Removal of PCB from indoor air and surface materials by introduction of additional sorbing materials

Gunnarsen, Lars; Lyng, Nadja; Kolarik, Barbara; Andersen, Helle Vibeke

Published in:
Healthy Buildings Europe 2017

Publication date:
2017

Document Version
Publisher's PDF, also known as Version of record

Healthy Buildings 2017 Europe
July 2-5, 2017, Lublin, Poland

Paper ID 0272 ISBN: 978-83-7947-232-1

Removal of PCB from indoor air and surface materials by introduction of additional sorbing materials

Lars Gunnarsen[1*], Nadja Lyng[2], Barbara Kolarik[1] and Helle Vibe Andersen[1]

[1] Department of Energy Performance, Indoor Environment and Sustainability of Buildings, Danish Building Research Institute, Aalborg University, Denmark
[2] Indoor Environment and Building Examinations, Danish Technological Institute, Denmark

[*]*Corresponding email: lbg@sbi.aau.dk*

SUMMARY
Alleviation of indoor PCB contamination is extremely expensive because PCB from old primary sources has redistributed to most other surfaces over time. This study investigates the introduction of new removable sorbing materials as a method instantly lowering the concentration of PCB in indoor air and slowly decontaminating old surface materials. In three bedrooms of a contaminated apartment respectively new painted gypsum boards, sheets of flexible polyurethane foam and activated carbon fabric were introduced. The PCB concentrations in room air were monitored before the intervention and several times during the following 10 months. The PCB concentrations in the old surface materials as well as the new materials were also measured. An immediate reduction of PCB concentration in indoor air, a gradual increase of PCB in new material and as well a gradual reduction in old surface materials were demonstrated.

KEYWORDS
SVOC, Sources and sinks, Emission, Sorption, Renovation method.

1 INTRODUCTION
Polychlorinated biphenyls (PCBs) have been used commercially since 1929 as dielectric and heat exchange fluids as well as in a variety of other applications including as plasticizer (WHO, 2000). PCBs have been used as plasticiser in building materials including sealants and were used in Denmark from the 1950s until banned in 1977 (Danish Environmental Protection Agency, 1983). Indoor sealants with PCB can cause a contamination of the indoor air (Frederiksen et al., 2012) and a need for alleviation. Over time the PCBs have redistributed from the original sources (primary sources) to the surfaces of the indoor environment (tertiary sources) through evaporation, dispersion and sorption with the indoor air as transport medium. Further PCB from the original source can migrate into adjacent material (secondary sources). Although the quantitative amount of tertiary sources can constitute only few percentages of the primary sources (Gunnarsen and Kolarik, 2014) alleviation becomes a challenge. The surface contamination is a reversible process and eliminating other sources can change the behaviour of the contaminated surface areas from sinks to sources. Experiments in contaminated rooms with encapsulated primary and secondary sources have shown that the surfaces have the potential to maintain a high indoor air concentration of PCB and also increase their emission rate with increasing air change rate (Lyng et al., 2015). Further the experiments indicated that

the bulk air concentration were less than 25% lower than the surface air concentration very close to the area sources (Lyng, 2016).

Redistribution of tertiary contaminations will happen when new material with sorptive properties is introduced to the environment. The new material will act as a sink depleting PCB from the bulk air. The old contaminated surfaces in the room will respond to the depletion and act as sources emitting PCB in response to surface concentrations and bulk air concentrations. Weschler and Nazaroff (2008) made some considerations concerning Semi-Volatile Organic Compounds (SVOCs) and the interaction of sources and sinks in relation to the gas-phase concentrations of SVOC in the indoor air. They identify three phases with different mechanisms dominating the redistribution processes. In the initial phase shortly after introduction of the source they estimate the bulk room concentration of the contaminant to be approximately the surface concentration close to the source times the ratio of the area of source material to total surface area.

This experiment introduces new clean materials into an old contaminated room, i.e. the opposite situation as Weschler and Nazaroff (2008). We consider the initial phase shortly after introduction of the new material with an area A_{new} into the room with old contaminated surfaces with a source area A_{old}. In line with Weschler and Nazaroff (2008) the emission rate, R_{old} from the old surfaces of the room is described as:

$$R_{old} = h_{old} \cdot A_{old} \cdot (C_{0old} - C_r) \quad (1)$$

with a mass-transfer coefficient for the emission, h_{old}, a surface concentration of the contaminated surface of C_{0old} and a bulk room concentration of C_r.

