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## **MIGRATION OF PCBs FROM SEALANTS TO ADJACENT MATERIAL**

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### **SUMMARY**

PCB in sealants can migrate into porous surfaces like concrete and brick and contaminate these materials. The study focuses on an analysis of concentration profiles in relation to alleviation of PCB contamination. The PCB content was determined in 20 samples of concrete and 6 samples of brick adjacent to outdoor PCB-containing sealants. The samples were submitted to the laboratory for normal commercial assessment of PCB content in relation to the handling of waste from renovation works in Denmark. The results showed that the PCB concentration in the sealant not necessarily relate to the concentration of the adjacent material. The profiles of PCB concentration into the materials showed some variation, though in general there is a tendency of relative higher concentration near the primary source in concrete and longer migration at low concentrations in brick. The concentration profiles seemed to fit an exponential function for concrete and a power function for brick. Based on the sample series, a worst case scenario for migration was defined. For concrete, it showed that a concentration of less than 50 ppm in the material (hazardous waste) was reached at a distance of 5 cm, whereas for bricks the distance was 3.5 cm.

### **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 were first recognized as an environmental contaminant in the 1960s (Jensen et al., 1969) and now PCBs are considered as global environmental contaminants (Breivik et al., 2002). Due to their toxicity and persistence, many countries and intergovernmental organisations have now banned or severely restricted the use, transport, handling and disposal of PCBs (WHO, 2000; European Council, 2004). PCBs have been used as plasticiser in building materials including in sealants and were used in Denmark from the 1950s until banned in 1977 (Danish Environmental Protection Agency, 1983). A recent investigation showed that it was most frequently used in sealants in buildings from the period 1965-1974 in Denmark (Grontmij & COWI, 2013).

PCB containing sealants can evaporate PCB to the indoor air, causing concentration levels of great concern regarding human health (e.g. Balfanz et al., 1993;

Frederiksen et al, 2012; Kohler et al., 2005). Over the years PCB from sealants has migrated into adjacent materials creating “secondary sources”, thereby increasing the surface area of contaminated parts. This adsorbed PCB can be re-emitted into the air when the primary sources are removed or reduced (U.S. EPA, 2015; Kolarik et al., 2012). Although the adjacent materials have a low content of PCB compared to the sealants, they often have the emission strength to impact the air quality. During remediation one therefore has to be aware of these secondary PCB sources and the possible needs for removal or remediation to ensure acceptable indoor air quality. If the contaminated building material is removed, it is of great importance to ensure correct handling in relation to both workers safety and proper waste handling. As the interest in reusing building material is increasing, it is also important to be aware of these potential contaminations and ensure, that reuse of bricks or concrete do not introduce contaminating compounds into new buildings or the environment.

To our knowledge, little is known about the transport process of PCBs from sealants into adjacent materials like porous surfaces of concrete or brick. Assumptions of diffusion according to Ficks second law are proposed (Ljung et al., 2002; Liu et al., 2014). Rex et al. (2002) assume the penetration depth to be related to the porosity of the material and type of concrete or brick. Rex et al. (2002) gathered Swedish data on migration of PCBs from sealant (mainly outdoor) to building materials of concrete, brick, wood and lightweight concrete. Although the variations are large, they propose a power function (log-log) relation between the PCB concentration in the material and the distance from the sealant. The results show comparable migration of PCBs in concrete and brick, whereas the migration in wood seems less and in lightweight concrete it is higher.

Chang et al (2002) examined brick and masonry surfaces for PCBs from sealants. With the criterion for an acceptable PCB concentration at 1 mg/kg (ppm) PCB as a limit, they found that the depth of migration was approximately 1-5 cm from the sealant though sometimes the whole brick was contaminated. They conclude that porosity and surface treatment influence the migration. Woodard and Curran (2010) analysed indoor plaster, brick and concrete adjacent to sealants containing PCBs and concentrations greater 1 ppm were detected in the majority of the samples up to a distance of 33 cm.

