gabrio et al. (2000) found mean air concentrations of PCB_{tot} (6 indicator PCBs × 5) in two schools much higher than in our study (3541 ng/m^3 and 7490 ng/m^3). A few years later researchers from the same group presented air measurements from a contaminated school with even higher concentrations of total PCBs beyond 12,000 ng/m^3 (Schwenk et al., 2002). The plasma values of PCBs in that study, even though elevated 2–8-fold for exposed compared with non-exposed, were somewhat lower than in the present study: mean PCB 28 was 0.24 μg/l compared with our median of 1.37 μg/l, mean PCB 52 of 0.07 μg/l compared with 0.22 μg/l, and mean PCB 101 of 0.02 compared with 0.03 μg/l. Plasma concentration in the study by Gabrio et al. (2000) as well as in another German study comparing 377 pupils from a contaminated school, with 218 pupils from a non-contaminated school (Liebl et al., 2004) were similar to Schwenk et al. (2002). Furthermore, Schwenk et al. (2002) found elevated levels of PCBs 28 and 52 among 18 teachers from a highly contaminated school compared with 11 teachers from a control school. Schettgen et al. (2012) examined 209 persons employed in a contaminated public university building and 98 matched controls. They found significant differences in plasma levels for the lower chlorinated PCBs 28, 52, and 101, and for the DL-PCB 105 and 118. However the differences were much smaller than in the present study. When comparing concentrations in dwellings and public institutions, it must be kept in mind, that people spend far more time at home than in school or at work. This is probably the explanation why the higher air concentrations in German schools are not as clearly reflected in the blood values like in the present study.

Strengths and weaknesses of the study

The present study is the first on residential buildings where design and sample size allow thorough multivariable analyses to control for potential confounders. Moreover, performing this study in Farum Midtpunkt, where approximately 1/5 of the flats are built

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with PCBs in the sealants, provided us with a unique design of two groups of people with theoretically equal background, but different PCB exposures.

The main weakness of the study was that the participants were not included after a random sampling, due to no access to a list of residents. Instead the principle first come, first served was applied, which may have resulted in selection bias, because certain groups felt a stronger motivation to turn up, i.e. people with health problems. Since this study was strictly an exposure study, the questionnaire contained no questions about health; and therefore we have no impression of the size of this possible bias. We are convinced though, that this potential bias is not a problem, since even the participant with the highest plasma concentrations of PCBs is well below the lowest effect level found in animal studies. We did not have information about socio-economic status, and a clustering of certain social classes in some of the blocks cannot be ruled out. However, the monthly rent per square metre of the flat, and the size of the flats were equal in the contaminated and non-contaminated blocks.

Conclusions

Our study confirms that indoor air exposure to polychlorinated biphenyls (PCBs) by the emissions of PCB containing sealants may result in a considerable internal PCBs exposure of the residents. This was true for both the lower and many of the higher chlorinated congeners. The significant associations between indoor air PCB exposure and sums of low and medium chlorinated PCBs in plasma persisted in multivariable analyses controlling for potential confounders such as age, gender, dietary habits, breastfeeding and body mass index.

For the first time we were able to demonstrate that the internal exposure to low chlorinated PCBs is significantly associated with the indoor air concentration of these congeners. Furthermore, number of years in the flat was associated with higher plasma values of lower and medium chlorinated PCBs among the exposed only. Finally, our results indicate that the monitoring of the internal exposure to low chlorinated PCBs is valuable for the risk assessment of individuals, who were exposed by PCB-polluted indoor air.

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