Similar a deposition rate R_{new} to the new introduced surfaces can be described as:

$$R_{new} = h_{new} \cdot A_{new} \cdot (C_{0new} - C_r) \approx - h_{new} \cdot A_{new} \cdot C_r \qquad (C_{0new} << C_r) \quad (2)$$

with a mass-transfer coefficient for the deposition, h_{new} and an insignificant surface concentration, C_{0new}. Initially these surface interactions by far exceed the impact of primary source emissions and continuous dilution by ventilation of the rooms. In the initial period we assume the gas-phase transport from the interface into the bulk air as the rate-limiting step for both emission and sorption and that the respective mass-transfer coefficients are of comparable magnitude. Combining equation (1) and (2) we get:

$$C_r \approx C_{0old} \cdot A_{new} / (A_{old} + A_{new}) \quad (3)$$

We assume that the surface concentration of the old area source C_{0old} are only slightly higher than the bulk room concentration before introducing the new material. As the redistribution of PCB from old to new materials continues the surface concentrations of both air and material on old as well as new surfaces are expected to change.

2 MATERIALS/METHODS

The experiments took place in a 15 floor building being part of an estate erected in 1969-1973 including 5 blocks having PCB containing sealants. Up to present time PCB have redistributed and contaminated most inner surfaces. The experiment was done in three similar rooms in an apartment on the 15th floor with indoor air concentrations of PCB_{total} exceeding 3000 ng/m^3 and

therefore vacated. Information about size and old surface materials of the rooms are summarized in Table 1.

Table 1. Descriptive data for the three experimental rooms. Surface area is sum of all inner visible areas including floor, ceiling and walls.

Room	**Floor area (m^2)**	**Room volume (m^3)**	**Surface area (m^2)**	**Air change (h^{-1})**	**Floor covering**	**Wall cover**
1	7.4	18.5	42.1	0.15	Wall to wall carpet	Acrylic paint on woodchip wallpaper
2	7.4	18.4	41.9	0.18	PVC	Acrylic paint on flat wallpaper
3	10.8	27.0	55.0	0.18	Wall to wall carpet	Acrylic paint on woven glass fibre

One year as well as 30, 29 and 1 day before introduction of the new materials the air concentration of PCB were measured in each room. The 8th of April 2016 three types of new materials were hung up in the rooms with one type of material in each room. The surface area of the new materials was equal to the total visible inner surface area of the empty rooms. This significant added material loading with width of lanes between new materials of approximately 0.4 m was expected to give some obstruction to the mixing and flow of air and stand fans operating at their lowest setting was introduced to compensate for this. Information about the new materials is found in Table 2.

Table 2. Description of the three new materials. Surface area refers to the exposed surface area (both sides of the new materials).

Room	**Material**	**Weight per surface area (g/m^2)**	**Surface area (m^2)**	**Total weight (kg)**
1	Three coats of paint on both sides of 13 mm gypsum board.	Dry paint: 326 Gypsum: 4500	42.1	Dry paint: 13.7 Gypsum: 190.0
2	10 mm polyurethane foam (PUF)	115	41.9	4.8
3	0.5 mm activated carbon cloth	60	55.0	3.3

Air concentrations were measured 1, 3, 7, 16, 33, 61 and finally 271 days after introduction of new material. Air samples were taken on sampling tubes as reported by Lyng et al. (2016). The seven congeners PCB-28, PCB-52, PCB-101, PCB-118, PCB-138, PCB-153 and PCB-180 were determined and their sum named "PCB_{sum7}". The total amount of PCB is defined as 5 times PCB_{sum7}. Material samples from surface materials (wall and floor cover) of the rooms were taken just before introduction of new materials and after 271 days. Samples from new materials were taken just before they were introduced, and after 34 and 271 days. Paint samples were taken by scraping down to a visible colour or texture change. Samples of flooring material was cut with a knife and peeled of including remains of glue stuck to the surface material. Samples of PUF and carbon cloth were taken with a pair of scissors. All material samples were individually wrapped in aluminium foil and placed in sealed Rilsan bags before transport to the laboratory for analysis. Homogenization, extraction and analysis were done according to Lyng et al. (2016).

