Highly PCB-contaminated schools due to PCB-containing roughcast

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ABSTRACT
The Bremer Umweltinstitut (Environmental Institute of Bremen) has examined more than 200 schools and kindergartens. The indoor air contamination with polychlorinated biphenyls (PCBs) varied between several ng/m^3 up to 40 000 ng/m^3 (40 µg/m^3). The most important sources were sealants and leakage from capacitors. The parameter responsible for extremely high indoor air concentration was the use of PCBs as flame retardants on ceiling panels and the usage of high PCB containing roughcasts. Roughcast as a source has not been discussed in the literature till now. Very high indoor air contamination with PCBs is leading to secondary contamination of different materials up to more than 1000 mg/kg.

INDEX TERMS
Polychlorinated biphenyls (PCBs); Building products; Roughcast; Measurement strategy

INTRODUCTION
Polychlorinated biphenyls (PCBs) are a class of substances industrially produced since 1929 and now being used all over the world. Production and uses were banned in many countries beginning in the late 1970s due to their toxicological properties.

Restrictions were provoked, e.g. by several accidents in industrial plants in Japan and Korea in 1968 and 1979, when PCB-contaminated rice oil was liberated. In the USA, the production of PCBs was stopped in 1977. PCBs are not produced in Germany since 1983 and since 1989 even their use has been restricted.

Apart from many industrial purposes, PCB-containing products were used in numerous building products. Electric appliances equipped with capacitors containing PCBs were often installed in bigger buildings like schools and public houses. In Germany, many of these capacitors were replaced due to problems, especially PCB-releasing accidents. Another important application of PCBs in buildings was their use in elastic sealants as a flame retardant and as plasticizers. For these properties, PCBs were added to paints and other material.

For the evaluation of the PCB load within a building, the examination of indoor air has been established especially because unknown sources can contribute to the air contamination. If only sealants are tested for PCB loads—a usual procedure in Germany—buildings with other PCB sources except sealants may be declared free of PCBs by mistake.

Especially, indoor air from buildings with flame retardant coatings or other extensive sources show significant higher PCB loads compared to the air from buildings containing ‘only’ PCB-loaded sealants.

The measuring strategy of the Bremer Umweltinstiut for the determination of PCB loads in buildings intends a spot-check-like examination of the indoor air, considering different stages of building, of renovation and of suspicious materials.

In this manner, in numerous buildings, building parts or single rooms with high PCB burdens (significantly greater than 300 ng/m^3) were identified.

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METHODS

Before taking air samples, the room must be ventilated for 8 h. After ventilating, doors and windows have to be kept closed until sampling. Room temperature while sampling should be at least approximately 20°C (68°F) to ensure conditions like normal usage of the room. In order to simulate the swirl-up of dust during normal usage dust should be raised by a fan.

The air samples were taken with calibrated pumps (rate 50 l/min) and collection of the PCBs using adsorption cartridges (according to VDI-guideline 4200.2, 1997). The cartridges contain glass fibre filter plates and highly cleaned soft polyurethane foam as adsorbents. The foam is spiked with a solution of ^{13}C-labelled PCB 28 as an internal standard. At least 4000 (better 6000 or more) litres of air should be collected per sample.

Loaded cartridges were eluted with *n*-hexane (research grade) after adding 1 ml of *n*-nonane as a keeper and ^{13}C-labelled PCB 153 as a further internal standard. This was followed by concentrating the hexane and clean up by shaking with concentrated sulfuric acid. Separation, identification and quantification was done by capillary gas chromatography (Shimadzu, type GC-17A) equipped with a DB-5MS type column with the dimensions 60 m × 0.25 mm × 0.25 µm connected to a mass selective detector (Shimadzu, type QP-5050). Samples were injected into a split/splitless injector with an autosampler (Shimadzu, type AOC-20i/s).

The surface contamination was determined by analysing wipe samples. They were taken by repeated wiping of a defined area of the surface with a highly cleaned, solvent-soaked cloth. (Lammers *et al.*, 2001; Weis *et al.*, 2002).

