PCB IN INDOOR AIR AND DUST IN BUILDINGS IN STOCKHOLM

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ABSTRACT
PCB levels in indoor air were low, (>50 ng/m^3) in 9 flats and 3 schools. In 15 flats levels varied more (23-270 ng/m3). Sealant contained a less volatile high-chlorinated PCB-mixture except in one case. Levels were over 300 ng/m^3 in 2 flats analysed from passive samples, but were below this from active samples. The most toxic PCB congeners were very low. PCB in house dust varied considerably (0.01 - 5.4 µg/g) and was also found in buildings without PCB in sealant. Average PCB level in dust is of the same magnitude that one-gram of dust represents the average daily intake of PCB from food. Small children should not be exposed to this dust. PCB levels in indoor air may correlate with ventilation, floor level and dust. More study is needed on dust and/or gas paths and exposure. Quality control of the indoor environment should be improved when dismantling PCB sealant and a safe-level needs to be established.

INDEX TERMS
PCB indoors, Air & Dust, Monitoring, Health effects, Safe-level

INTRODUCTION
As part of the City of Stockholm's Environmental Monitoring a study of polychlorinated biphenyl's (PCB) concentrations in the indoor environment was carried out. Two areas were studied, area 1 in the city, (9 flats) and area 2 a northern suburb (15 flats & 3 schools). The primary aim has been to ascertain PCB-levels in domestic buildings with PCB sealant on the outer part of the building envelope and relate to possible exposure. The choice of buildings has not been random or completely systematic. In all cases a PCB-inventory had been made and sealant was to be removed after the survey. Building owners, administrators and tenants interest to take part in the study have limited final choice of buildings. Results are not completely representative of the PCB buildings in Stockholm. Elastic sealant is the only known source of PCBs.

Sealant containing PCB has been used in Sweden from 1956 to 1973, mainly in building facade constructions in joints between cement blocks, around windows, balconies, etc. PCBs are a group of 209 fat-soluble congeners that have a number of adverse health effects to both man and animals, e.g. cancer, effects on reproduction and the nervous- and immune systems. PCBs are persistent chemicals and accumulated higher up in the food chain through biomagnification. Risk for human exposure has been primarily from foods such as fish, milk products and breast milk. PCB levels have deceased in foods recently. Present day health assessment is based upon the most toxic dioxin-like PCBs expressed as Toxicological Equivalents (TEQs), and does not take into account all possible paths of exposure or all effects that different PCBs can give [Bernes, et al. 1998]. The Swedish Food Administration gives recommendations and has limit-levels for foods such as fatty fish. Congeners vary in

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degree of chlorination and in spatial configuration of the biphenyl molecule. Different producers have developed different technical PCB-mixtures to soften sealant. These mixtures contain large numbers of PCBs. Primarily because more volatile, less chlorinated PCBs, are emitted from the source during transition to vapour phase, the distribution of PCBs is different compared to that in the original source. The distribution of PCBs in sealant and in indoor air differs from that in foods and blood [Johansson, 2001]. In Sweden focus has been on the removal of PCB. In a first phase PCB above 500-ppm in sealant is to be removed by 2002/03. Contractors dismantling and disposing of PCB sealant must follow guidelines set by the Environmental Administration.

METHODS

Air samples were taken in all localities, including two references (building 2 & 4), using a standard pump sampler OVS-2 (OSHA versatile sampler, SKC 226-30-16) containing XAD-2 adsorbent, a back-up section and filter. Sampling flows were about 1 l/min. Samplers were placed where convenient for tenants over 2 days. Extraction of the filter and adsorbent was made using an ultrasonic bath with 5-ml toluene to which an internal standard - 1,2,3,4-tetrachlornaftalen - was added. Total PCBs were calculated from ∑6 PCB (IUPAC number: 28, 52, 101, 138, 153, 180) multiplied by a factor 5 [Bossenmayer et al, 1996]. Based on the results from active sampling passive samples were taken using a semipermeable membrane device (SPMD) set out over 2 weeks. Sampling was based on highest PCB-levels, 2 from area 1, 7 from area 2 and 1 from a school. The SPMD sampling equipment has been developed by the Institute of Environmental Chemistry, Umeå University [Strandberg et al, 1997].