Temperatures were monitored in the rooms by Hobo loggers. The loggers were connected to CO_2 sensors from Vaisala. Readings were taken every 10 minutes. Air changes in the rooms

were measured several times repeatedly by dosing CO_2 and using the logged concentrations. PCB concentrations are highly dependent of room temperatures. In order to compensate for this all air concentrations reported are normalized to a reference temperature of 21 ^{o}C according to Lyng et al. (2016). The temperatures used here are the average temperatures for the sampling periods.

3 RESULTS

The air samples taken one year before the experiment were at the same concentration levels as the samples taken the month before introduction of new material. The average concentrations of PCB_{sum7} were 703 ng/m^3 (n=5), 584 ng/m^3 (n=6) and 684 ng/m^3 (n=6) in room 1, 2 and 3 with a coefficient of variation of maximum 11%. Figure 1 shows the air concentrations in each of the three rooms before and after introduction of the new materials at day zero.

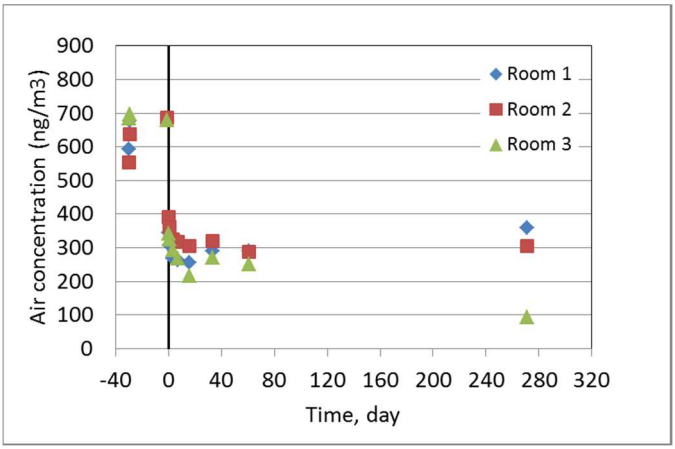

Figure 1. The air concentration of PCB_{sum7} in the three rooms at different days. Zero represent the day of introduction of the new materials.

Initial significant decreases in air concentrations are seen after the introduction of the new materials with room 1, 2 and 3 dropping to respectively 54 %, 63 % and 50 % of previous level of PCB_{sum7}. The next few days the concentrations decrease further in an intermediate period and then in the following levelling period room 1 and 2 concentrations seem constant while room 3 decreases further. Taking the average value of the air concentration of PCB_{sum7} measured day 16, 33 and 61 in the levelling period the concentration in the three rooms are lowered further to a level of respectively 44%, 49% and 36% of previous level of PCB_{sum7}. After 271 days the levels are 56, 49 and 14 % of the concentration before introduction of new materials. It may be noted that the remarkable decrease at 271 days in room 3 caused us to repeat the measurements and the repeated air sample confirmed the result.

Figure 2 shows the concentration of the seven congeners measured in all material samples. PCB-52 is the dominant congener for all samples, followed by PCB-28 for all valid samples. For the old materials, samples of wall paint show a decrease in concentration over time in all rooms. Room 1 and room 3 have a floor cover of carpet and these also show a decrease over time. The floor cover in room 2 is vinyl and no significant change is seen. For the new materials both paint and PUF showed an increase in concentration over time. For carbon cloth an increase is observed at day 34 though the increase does not continue until day 271. This gave rise to suspicion of an inadequate extraction of PCB. A new sample was taken and the time of extraction was prolonged. The concentration was still low and therefore carbon cloth concentrations were all considered invalid.

