

PCB contamination from polysulphide sealants in residential areas—exposure and risk assessment

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Abstract

From the late 1950s to the early 1970s elastic polysulphide sealants were used in outdoor seams between concrete blocks in prefabricated buildings. The sealants contained 5–30% polychlorinated biphenyls (PCBs). Due to the weathering of sealants in general and the replacement of seams with new PCB-free materials in the 1990s, PCBs have drifted to the soil adjacent to buildings. The objectives of this study were to survey PCB contamination in the surroundings of former PCB-containing buildings and to evaluate the risks to human health. Samples from soil, and also from blood serum of residents, were collected to obtain data for exposure assessment. The health risk assessment was based on deterministic and probabilistic calculations for cancer and non-cancer risks. Soil ingestion and dermal contact were considered the main routes of exposure and children the most important exposed group.

The mean total PCB concentration was 6.83 mg/kg within 2 m of the buildings and 0.52 mg/kg within 3–10 m from the buildings. The deterministic risk assessment with conservative parameters resulted in lifetime cancer risk estimates on the order of 10^{-6} – 10^{-7} . The lifetime average daily dose (LADD) for PCBs was less than 10% of the reference dose (RfD) 0.02 µg/kg day, which is based on immunosuppression in monkeys. The LADD corresponding to the total site attributable exposure was less than 10% of the estimated average dietary PCB intake in Finland. Children can, however, in worst cases be exposed to daily doses near the level of the RfD. Low cost measures are recommended to reduce possible exposure of children.

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1. Introduction

A new source of polychlorinated biphenyl (PCB) contamination, elastic polysulphide sealants of prefabri-

cated residential buildings, was identified in the 1990s in Germany, Sweden and Finland (Benthe et al., 1992; Jansson et al., 1997; Pyy and Lyly, 1998). The elastic polysulphide sealants used in 1960–1975 contained 5–30% PCB as a plasticizer. PCB-containing sealants were mainly used in the outdoor seams of concrete buildings, and it was discovered that the PCBs in the sealants outdoors spread to the environment through erosion and the removal of old sealants.

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In Germany, PCB-containing sealants were used more often in indoor seams and thus were considered mainly an indoor air problem (Benthe et al., 1992; Gabrio et al., 2000). In Finland, however, PCBs were mainly used in outdoor seams between concrete blocks. Estimates of the volume of PCBs used in sealants vary from 130 t to 270 t (Haukijärvi and Pentti, 2000). The most frequently applied commercial mixture was Aroclor 1260 or Aroclor 1254. A large block of flats can contain as much as 40–50 kg of PCB (Jansson et al., 1997; Haukijärvi and Pentti, 2000).

Polychlorinated biphenyls have mainly long-term effects on humans. Although the acute toxicity of PCB mixtures is low, they are expected to increase cancer risks in humans and have neurodevelopmental and hormonal effects. They can also cause immunosuppression. Dermal manifestations such as chloracne have been noted in cases of high exposure. Immunosuppression occurs in monkeys already at an exposure level of 5 µg/kg/day (Tryphonas et al., 1991), and this effect has been used as a basis for exposure standards. On the basis of this finding and a safety factor of 300 the United States Environmental Protection Agency (US EPA) has issued a dose of 0.02 µg/kg/day as a reference (RfD) for the chronic acceptable daily intake of PCBs (RAIS, 2003). The subchronic oral RfD is 0.05 µg/kg/day (RAIS, 2003).

The recommended limit value for soil remediation is 0.5 mg PCB/kg soil in Finland (Puolanne et al., 1994), but according to some earlier Scandinavian studies this value is sometimes exceeded (Jansson et al., 1997; Pyy and Lyly, 1998; Hellman, 2000). PCBs can contaminate soil in the vicinity of old concrete buildings and thereby create a risk to residents and the environment.

The objectives of this research project were to study soil contamination in the yards of PCB-containing buildings and to evaluate the possible consequential health risks to residents.