Grontmij and Carl Bro (2011) reported results from a housing estate in Farum in Denmark. They found a variation in migration, due among other factors to porosity. Concentrations above 50 ppm PCB (hazardous waste) were at distances up to almost 3 cm from the sealant in concrete, whereas they were 1 cm in woodwork and 7 cm in Masonite.

This study focuses on an analysis of concentration profiles of PCBs caused by migration from primary sources.

## **METHODOLOGIES**

In the time period 2008-2012, 20 samples of concrete and 6 samples of brick adjacent to outdoor PCB-containing sealants were taken in order to assess the PCB content in relation to the handling of waste from demolition or renovation works in Denmark. The samples are from buildings built in the period 1950-1977 and were

taken by professionals with attention to minimising cross contamination from tools or sealants. No information is available on the structure of the material or impacts like weathering. Samples of concrete were drilled out as a cylindrical core, cut out with angle cutter or chiselled out. The brick samples were chiselled out. The samples were submitted to the laboratory. At the laboratory the first 1.5 mm of the surface was removed due to residues of sealant. All samples were subsequently cut in pieces of about 1 cm in the direction of the migration of PCBs. The pieces were cut with a concrete saw with diamond blade and measures were taken to avoid heating above 70°C. The pieces were subsequently crushed in a ball mill and extracted with cyclohexane/acetone (1:1 vol/vol). The extracts were analysed by GC-ECD or GC-MS for the content of the PCB congeners PCB-28, PCB-52, PCB-101, PCB-118, PCB-138, PCB-153 and PCB-180. The sum of these 7 congeners is defined as PCBsum7. The concentrations of each subsample are related to the mean of the distance from the sealant, e.g. a sample taken in a distance from 10 mm to 20 mm is related to a distance of 15 mm. A set of concentrations originating from the same sample are defined as a series. 10 of the series of concrete and 6 of the bricks had 3 or more samples and are used for statistical analysis.

## RESULTS AND DISCUSSION

Figure 1 shows the concentration of PCBsum7 in the sample adjacent to the sealant (distance 1.5-10 mm) in relation to the concentration of the sealant. The content of PCBsum7 in the sealants range from 4 ppm to almost 100000 ppm (the latter identified as a Aroclor 1260), whereas the content of the adjacent sample in concrete and brick ranged between 0.016 ppm and 1410 ppm and between 44 ppm and 747 ppm, respectively. For concrete, the PCB content in the adjacent sample ranged from less than 0.1% (9 observations) to 7% of the concentration in the sealant, whereas for brick the content was 0.2-8%. As seen from the figure there is no clear relation between the two variables.

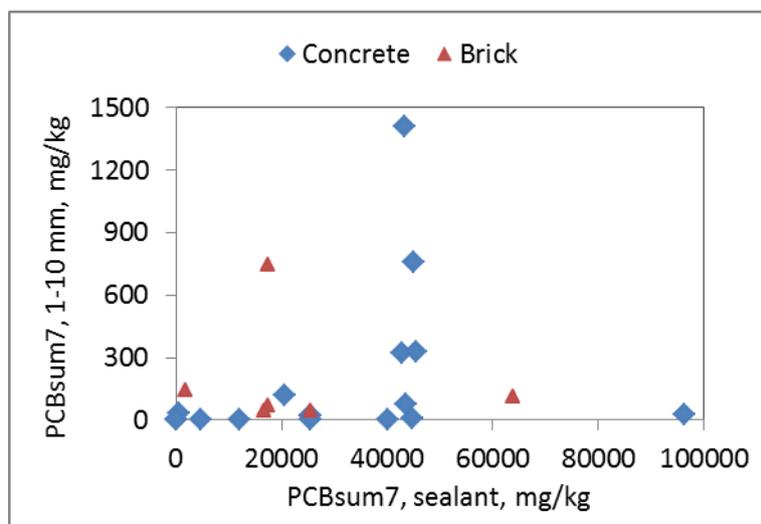


Figure 1. Concentration of PCBsum7 in the sample adjacent to the sealant (distance 0,1-1 cm) versus the concentration in the sealant.

Figure 2 shows the concentration of PCBsum7 for concrete and brick, respectively, versus the distance to the sealant for all samples. Defining the total concentration of

all PCB congeners in a sample as 5 times PCBsum7, 10 ppm PCBsum7 corresponds to a concentration of 50 ppm PCB total, defined as hazardous waste.