Dust samples were taken using a VacuuMark-sampler and a vacuum cleaner. Surfaces sampled varied according to availability and amounts of dust. Tracking dust paths was not included. PCB in dust was extracted twice in heptane then shaken with sulphuric acid. The heptane phase was evaporated to 1 ml to which the internal standard was added. PCBs in dust are expressed as ∑7PCB µg/g. PCB patterns in dust varied quite considerably and at present there is no standard procedure for calculating total values.

Sealant was taken with a knife that was cleaned between samples. About 0.1 g of the material was extracted twice in heptane, the extract was shaken with sulphuric acid, the heptane phase was transferred to a 100 ml measuring cylinder and diluted with heptane up to the mark. From this10 ml was taken to which the internal standard was added. Total PCBs were calculated as the concentration of ∑7 PCB times a correction factor depending upon the technical PCB-mixture [Schulz et al, 1989].

PCB concentrations in toluene and heptane extracts were determined by means of gas chromatography and mass spectrometry in which selected ion monitoring (GC-MS SIM) was used for quantifying 7 indicator PCBs (IUPAC number: 28, 52, 101, 118, 138, 153 and 180).

Ventilation measurements were made using a passive tracer gas technique over a period of 2 weeks, developed by Pentiaq AB [Stymie & Boman, 1994], and include air exchange rate, average age of the ventilated air and the main air flow for each room in which measurements were made.
Indoor temperature was also registered during measuring periods.

Questionnaire

Tenants in apartments were asked 3 questions after measurements; how well ventilated is the flat, how often do you air and how often do you vacuum clean.

RESULTS
Table 1 shows PCB concentrations in indoor air, house dust and sealant respectively as well as the performance of the ventilation system.

PCB in sealant
In most cases the PCB mixture is of a high-chlorinated type (Clophene A60). In two cases low-chlorinated PCB mixtures were found (Clophene A40), in building 2, area 1 and in building 6, area 2. PCB levels in sealant are comparable to other buildings in Stockholm, however, the amount of PCB sealant in the buildings was less. In building 1, area 1 and two schools in area 2 PCB-levels were low. In a third school sealant levels were high and only here could PCB be detected in indoor air. In the apartment buildings in area 2 sealant-levels were generally higher.

PCB in indoor air
Total PCB concentrations from active pumped samples vary from under detection level to 270 ng/m^3 (Table 1 & Figure 2). The lowest level was in reference locality 2 in the city and the highest was in a basement flat where the PCBs in sealant were less chlorinated (A40).

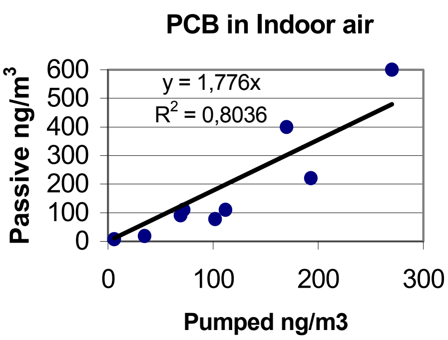

Figure 1 Correlation between 9 active and passive indoor air samples

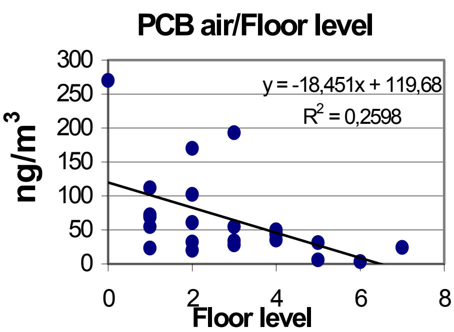

Figure 2 Correlation between floor level and pumped indoor air samples (23)

Lowest levels were in building 1. Concentrations varied more in area 2 for both active (a) and passive (p) samples and were low to moderately high; ten cases <100 ng/m^3, four 100-200 ng/m^3 and one apartment with 270 ng/m^3. Only one school had PCBs in indoor air (37 ng/m^3). There is a reasonably good correlation between active and passive samples (Figure 1) especially at concentrations around and below 100 ng/m. In two cases passive samples were twice the level of pumped samples (400/170-ng/m^3 and 600/270 ng/m^3).