2. Materials and methods

2.1. Study areas and soil sampling

We studied 11 buildings that were built in 1960s and had undergone sealant replacement within 1–3 years of our study. The yards of the buildings were partly covered with asphalt, grass or sand. The distance to the nearest aquifer was, in every case, more than 1 km. In order to study the distribution of PCBs, we took soil samples by applying the systematic grid sampling method. All the cardinal points were sampled separately. The sampling distances were 0.1 m, 0.5 m, 1 m, 2 m, 3 m, 5 m, and 10 m from the buildings. Composite samples were taken from children's playgrounds and sandpits near the buildings.

Glass jars (300 ml) were used as containers. They were covered with aluminium foil and closed tightly with a lid. The sampler (shovel) was cleaned with distilled water and dried with a paper towel between each sampling procedure.

2.2. PCB analyses

Samples were roughly homogenized by mixing with a rinsed steel spatula after being passed through a 6 mm sieve. About 2 g of soil was weighed accurately in a 15-ml screw-capped test tube, and 3 ml of hexane–acetone mixture (1:1) was added. The tubes were vortexed and ultrasonicated for 1 h. Thereafter the soil samples were centrifuged for about 1 min, and the supernatant was removed. This extraction procedure was repeated once, and the supernatants were combined. Acetone was removed by distilled water (2 × 3 ml). The hexane phase was purified by Florisil Norma Phase columns (Supelco Ltd.) according to the instructions of the manufacturer. Samples were concentrated by nitrogen flux before GC–MS analysis.

PCBs were analysed by a gas chromatograph (HP 6890 Series) equipped with a mass selective detector (HP 5971A) and a HP-5MS capillary column (length 30 m, diameter 250 µm, film thickness 0.25 µm). The carrier gas (helium) flow was 1 ml/min. The oven temperature programme was as follows: 80 °C (1 min), 15 °C/min to 150 °C, 15 °C/min to 250 °C, 15 °C/min to 315 °C (1 min). Fifteen different PCB congeners were used in the quantitative analysis (PCB 28, 31, 52, 77, 101, 105, 118, 126, 128, 138, 153, 156, 169, 170, 180). Because the peak pattern resembled that of Aroclor 1260, the total PCB concentration was calculated by comparing the sum of the respective peaks in the samples and in an Aroclor 1260 standard (Schulz et al., 1989). The detection limit for the PCBs in soil was about 0.01 mg/kg. The calculated overall accuracy of the method was about 20% for PCB mixtures (Mäkinen et al., 2001). The soil PCB results, used for the risk assessment, are expressed in mg per kg soil wet weight, if not otherwise mentioned.

In October 2001 the Finnish Environment Institute carried out an interlaboratory comparison test (Mäkinen et al., 2001) on methods used for analysing PCBs in contaminated soils. The method described in this report was also evaluated, and the results proved to be acceptable (*z*-value 0.453).

2.3. Health risk assessment

We estimated residents' exposure to PCB and the consequent health risk by using the equations originally presented by the US EPA (1989). The lifetime average daily dose (LADD) was calculated for each exposure route, and incremental cancer risk was further defined

by multiplying the LADD by the cancer slope factor (CSF) 2 per mg/kg (US EPA, 1996). Non-carcinogenic risks were assessed by comparing exposure with the EPA reference dose (RfD) of 0.02 µg/kg/day for Aroclor 1254 (RAIS, 2003).

In selecting the parameters for the exposure calculations, we primarily followed the recommendations presented by the US EPA (Table 1). The time of exposure was, however, based on Finnish climatic conditions (i.e., exposure to soil only 180 days/year).

We concluded that the ingestion of PCB-contaminated soil and dermal contact with soil are the major exposure routes. According to our measurements, the PCB levels in the outdoor air were low (concentrations < 0.05 µg/m³). We expected this to be the case since the vapour pressure of highly chlorinated PCB mixtures (mainly Aroclor 1260) is very low (about 1% of that of Aroclor 1242). We focused on children from 1 to 6 years of age for the risk assessment since this group is the most susceptible to hand-to-mouth behaviour. The deterministic exposure and dose–response models presented by the US EPA (1989, 1997) were adopted for the assessment of excess lifetime cancer risks. The exposure factors were adjusted to Finnish climatic conditions.