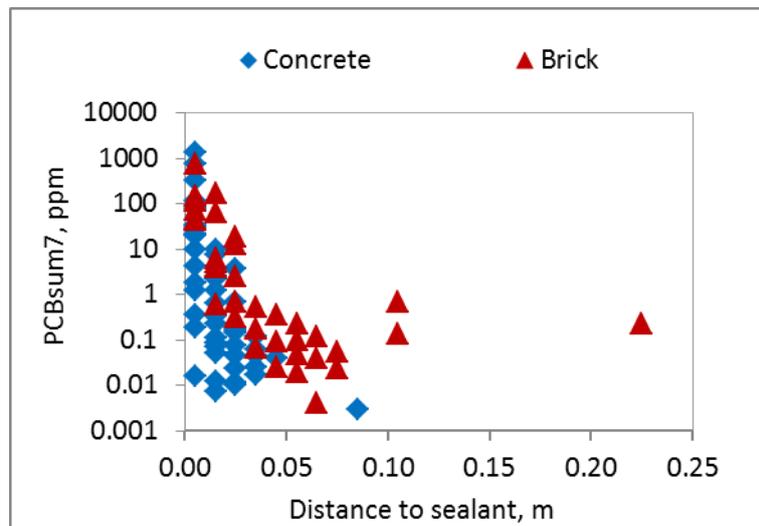


Figure 2. Concentration of PCBsum7 in concrete or brick in relation to the distance to the sealant.

Two values in one series for brick (at a distance of 0.105 m and 0.225 m) have a composition of the seven congeners that do not match the other samples in the series. This indicates that these samples are contaminated from another source and therefore the samples were excluded from further analysis. In general, concentrations show a large variation, though with decreasing concentrations at an increasing distance to the sealants. The decrease is steeper for concrete than for brick.

Figure 3 shows the concentration (log) of PCBsum7 versus the distance from the sealant for the 10 series of concrete with three or more samples. The linear regression lines are shown. The regression coefficient  $R^2$  varies from 0.79 to 0.98.

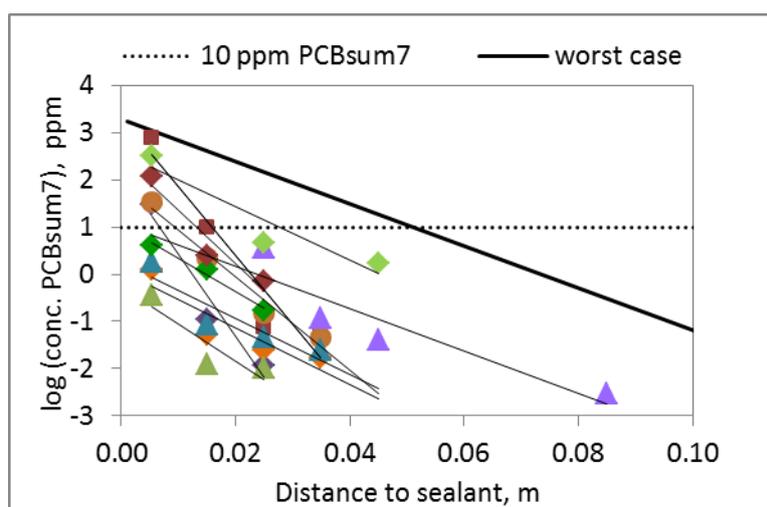


Figure 3. The concentration (log) of PCBsum7 versus the distance from the sealant for the 10 series of concrete with 3 or more samples. Linear regression lines together with a worst case scenario (see text) are shown.

On the basis of these data sets, a worst case scenario for migration of PCB into concrete is made combining the intercept of the regression line giving the highest concentration in the surface and the deepest penetration represented by the regression line with the smallest slope (numeric). This worst case shows that at a distance of approximately 5 cm, the concentration drops below 50 ppm PCB (10 ppm PCBsum7).