Concentration of dioxin-like PCBs in 9 flats and 1 school, expressed as Toxic Equivalents (TEQ), were very low, 0.001-0.1 pg/m^3 and represent less than 1% of the normal total intake from food, if calculated as 100% uptake and a given breathing rate.

PCB in indoor dust
PCB concentrations in dust from flats varied considerably, 0.01-5.4 µg/g ∑7PCB (Table 1), and were even registered in buildings without or at low levels of PCBs in sealant. Dust levels do not seem to correlate with other parameters investigated. Some degree of correlation may exist between PCB in dust and indoor air (a), but distributions between flats are large. In 8 flats with PCBs in dust below 1 µg/g indoor air levels were below 50 ng/m^3, and in 6 flats with dust-levels of 1-2 µg/g the indoor air levels were 50-100 ng/m^3. Reference localities had

0.06 µg/g, area 1 and 1.8 µg/g, area 2. The latter gives evidence of variable tracking sources of PCBs in house dust.

Table 1: Summary of measurements: area 1, buildings 1-3 in the city, and area 2, buildings 4-9 in a northern suburb. Number of samples in brackets, reference localities (ref).

Area/ Buildings 1-9 (No. samples)	**Indoor air active(a)** ng/m^3	**Indoor air passive(p)** ng/m^3	**Dust** ∑7PCB µg/g	**Sealant %** (A60)	**Ventilation ACH $^{-h}$**
Area 1: (9)					
1 (3)	3-6	7 (1)	0.2-3.6(4)	0-0.0014	0.47-1.26
2 (ref)	0	-	0.06	0	1.06
3 (5)	20-35	18(1)	0.37(6)	14-24	0.30-1.01
Mean (8)	**19**		**0.8**(10)	**12.3**	**0.6**
Standard dev.	**12.8**		**1.2**	**10.6**	**0.3**
Area 2:(15)					
4 (ref)	31	-	1.8	0	0.35
5 (14)	23-193	78-400(6)	0.01-5.40	17-25	0.32-1.69
Mean (14)	**76**	**186**(8)	**1.4**	**19.7**	**0.7**
Standard dev.	**51.5**	**130**	**1.3**	**2**	**0.4**
6 (1)	270	600	0.53	12 (A40)	0.32
Area 1+2: All flats					
Mean (23)	**64.6**	**180.4**(9)	**1.1**(25)	**16.8**	**0.67**
Standard dev.	**66.1**	**198.1**	**1.2**	**7.2**	**0.34**
Schools (3)					
7-9	0-37	37(1)	0.27-3.1	0.007-12	

Ventilation

Air exchange rates represent total ventilation in every apartment and include airing (Table 1). Flats have exhaust ventilation with possibilities for extra exhaust above stoves in kitchens. Air exchange rates are on average above 0.5, but variations are quite large. In 9 flats rates were below 0.5 even though airing was quite frequent. Air exchange rates are probably lower in wintertime (less airing). These results are in general accord with previous studies in Sweden. The three schools in area 2 had mechanical supply and exhaust ventilation, as did the reference building office in area 1. Air exchange rates in schools were between 1.5 and 3 h $^{-1}$.

Questionnaire

Tenants were generally dissatisfied with ventilation, particularly in area 2. A vast majority of tenants in both areas aired their apartments at least once a day and most of them vacuum cleaned at least once a week. The basement flat in building 6, where indoor air levels of PCBs were highest was also less frequently aired and cleaned.

COMPARISON OF RESULTS AND DISCUSSION

In the three schools in area 2 there was a clear relationship between PCB levels in sealant, indoor air and dust. Here it is possible that PCB in house dust could be more of a problem than in indoor air, especially as the quality and frequency of cleaning procedures has deteriorated in many schools over the last few years.

The following is a comparison and discussion of results in apartments in the two different areas. Indoor air concentrations refer to pumped samples if not otherwise stated.