The probabilistic approach was used to study both the uncertainties and the distribution of the risk estimates. Distribution curves were generated by commercial software (Crystal Ball™ with Monte Carlo simulation). Our sampling size was 10000. All the parameter values used in the calculations are presented in Table 1.

Aroclor 1260 contains about 4–5% of coplanar PCBs (IPCS/WHO, 1993; Van den Berg et al., 1998). In order to consider congener-specific data and dioxin-like compounds, we estimated the toxicity mainly by using a conversion factor of 11.3 (ng TEQ/mg product) for Aroclor 1260 (Environment Canada, 1998).

2.4. PCB levels in blood serum

We also studied the PCB concentrations in the blood serum of 24 residents of buildings containing PCB sealants. Residents ($n = 27$) in non-PCB buildings served as a control group. The age of the participants varied between 30 and 70 years (average = 58 years) for the PCB-building residents and between 30 and 61 years (average = 48 years) for the control group. We restricted the study to those who had lived in a PCB building for at least the last 5 years. The work history of each inhabitant was registered so that we could exclude persons with possible occupational exposure.

Samples were centrifuged, frozen immediately and kept frozen (–20 °C) until the analysis. The Biomonitoring Laboratory of the Finnish Institute of Occupational Health was in charge of the analyses. The following PCB congeners were determined: PCB 28, 52, 77, 101, 118, 126, 138, 153, 169 and 180. Serum (2 ml) was extracted with hexane-diethyl ether. The extract was purified with sulphuric acid and column chromatography. Samples were analysed with a gas chromatograph equipped with mass selective detector (negative chemical ionization). Methane was used as the reagent gas, and a PAS-1701 capillary column (30 m, 250 µm, 0.25 µm) was used. Details of the method have been described in an earlier paper (Kontsas and Pekari, 2003).

3. Results

3.1. PCB concentrations in soil

The total PCB concentrations in our soil samples ranged from 0.11 mg/kg to 26.9 mg/kg (w.w.). The PCB concentrations were highest in areas adjacent to the buildings, and they declined as the distance increased

Table 1
Calculation parameters used in the health risk assessment

Parameter	Unit	Point estimate	Distribution	Type	Source
CS	mg/kg	6.83	$\mu = 7.42, \sigma = 14.64$	LN	Our data
IR	mg/day	100	45 (50%), 208 (95%)	LN	US EPA (1997), Lijzen et al. (2001)
SA	cm ²	2194	1799 (50%), 2194 (95%)	LN	US EPA (1997)
AF	mg/cm ²	0.52	$\mu = 0.52, \sigma = 0.90$	LN	Finley et al. (1994)
BW	kg	21.3	$\mu = 16.6, \sigma = 2.45$	LN	US EPA (1997)
ED	a	5	min = 4, max = 5	U	1–6 a
EF	d/a	180	min = 60, max = 180	U	Own judgment
ABS	–	0.06		U	US EPA (1992)
CSF	(mg/kg day) ^{–1}	2			US EPA (1996)
AT	day	70 × 365 Cancer risk			US EPA (1989)
	day	5 × 365 Children			US EPA (1989)

CS = concentration of soil, IR = intake rate, SA = skin area, AF = adherence factor, BW = body weight, ED = exposure duration; EF = exposure frequency, ABS = dermal absorption factor, CSF = cancer slope factor, AT = averaging time, μ = average, σ = variance, LN = lognormal, U = uniform.