Figure 4 shows the concentration (log) of PCBsum7 versus the distance (log) from the sealant for the series of brick with 3 or more samples. These data series show a better fit to the logarithmic distance than the linear distance as observed for the concrete. This behaviour is caused by the sample closest to the sealant. The linear regression lines are shown. Apart from the series marked with green triangles and a green regression line having a poor correlation (the regression coefficient  $R^2$  is 0.25), the other series have a  $R^2$  of 0.93 or better.

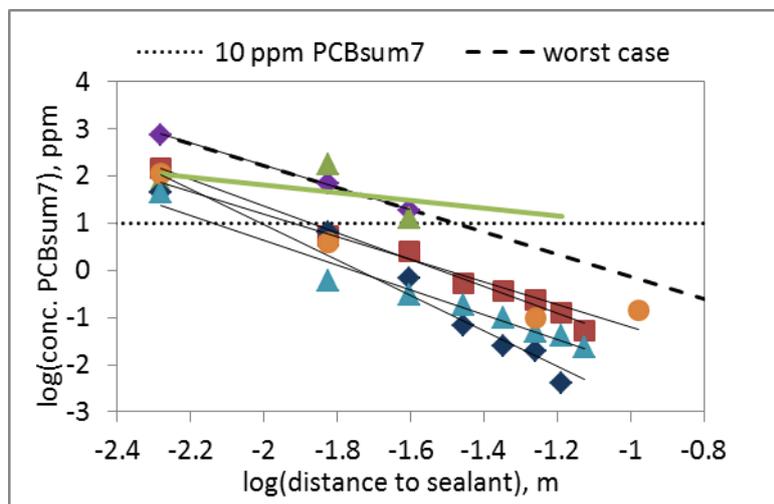


Figure 4. The concentration (log) of PCBsum7 versus the distance (log) from the sealant for the 6 series of brick with three or more samples. Linear regression lines together with a worst case scenario (see text) are shown.

As for concrete a similar worst case scenario is made for brick, though one data set has been excluded (Figure 4, green triangles and line) as this data set has a poor relation. For the remaining five data sets, a worst case scenario becomes equal to one of the data sets (Figure 4, purple rhombus). This worst case shows that at a distance of approximately 3.5 cm the concentration drops below 50 ppm PCB (10 ppm PCBsum7).

The worst case scenarios are rather rough as very little is known about the origin of the samples. Further a conversion factor of 5 between PCBsum7 and total amount of PCBs are used despite that this might lead to an overestimation for the highly chlorinated PCB products, though taking all the other uncertainties into account, this is ignored. To compare the worst case scenarios, Figure 5 shows the scenarios together with the relation for diffusion of PCB into concrete set up by Ljung et al. (2002), the relation for migration of PCBs into concrete and brick found by Rex et al. (2002) and observations made in the Farum housing estate on the indoor migration of PCBs in concrete (Gunnarsen & Kolarik, 2012). The relation set up by Ljung et al. (2002) requires a surface concentration and a period of time together with the diffusion coefficient. In this example, the time is set to 40 years and the surface

concentration to 3% of a sealant having a concentration of 10% (w/w) and with PCBs deposited in the first 2 mm of a concrete having 6% cavity. Ljung et al. (2002) found a mean diffusion coefficient of  $3 \cdot 10^{-15} \text{ m}^2/\text{s}$  (ranging from  $1.7 \cdot 10^{-15} \text{ m}^2/\text{s}$  to  $3.5 \cdot 10^{-15} \text{ m}^2/\text{s}$ ), assuming isotropic material and a diffusion independent of the concentration. The mean diffusion coefficient is used here, though the curve does not change significantly taking the other diffusion coefficients from their interval. For a comparison, the worst case based on the present data set for concrete is also presented for a sealant with 10% (w/w) PCB, giving a surface concentration of 3% like for the calculation made on the basis of Ljung et al. (2002). The relations in Rex et al. (2002) are reported as power functions with normalisation to sealant content of PCBs of 10% (w/w). The relation for concrete is  $C(x)=0.0027 \cdot x^{-2.1242}$  and for brick  $C(x)=0.0007 \cdot x^{-2.4564}$  with  $C(x)$  as the concentration in the material (ppm PCB total) and  $x$  as the distance from the sealant (m). The relation from the Farum housing estate is based on a series of concrete samples adjacent to an indoor sealant and the relation was found to be  $C(x)=7500 \cdot e^{-181x}$  (Gunnarsen and Kolarik, 2012).

