Chemosphere 179 (2017) 101–111

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Laboratory investigation of PCB bake-out from tertiary contaminated concrete for remediation of buildings

Barbara Kolarik [a, b, *], Helle Vibeke Andersen [a], Marie Frederiksen [a, c], Lars Gunnarsen [a]

[a] Department of Energy, Environment and Indoor Climate, Danish Building Research Institute, Aalborg University, A.C. Meyers Vaenge 15, DK-2450 Copenhagen SV, Denmark
[b] Danish Environmental Analysis (Dansk MiljøAnalyse), Skelstedet 5, DK- 2950 Vedbaek, Denmark
[c] National Research Centre for the Working Environment, Lersø Parkalle 105, DK-2100 Copenhagen Ø, Denmark

HIGHLIGHTS

- Painted and sandblasted samples of PCB contaminated concrete either exposed to 65 °C or 50 °C for 20 or 10 days.
- Significant impact of temperature and duration on PCB removal based on air results.
- Significant decrease of PCB concentration only in the paint fraction.
- No significant effect of bake-out on fractions of the sandblasted samples.
- Different types of the same material might behave differently under bake-out.

ARTICLE INFO

Article history:
Received 5 January 2017
Received in revised form
21 March 2017
Accepted 22 March 2017
Available online 25 March 2017

Handling Editor: J. de Boer

Keywords:
Polychlorinated biphenyls
Mitigation
Thermal stripping
Indoor air
Building materials
Tertiary sources

ABSTRACT

Polychlorinated biphenyls (PCBs) have been used in flexible construction products from the 1950s to the 1970s. Despite a relatively low vapour pressure, PCBs have over the years emitted to the indoor air and adsorbed to other surfaces, creating tertiary sources. While necessary to protect the environment and health of building occupants, remediation of the contaminated buildings is an expensive and difficult process. The aim of this study was to investigate the effect of bake-out as a method for removing PCB from contaminated indoor surfaces. Four painted and four sandblasted samples of concrete wall were kept at 50 or 65 °C for 10 or 20 days in small scale chambers, which were ventilated with clean air at area specific rates comparable to full scale rooms. The air concentrations were measured several times during the bake-out. Concentrations of PCB in paint, plaster and concrete were measured before and after the experiment. For the painted samples, PCB28 and PCB52 followed by PCB138 and PCB153 were the predominant congeners measured in air. A significant reduction of the PCB concentration was only found for more volatile congeners in paint, while no significant effects were observed in the plaster or concrete. For the most effective setting (65 °C, 20 days), the concentration of $PCB_{\sum 7}$ in the paint was reduced by 40%. For the sandblasted samples, the less volatile congeners PCB138 and PCB153 were predominant in the air during bake-out. No effect was observed on the initially low concentrations of the sandblasted samples, possibly due to analytical limitations.

1. Introduction

Polychlorinated biphenyl (PCB) is a group of chemicals, having 1–10 chlorine atoms replacing hydrogen in the biphenyl ring. Due to the physico-chemical properties PCB has been favourable for many technical purposes. Since the 1950s PCB was used in closed applications e.g. dielectric fluids, cooling or hydraulic liquids and in open applications like flexible construction products. Due to increasing evidence of adverse environmental and health effects, open applications were banned in many countries in the 1970s (Diamond et al., 2010) and closed applications in the 1980s (Breivik et al., 2002). High concentrations can still be found in indoor air in many buildings erected or renovated in this period both from

---

[*] Corresponding author. Danish Environmental Analysis (Dansk MiljøAnalyse), Skelstedet 5, DK- 2950 Vedbaek, Denmark.
E-mail address: bk@dma.nu (B. Kolarik).

http://dx.doi.org/10.1016/j.chemosphere.2017.03.094
0045-6535/

remaining open applications and leaky closed applications. Due to the relatively low volatility and high chemical stability of PCB, it both migrates from the original primary sources to adjacent materials and evaporates to the indoor air and deposit to other indoor surfaces including concrete (Jartun et al., 2009; Pizarro et al., 2002) and paint (Andersson et al., 2004; Jartun et al., 2009). As a result, three types of PCB sources can be defined: primary sources (materials in which PCB was originally used), secondary sources (adjacent materials contaminated by diffusion) and tertiary sources (materials contaminated by adsorption from contaminated indoor air) (Kolarik et al., 2016). In a dynamic indoor climate these slowly developed secondary and tertiary sources sometimes act as sources and sometimes as sinks (Liu et al., 2015). Therefore, these sources should not be overlooked in the remediation of contaminated buildings.