Loaded cloth pieces were spiked with PCB 209 as the internal standard and eluted with *n*-hexane (research grade) by Soxhlet extraction. Clean-up was performed by shaking with concentrated sulfuric acid. Separation, identification and quantification was done by capillary gas chromatography (Shimadzu type; 14A) equipped with a DB-5 type column with the dimensions 60 m × 0.25 mm × 0.32 µm connected to an electron capture detector (ECD). The samples were injected into a split/splitless injector with an autosampler (type AOC 14).

Samples taken from sealants were spiked with PCB 209 as the internal standard and eluted with iso-octane (2,2,4-trimethylpentane) in an ultrasonic bath. The solvent was cleaned by means of ready-to-use sulfonic acid cartridges. Analysis was performed as described for the wipe samples.

Samples from materials other than sealants, e.g. roughcast, were spiked with PCB 209 as the internal standard and eluted with toluene, the clean-up and the analysis procedure being performed as described for the wipe samples.

The PCB content of all samples was calculated by summarising the concentrations of the following congeners: IUPAC # 28, 52, 101, 138, 153 and 180 and multiplying by a factor of 5, which is a well-established method (DIN 51527-1, 1987; Bossenmayer *et al.*, 1996).

RESULTS

During the investigation of a school with two phases of construction (1955 and 1963), where no sealants could be seen, we measured indoor air concentrations between 26 and 61 ng/m^3 in a spot-check. The indoor air of a single room contained 1700 ng/m^3. In this room, sound insulating ceiling panels were installed. The flame retardant coating of these panels contained 110 000 mg/kg PCB. This room was cleared out and the walls were painted; a further examination of the indoor air performed 3 weeks later showed an indoor air burden of 4400 ng/m^3!

In the indoor air of numerous schools in northern Germany, concentrations up to approximately 40 000 ng/m^3 were measured. As the major source causing these extremely high indoor air loads, we identified a special kind of plaster: roughcast. This plaster consists of small pebbles, plastics and PCBs as plasticizer.

In Table 1 we present the indoor air concentrations from some schools with roughcast as a major source of PCB contamination.

Table 1 Single PCB concentrations in rooms built with roughcast

	Room with PCB source (roughcast)	PCB indoor air loads (ng/m^3)
School 1	Corridor A	14 000
	Corridor B	8400
	Hall	4400
	Library	8600
School 2	Corridor A	34 000
	Corridor B	31 000
School 3	Corridor 1 gymnasium	9200
	Corridor 2 gymnasium	39 000
	Hall	7700
	Corridor 3	11 000
	Corridor 4	7400
School 4	Corridor A	11 000
	Corridor B	8000

Especially in the corridor areas of these schools, durable roughcast was used as wall coating. The PCB contents of the examined roughcasts ranged between 15 000 and 47 000 mg/kg.

In Table 2 the indoor air loads in rooms adjacent to the primarily burdened rooms are shown. No PCB sources had been found in these rooms.

Table 2 Rooms without PCB sources, adjacent to primarily burdened rooms

Building	Rooms without sources	PCB indoor air loads (ng/m^3)
School 1	Class rooms	770–3500
School 2	Class rooms	300–6300
School 3	Class rooms	550–4500
School 4	Class rooms	950–1600

If furnishings and fittings were exposed to indoor air burdens at levels of 10 000 ng/m^3 for years, they show secondary contaminations in the range of the so-called primary sources (more than 1000 mg/kg). In Table 3, some selected secondarily contaminated materials are shown.

Table 3 Secondary contamination in rooms with roughcast as a PCB source

Building	Material	PCB load (mg/kg)
School 1	Sound insulating ceiling panel	2800
	PVC tiles (dark)	1600
	Silicon sealant from window frame	2500
	Lacquer on window frame	7600
	Anti-skid edge (rubber) on stairs	2200
	Book cover	1900
School 2	Lacquer on window frame	1600
	Lacquer on banisters	1800
	PVC folding wall	660
	Silicon sealant from window frame	950

School 3	Silicon sealant from window frame	2000
	Sound insulating ceiling panel	820
	Wall paper	2200
	Wall paint	1200
School 4	Lacquer on new banisters	360
	Lacquer on door frame	390

In Table 4 the results of surface examinations (performed by taking wipe samples from secondarily contaminated materials) are shown.