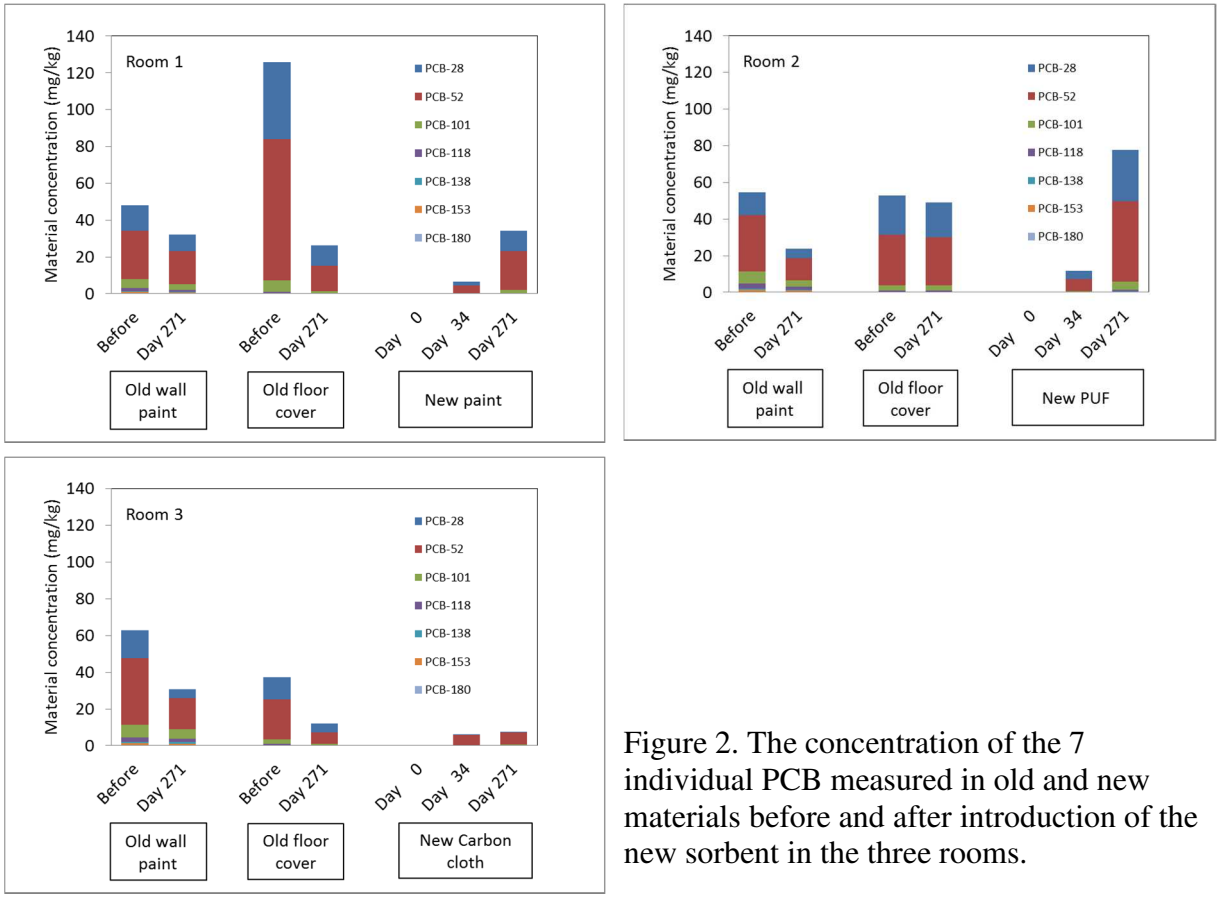

Figure 2. The concentration of the 7 individual PCB measured in old and new materials before and after introduction of the new sorbent in the three rooms.

4 DISCUSSION

After introduction of the new materials the temporal changes in air concentrations reflect changing mass transfer processes with exchange rates dependent on area densities and partition coefficients for the surface materials. The initial drop to about 50 % of previous stable concentrations may be expected according to equation (3).

The decrease in bulk air concentrations in the following intermediate period lasting approximately 1 week is probably the result of reduced availability of PCB for desorption in the old materials. The old paint surfaces may have some PCB molecules with stronger surface bonds and possibly develop a small resistance to emission in the paint film. This period is followed by a levelling period with more stable air concentrations. In this period more significant mass transfer occurs and PCB concentrations in the air immediately above the material surfaces tend to equilibrate causing PCB affinity and area density of surface materials to become increasingly important.