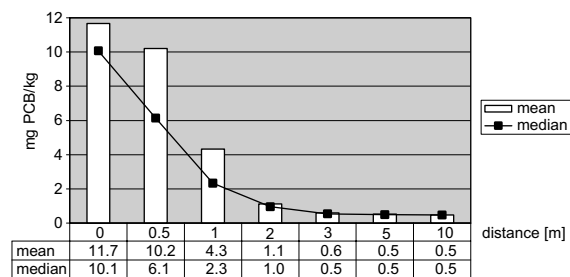


Fig. 1. PCB concentrations in the soil around the studied buildings.

(Fig. 1). The average concentration in the samples from the area within 2 m of the buildings was 6.83 mg PCB/kg (w.w.) and in the distance of 3–10 m from the walls 0.52 mg/kg-w.w., respectively. The chromatogram primarily resembled the profile of Aroclor 1260 and, in some cases, Aroclor 1254. The chromatogram was almost identical with that of the technical mixtures. The major congeners found in soil were PCB 101, PCB 118, PCB 138, PCB 153 and PCB 180. The most toxic congeners (PCB 126, TEF = 0.1; PCB 169, TEF = 0.01) were not found in our samples.

The PCB concentrations in the soil clearly varied according to the cardinal point. They were the highest on the southern side of the buildings. The average concentrations were the following: 16.6 mg/kg to the south, 2.00 mg/kg to the west, 2.39 mg/kg to the east, and 3.96 mg/kg to the north.

We also studied PCB concentrations from four sandpits on children's playgrounds within about 5–15 m of the buildings. The concentrations varied from <0.01 to 0.19 mg/kg. High PCB concentrations were not found, probably because the sand is normally changed annually.

3.2. PCBs in blood serum of residents

There were no statistically significant differences between the blood PCB levels of the inhabitants of the PCB-building and those of the control group (Table 2). The member of the control group with the highest PCB serum level had had some extra exposure during leisure-time activities. The high exposure was mainly attributed to PCB 28 and PCB 52 congeners, which are not prominent in the Aroclor 1260 or 1254 used in sealants.

3.3. Health risk assessment

According to our calculations, the average daily PCB intake of a child (1–6 years of age) through ingestion was about 0.34 $\mu\text{g}/\text{day}$ or 0.016 $\mu\text{g}/\text{kg}/\text{day}$ (ingestion 100 mg/day, 180 days/year) corresponding to 1.13 $\times 10^{-6}$

Table 2

Blood serum PCB concentrations of residents of PCB buildings and controls (the results are expressed as the sum concentration of 10 PCBs (PCB 28, 52, 77, 101, 118, 126, 138, 153, 169, 180))

Parameter	Residents of PCB buildings (n = 24), $\mu\text{g}/\text{l}$	Control group (n = 27), $\mu\text{g}/\text{l}$
Mean	2.09	1.79
Median	1.84	1.17
Standard deviation	0.96	2.33
Range	0.95–4.13	0.23–12.56

$p = 0.559$, not statistically significant.

Table 3

Calculated PCB exposure and cancer risk for exposed children aged 1–6 years

Soil PCB, mg/kg	LADD, mg/kg/day	Risk
<i>Ingestion</i>		
6.83	1.13×10^{-6}	2.3×10^{-6}
0.52	8.60×10^{-8}	1.7×10^{-7}
<i>Dermal</i>		
6.83	7.7×10^{-7}	1.5×10^{-6}
0.52	5.9×10^{-8}	1.2×10^{-7}

Combined risk 3.8×10^{-6} , when the PCB level of the soil is 6.83 mg/kg and 2.9×10^{-7} at a PCB level of 0.52 mg/kg.

10^{-6} mg/kg/day as the LADD, when the soil PCB concentration was 6.83 mg/kg (Table 3). This LADD corresponds roughly to 1.3×10^{-5} ng I-TEQ/kg/day (= 0.09 μg WHO-TEQ/kg in a week), which is a low level compared with the recommended tolerable weekly intake (TWI) of 14 μg WHO-TEQ/kg (SCF, 2001). Furthermore, the LADD corresponds to less than 10% of the estimated daily dietary intake (1.5 $\mu\text{g}/\text{day}$) of PCBs in Finland (Mustaniemi et al., 1995). The exposures calculated for dermal contact were 7.7×10^{-7} mg/kg/day (corresponding soil to a soil concentration of 6.83 mg/kg) and 5.9×10^{-8} mg/kg (corresponding soil concentration of 0.52 mg/kg), both expressed as the LADD. The combined cancer risk via both exposure routes (ingestion, dermal) is 3.8×10^{-6} .