Table 4 Results of the surface examinations (wipe samples)

Building	Wiped material	PCB load (µg/m^2)
School 1	Window frame	59 000
	PVC tiles (light)	11 000
	Lacquer on radiator	2300
School 2	Floor tile made of artificial stone	7700
	Lacquer on banisters	17 000
School 3	Window frame	11 000
	Skirting-board made of slate	34 000
	Lacquer on door	7000
	Stairs made of artificial stone	66 000
School 4	Slats in metal ceiling	7200
	Floor tiles	3100

In house-dust settled on ceiling panels or in radiation systems over a long period of time, a PCB contamination between 20 and 980 mg/kg was determined.

The PCB load in the indoor air of 15 identical rooms of a building constructed in one phase, the concentration varied between 110 and 4400 ng/m^3. A sealant around window frames and door frames was identified as the PCB source. Although all sealants seemed to be identical, unexpectedly their PCBs varied widely—from extremely high to low. For this reason, differences in the air concentration were found.

Also, the distribution of sealants in the building and the rooms was irregular, so that in some rooms all sealants were loaded with PCBs, while in others only parts or none of the sealants were contaminated.

DISCUSSION

With our investigation we proved, that the spot-check-sampling of material is not suitable for establishing the PCB status of a whole building. Some materials may contain PCBs while others are clean. Even in buildings containing only sealants as a possible source, this spot-check can lead to a wrong assessment.

From our experience, an irregular distribution of PCB burdens may occur even in buildings constructed in one phase. One reason for these variations is that the PCBs were added to the sealants as a plasticizer directly during construction. The amount of PCBs added to the sealants depends on the actual temperature, because lower temperatures demand more plasticizer.

The closer examination of buildings by indoor air sampling followed by source sampling revealed a source, which has not been described before: roughcast. This plaster has been developed intentionally for outdoor use, but due to its durability, it was also applied to rough-worn areas like gyms and corridors of schools. The extended use of this PCB source leads to extreme high indoor air loads. This impact has not been described before (Hassauer and Kalberlah, 1999).

This high burden can last for decades and can lead to secondary contamination of materials resulting in concentrations similar to primary sources of approximately 1000 mg/kg (Zwiener, 1997).

One of the most important problems concerning the redevelopment of PCB loaded buildings is the secondary contamination of materials and furniture. A redevelopment of loaded buildings with a decrease to significantly less than 300 ng/m^3 according to the German PCB guidelines (Lukassowitz, 1990) or even less than 100 ng/m^3 as it is demanded by more critical experts (Fobig, 1996; Kruse, 1996; Müller *et al*., 1999; Weis and Ruhnau, 2002) is only possible if both primary and secondary sources are identified and replaced in an expert manner. From our experience, the guideline value for redevelopment will not be reached, if secondary contamination is not considered.

Redevelopment will also not be successful if further contamination occurs while removing the PCB sources. Even simple renovation actions, where PCB sources are not touched, such as wall painting in a room containing loaded ceiling panels, can raise the PCB concentration in the indoor air significantly. This may be caused either by swirling up contaminated settled dust or by the mobilisation of PCBs from contaminated materials by the solvents in the applied paint. Therefore, a successful redevelopment requires both: the removal of every PCB source by an expert and the removal of loaded settled dusts.

From our experience, even in the case of very highly burdened buildings, indoor air concentrations clearly below 100 ng/m^3 can be reached, if both primary and secondary sources are identified thoroughly and if the redevelopment is performed by an expert.

CONCLUSIONS AND IMPLICATIONS

The determination of the PCB burden of a building should be performed by examination of several indoor air samples in the first place. Since PCB contents of products may vary even if the products seem to be identical, only material spot-checks may not be sufficient. Furthermore, unknown sources like roughcast should be taken into consideration.

If the PCB contamination shall be reduced effectively, it is necessary to remove both the primary and secondary sources by an expert. Other actions—before redevelopment—like painting can lead to higher indoor air contamination.

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