There is limited knowledge available regarding the range of possible remediation techniques, including their effectiveness on reducing PCB levels in both materials and air. The removal of primary sources is a straight forward process, however, it is often insufficient for decreasing high indoor air concentrations if used alone (Allen et al., 2011; Sundahl et al., 1999). Recently, our group has shown that the impact of the tertiary sources on the indoor air concentrations can be substantial (Kolarik et al., 2016). In that study, a concentration level of PCB in the indoor air of 859 ng/m^3 (median value) was measured in 83 contaminated apartments more than one year after the primary sources (sealants) had been covered with aluminium tape and wooden strips (Frederiksen et al., 2012). Statistical analyses of the influence of building characteristic and human behaviour on air concentrations of PCB revealed that the ‘area of walls and ceiling to volume ratio’ was the parameter with the strongest impact (Kolarik et al., 2016). These results were in line with results previously reported in two studies, where the importance of tertiary contamination was investigated. In the first study, PCB concentrations were measured in 411 samples of materials collected in PCB contaminated schools. PCB was detected in 93% of these samples, which all were either secondary or tertiary sources (Thomas et al., US EPA, Washington, 2012). The second study was a laboratory investigation, where re-emission of PCB from tertiary contaminated concrete panels was investigated. The authors of that study concluded that although PCB concentration in sink materials usually is low, they often cover large areas and thus may contribute to elevated concentrations in indoor air for a prolonged period after the primary sources have been removed from a building (Guo et al., US EPA, Washington, 2012).

Removal or mitigation of tertiary sources can be an expensive and cumbersome process. Beside source removal, chemical extraction, chemical degradation and encapsulation of the sources, ventilation is also used to minimize exposure to PCB emitted from contaminated building materials (Allen et al., 2011; Brown et al., 2016; Frederiksen et al., 2015). Bake-out, also known as thermal stripping, seems to be a promising method for remediation of tertiary sources with large surface area. The idea of bake-out has its background in the temperature dependency of the vapour pressure (Falconer and Bidleman, 1994; Paasivirta and Sinkkonen, 2009). It has been estimated that a temperature increase from 20 °C to 40 °C increases the vapour pressure of PCB by a factor of 6–13, depending on the congener, with relative higher increase for the less volatile ones. A temperature increase to 65 °C increases the vapour pressure by a factor 40–200 (Falconer and Bidleman, 1994; Paasivirta and Sinkkonen, 2009). Guo et al. (2011) has also shown a temperature dependency of PCB emission and emission factors increasing by a factor of 5–9 for each 10° increase between 10 and 50 °C. Bake-out has been used in a few remediation cases in Denmark; however, documentation of the effects includes air measurements only (Andersen, 2016; Østergaard Haven and Langeland, 2016).

In theory, bake-out would be a straight forward way of removing PCB from surfaces if the emission was solely a surface phenomenon with the rate limiting step being the resistance to mass transport in the boundary layer and assuming an efficient removal of PCB in the bulk air. To our knowledge, this simplified thinking is only valid initially. After some time, the emission will decrease along with the amount of available PCB at the surface and diffusion of PCB from the underlying layers to the surface will become the rate limiting step. Elevated temperature will increase the rate of diffusion in the material, but the impact will be much lower than the impact on the vapour pressure and the partitioning between material and air. It may be speculated, that the increased rate of diffusion might cause a contamination deeper into the material.

In the current study we wanted to explore the bake-out method with practicable temperatures and duration times. We wanted to see if the chosen parameters would stress the emission mechanism to a point, where it was no longer possible to mobilize and emit considerable amounts of PCB from the surface. Therefore, the aim of this study was to investigate the effect of bake-out at different temperatures and duration on tertiary contaminated concrete as well as PCB concentrations in the air.

2. Methods

The experiment was conducted in the laboratory at the Danish Building Research Institute in small scale climate chambers (Chamber for Laboratory Investigation of Materials, Pollution and Air Quality, CLIMPAQ; Fig. S1). The CLIMPAQ chambers are made of glass and have a volume of 51 L. The environmental parameters such as air velocity, air flow, temperature and humidity can be varied independently within ranges typically found in indoor environment, which make CLIMPAQ chambers suitable for emission tests (Gunnarsen et al., 1994; Nordtest, 1998).

The concrete samples used in the present investigation originated from PCB contaminated apartments of a housing estate located in Farum, Denmark. The housing estate was constructed in stages between 1970 and 1974, and PCB was used in the first completed section, approx. 300 out of 1645 apartments. PCB was used in sealants around windows, indoor doorways and in some cases between the concrete panels in inner walls. The air concentration of PCB_{total} in 83 contaminated apartments ranged from 168 to 3843 ng/m^3, with a median value of 859 ng/m^3. The median concentration of PCB in elastic sealants in these apartments was 16.5% (w/w). More details about the housing estate can be found in Frederiksen et al. (2012).

2.1. Sample preparation

The experiment was conducted using concrete blocks originating from painted inner walls considered as tertiary sources, i.e. contaminated by adsorption of PCB from indoor air. The samples were taken in a distance of at least 30 cm from primary sources. The concrete walls were 6 cm thick with a thin layer of plaster and a considerable layer of paint on each side. The paints were of a slightly different colour. One side of the concrete blocks was named “1” and the other side named “2” (Fig. S2). Consequently, the paint layer on side 1 was named “paint 1” and paint layer on side 2 “paint 2”; likewise for the plaster. Six rectangular blocks of concrete (100 cm long x 7 cm wide) were cut from the walls and delivered to our laboratory by a construction company. At the remediation site, half of the blocks had been taken from the original painted wall without any pre-treatment. The other half of the blocks were taken from walls where paint and plaster had been removed by sandblasting at the site as part of the remediation process. Concrete