The deterministic calculations gave risk estimates of 2.3×10^{-6} (soil ingestion) and 1.5×10^{-6} (dermal contact), corresponding to the 92nd and 88th percentiles, respectively (Table 4). The 90th percentile was 2.5×10^{-6} (soil ingestion) and 1.9×10^{-6} (dermal contact). According to Monte Carlo simulation (Crystal BallTM-programme), the maximum cancer risk was on the level of 10^{-4} .

According to our calculations, the non-carcinogenic risks remained below the acceptable risk level (HQ = 1) (Table 5). If children are exposed to soil PCB level of 6.83 mg/kg (within 2 m from the buildings) their exposure is only slightly lower than the recom-

Table 4
Results of the probabilistic risk assessment and comparison with the deterministic point estimates

	Risk	
	Ingestion	Dermal
<i>Deterministic</i>		
Point estimate	2.3×10^{-6}	1.55×10^{-6}
Percentage point	92%	87%
<i>Probabilistic</i>		
Mean	1.2×10^{-6}	9.6×10^{-7}
Median	3.1×10^{-7}	2.0×10^{-7}
Maximum	1.4×10^{-4}	1.5×10^{-4}
Minimum	8.1×10^{-10}	1.9×10^{-10}
90% percentile	2.5×10^{-6}	1.9×10^{-6}

Table 5
Point estimates calculated for PCB intake from soil in comparison with a reference dose of 0.02 $\mu\text{g}/\text{kg}/\text{day}$ for non-cancer risks and Finnish dietary intake (0.021 $\mu\text{g}/\text{kg}/\text{day}$)

Soil PCB concentration (mg/kg)	Calculated total intake ($\mu\text{g}/\text{kg}/\text{day}$)	Hazard quotient (HQ)	Percentage of Finnish dietary intake
6.83	0.016 (1–6 a)	0.8	76
	0.0018 (70 a)	0.09	2.7
0.52	0.0012 (1–6 a)	0.06	6
	0.0001 (70 a)	0.005	0.2
50 (Hazardous waste limit)	0.117 (1–6 a)	5.8	557
	0.014 (70 a)	0.7	20

mended reference dose of 0.02 $\mu\text{g}/\text{kg}/\text{day}$ and the average dietary intake of adults. However, the soil PCB concentration in sandpits and in the playgrounds is less than 0.5 mg/kg, and thus exposure is lower.

4. Discussion

According to our study, PCBs in the sealants of concrete buildings spread from the constructions to the soil adjacent to the building. The average PCB level was 6.83 mg/kg in the top soil within a distance of 2 m of a building. This value is about tenfold the concentration level found in some Finnish cities. The maximum concentration in our samples was 26.9 mg/kg. In some earlier studies even higher concentrations have been found (Jansson et al., 1997; Pyy and Lyly, 1998). The biodegradation of PCB compounds is slow due to their persistency in the environment. The variance of the PCB concentrations in the soil samples within 2–3 m of the buildings was high, probably due to the recent replacement of sealants.

According to a previous study, the average background PCB concentration levels in parks are 0.025 mg/kg (d.w.) (city of Tampere) and 0.053 mg/kg (d.w.) (city of Helsinki) (Salla, 1999). PCBs probably appear in soil bound in small sealant particles. The bioavailability of PCBs from such particles is still unknown. In our study, the concentrations in soil exceeded the Finnish soil limit value for PCBs (0.5 mg/kg). Despite the elevated concentrations, the health risks (expressed as the excess lifetime cancer risk) remained on the level of 10^{-5} – 10^{-6} . In our calculations, we assumed the bioavailability of ingested PCB to be 100%, but in practice, this value is lower due to the soil matrix. For the dermal route of exposure we used the 0.06 absorption factor, which is based on an experimental study with a PCB compound in soil matrix (US EPA, 1992).