In room 1 with new painted surfaces, the PCB_{sum7} in old paint have been reduced to 67 % of initial concentration. The PCB_{sum7} paint concentrations in the room have almost equilibrated within the 271 days. In room 2 with PUF the PCB in old paint was reduced to 43 % of initial concentration. The PCB concentration in PUF became much higher than initial paint concentration indicating a high affinity to PCB. In room 3 with carbon cloth the PCB concentration in old paint was reduced to 49 %.

Looking at the congener composition it may be seen that the volatile congeners PCB-28 and PCB-52 have higher concentrations in the new materials compared to the old while the less volatile congeners have 2-8 times lower concentrations in the new materials. It could therefore

be assumed that PCB affinity to the investigated new materials is slightly higher but the equilibrating process is much slower for the less volatile congeners. The PCB concentrations in the carpets are significantly reduced compared to the vinyl flooring. The fluffy carpets may have weak bonding to PCB and an immediate response to changes in air concentration. The complete mass balance of PCB in the surface materials of the room include some removal of PCB with ventilation and some further PCB emission from PCB containing sealants acting as primary sources. The effect of these mass flows may however in the time frame of the experiment be small. Assuming 300 ng/m^3 of PCB_{sum7} in room air and an air change rate of 0.15 h^{-1} the amount of PCB removed by ventilation in room 1 during 271 days is 5.4 mg while the adsorbed amount of PCB in new paint is 443 mg.

5 CONCLUSIONS

Introduction of material with adsorbing properties and a surface area comparable to the contaminated surfaces of a room can act as a fast way of reducing the air concentration of PCB as well as way to decontaminate old surface materials in the time frame of months to years. The rate of mass transfer from old to new materials may exceed PCB removal by ventilation by orders of magnitude. For practical applications of these results the PCB affinity and sorption capacity of the new materials are key issues since the thick old layers of paint and other surface materials may cause equilibrium with new materials with limited sorption capacity at PCB concentrations exceeding acceptable levels for occupancy. Slow desorption caused by introduction of new materials may be a promising decontamination method with economic advantages compared to faster methods including removal of surface coatings. Further validation and optimisation of methods are recommended.

6 ACKNOWLEDGEMENT

This study has received financial support from Realdania Foundation and The Danish Transport and Construction Agency.

7 REFERENCES

Danish Environmental Protection Agency 1983. *PCB/PCT pollution. An investigation on consumption, pollution and transport pathways of PCBs and PCTs in Denmark. (in Danish)* Lyngby: COWIconsult. Danish Environmental Protection Agency, Copenhagen.

Frederiksen M., Meyer H. W., Ebbehøj N. E. & Gunnarsen L. 2012. Polychlorinated biphenyls (PCBs) in indoor air originating from sealants in contaminated and uncontaminated apartments within the same housing estate. Chemosphere, 89(4), 473-479.

Gunnarsen L. & Kolarik B. 2014. PCB in sealant, concrete, paint and lacquer 40 years after use of sealant with PCB – calculation of total remaining mass in a contaminated dwelling. Healthy Buildings 2012: 10th International Conference. Brisbane, Australia : Queensland University of Technology, 2012.

Lyng N.L., Gunnarsen L. and Andersen H.V. 2015. The effect of ventilation on the indoor air concentration of PCB: An intervention study. *Building and Environment 95*: 305-312.

Lyng, N. L. 2016. Emission of PCBs in Contaminated Buildings: Field investigation of the impact of ventilation and temperature and development of an emission test cell for in situ measurements. PhD thesis, Aalborg University, link: 10.5278/vbn.phd.engsci.00077

Lyng, N.L., Clausen, P.A., Lundsgaard, C., & Andersen, H.V. 2016. Modelling impact of room temperature on concentrations of polychlorinated biphenyls (PCBs) in indoor air. *Chemosphere 144:* 2127-133.

Weschler C. J. and Nazaroff W. W. 2008. Semivolatile organic compounds in indoor environments. Atmospheric Environment, 42(40), 9018-9040.

World Health Organization 2000. Air quality guidelines for Europe (Second Edition).