Indoor air

Compared to other measurements of PCB in indoor air in Swedish flats (spot samples), levels are lower than the highest concentrations measured, around 600 ng/m^3 [Sikander et al, 1999], but are generally higher than those previously measured in other areas in Stockholm, average of 59 ng/m^3 [Jansson et al, 1997]. PCB concentrations in indoor air are most probably caused by PCB-sealant on the outer part of the building structures and indicate continuous contamination of the indoor air. Most probably there is leakage in the building envelope and migration within the facade construction. Increases in air exchange rates could also increase PCB emission rates [Detlef, 1993] especially in buildings with exhaust ventilation. Outdoor temperatures, wind and degree of sealant weathering are probably other factors of importance. Floor level seems also to play a role; PCB-levels in indoor air tend to be higher lower down in buildings (Figure 2). A possible cause could be PCB leakage from the socle-sealant between the base and the lower unit.

According the Swedish Environmental Protection Agency the average daily intake of PCBs from food is 3 µg per day [Bernes et al, 1998]. Assuming 100% absorption by the body of the inhaled PCBs at an air rate of 20 m^3/day, then the daily intake from indoor air would be the same as for food if the PCB concentration in air were 150 ng/m^3. The highest PCB-levels are of that order measured in flats in area 2, even though the PCB-sealant load was relatively low and PCBs in the sealant are mainly highly chlorinated.

Comparing results with German Guidelines PCB levels in air in two flats, passive method, are such that the source of PCB should be found and removed (300 ng/m^3). Levels are lower from samples taken with the active method. There are three levels in the German Guidelines: 30 ng/m^3 - Long-term target, 300 ng/m^3 - Guideline level and 3000 ng/m^3 - Level of concern.

Differences in results between the two sampling methods could be due to a number of reasons [Detlef, 1993 and Bleeker, et al, 1999]. Measurements were carried out over different time periods and lengths of time (2 days Vs 2 weeks), winds and outdoor temperatures were not the same and ventilation conditions were different. There is no clear correlation between air exchange rates and PCB levels from active samples. There is some degree of correlation between air exchange rates and PCB-levels from passive samples, especially for higher values. In the case of the highest measurements - 600 ng/m^3 (p), 270 ng/m^3 (a) - the average outdoor temperatures were 14 ^{0}C and 11 ^{0}C respectively. A study has shown that PCB concentrations in a rum over a period of 9 months can vary from 200 to 2700 ng/m^3 and correlated to the outdoor temperature [Benthe, et al, 1992].

Dust

In a previous study in Sweden high concentrations of PCB in house dust were found, 4 µg/g $\sum$7PCB, in a house with PCB-sealant [Arnér et al, 1998]. Here too concentrations varied considerably. In the same study dust samples were taken from indoor air, but results were quite low. It is perhaps to be expected that PCB levels in house dust vary. Different surfaces were sampled, the majority of tenants clean quite regularly and air their apartments. House dust is of different age, can accumulate for example on textile surfaces over longer periods of time and can come into buildings in different ways. For example from contaminated soil in housing areas through personal tracking and/or weather and wind. PCBs can accumulate in house dust in buildings with low or no PCBs in sealant in PCB contaminated housing areas. More needs to be done to establish these paths and determine possible risks for exposure. The average PCB concentration in all dust samples is of the same magnitude that one-gram dust represents the daily intake of PCB from food, assuming total absorption by the body. This points to the importance of cleaning frequency and methods in PCB contaminated buildings, and that particularly small children should not be exposed to such dust.

CONCLUSIONS AND IMPLICATIONS

In Sweden there are no guideline or limit-levels for PCB in indoor air or dust in domestic, pre-school or school buildings. Nor are there any long term goals for the indoor environment after removal of PCB sealant from buildings. To minimise the spreading of PCB to the outdoor and indoor environments the Environmental Administration in Stockholm together with the Building Sectors Eco-cycle Council have specified general quality control procedures during dismantling of PCB contaminated material in buildings. To assure lower burdens in the indoor environment PCB in indoor air and dust should be measured before and after removal as part of quality control. Further monitoring of PCBs in the indoor environment is necessary, including tracking paths of contaminated house dust. A safe-level for the indoor environment needs to be established based upon risk assessment of all PCB congeners.

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