Our results indicate that direct exposure to PCBs in contaminated soil is lower than the exposure from food-stuffs, except during childhood, when it in worst case almost reaches the same level. This calculation is based on a soil contamination level of 6.83 mg/kg, which exists only in the immediate vicinity (0–2 m) of PCB-containing buildings. However, during the removal of old PCB sealants, and especially when old seams are being sanded, the exposure of residents can be considerably higher for a short period of time. Our assessment focused on children from 1 to 6 years of age. The exposure of adults is obviously lower due to their insignificant soil ingestion, and our biomonitoring results (serum PCB levels) are in agreement with this. “Pica children” may, however, eat several grams of soil per day instead of the 100 mg/day used in our calculations, and thus their exposure may be 10–100-fold greater. The prevalence of the Pica phenomenon among children is not known in Finland.

The method we used for assessing the risk of cancer is based on the linearized multistage model. Since PCBs are probably a promoter type of carcinogen, this approach probably overestimates the actual cancer risk. In addition, not much is known about several exposure factors, and thus the conservative assumptions used in our calculations increase the conservative nature of the risk estimates.

Exposure to carcinogens in childhood may be more dangerous than later in life and guidelines have been developed to address this problem (Ginsberg, 2003). According to the US EPA, child-specific safety factors should only be used for direct carcinogens, not for promoters such as PCBs. The calculated average daily intake as a child (1–6 years) 0.016 $\mu\text{g}/\text{kg}/\text{day}$ is, however, less than the RfD 0.02 $\mu\text{g}/\text{kg}/\text{day}$ based on immunosuppression and an uncertainty factor of 300.

Our probabilistic risk assessment showed that the maximum cancer risk via both exposure routes was on the level of 10^{-4} , and the 90% percentile was on the level of 10^{-6} .

The PCB concentrations in the blood serum specimens of our study did not differ significantly between

the residents of “PCB buildings” and the control group. Differences in the age distributions of the two groups caused some bias however. A clear correlation was obvious between age and the serum PCB level of the studied groups. The slope of the regression line was about 0.05 for both groups. This finding indicates that, if the age distribution of both groups had been the same, the average serum PCB concentration of the control group would have been about 0.5 µg/l higher. The blood PCB levels of the residents of the PCB buildings were lower than that of the workers involved in removal of old sealants (means 2.1 µg/l vs. 3.9 µg/l) (Kontsas et al., 2004).

5. Conclusions

Soil can be heavily contaminated with PCBs in the immediate vicinity of buildings in which PCB-containing sealants have been used. The area south of these buildings is more contaminated than those in other directions, and, therefore, the weathering of sealants is probably an important mechanism in the spread of PCBs to the surroundings.

Children between 1 and 6 years of age are the main risk group, especially because of their frequent and common hand-to-mouth behaviour. However, the health risks due to contaminated soil are probably small due to the low level of exposure.

The blood serum PCB levels of residents of “PCB-buildings” did not significantly differ from those of a control group.

Similar construction technology has been used at least in other Scandinavian countries (Sweden, Norway), and Germany. Recently PCB-containing caulking was found to be common in public buildings in the Boston area in the United States (Herrick et al., 2004). Probably the same construction technology has been used widely in Western countries, but polysulphide sealants have not been recognized as a PCB source. Our results can be applied to other Scandinavian countries, but in warmer countries the exposure of children may be higher due to longer summers. In our calculations we anticipated that children play in the yards of these buildings only 180 days per year.

Immediate actions to protect PCB-building inhabitants are not required. However, we offered some recommendations to reduce children’s PCB exposure further, for example, placement of both sandpits and children’s playgrounds outside the immediate vicinity of “PCB buildings” and the restriction of vegetable gardens in the yards of such buildings.

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