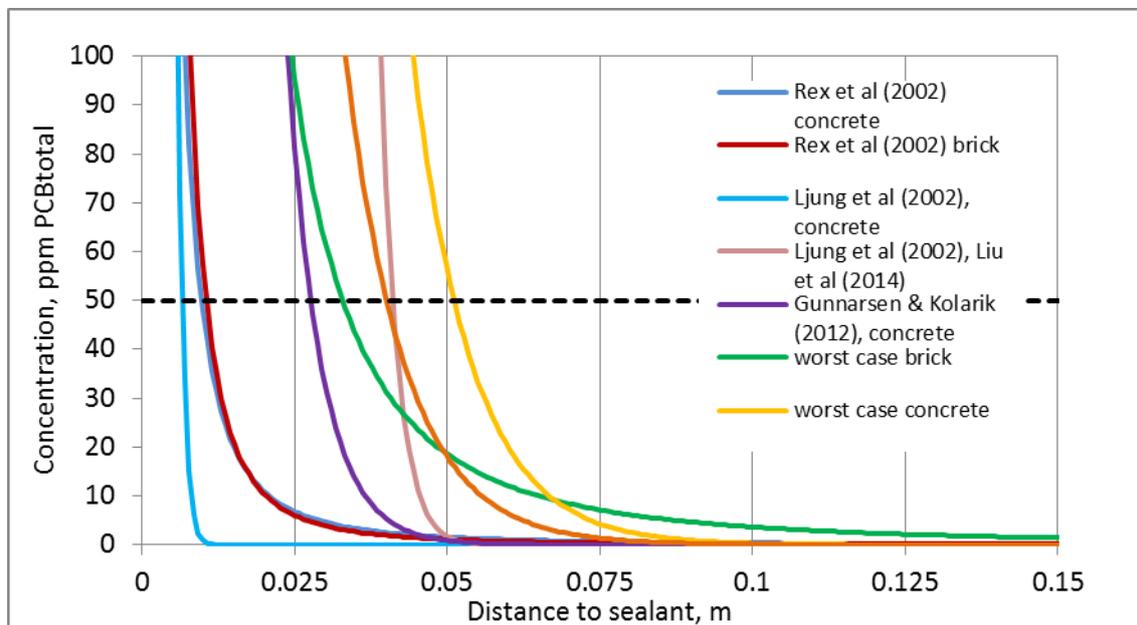


Figure 5. Concentration pattern for PCBs (ppm total) for relations proposed by Ljung et al. (2002), Rex et al. (2002), Gunnarsen & Kolarik (2012) and worst case scenarios, see text. The concentration of 50 ppm (hazardous waste) is shown.

It is seen in Figure 5 that the worst case scenario for concrete is comparable with the findings of Ljung et al. (2002), Rex et al. (2002) and Gunnarsen & Kolarik (2012) and in relation to these findings it defines a distance from the sealant that ensures a material content of PCBs below a 50 ppm PCB total in the concrete. This is also the case, if the worst case scenario for concrete is assumed to represent a sealant with a concentration of 10% (w/w) PCB. The worst case scenario for brick is comparable to the finding of Rex et al. (2002) and fulfils the purpose of estimating a safety distance compared with this.

As seen from Figure 5, the migration of PCB into brick compared with concrete for the worst case scenarios shows that the concrete reach a concentration level of 50 ppm PCB total at a larger distance to the sealant. This pattern changes at concentrations below 10 ppm PCB total, where the brick has a higher PCB content

than the concrete. For concrete, this worst case relation is smaller for the lower concentrations than the finding of Woodard and Curran (2010) having a concentration of almost 3 ppm PCB at a distance of 30-33 cm from an indoor sealant. We have no explanation for this, though we speculate whether this finding can be influenced by the structure of the concrete and/or by PCB migrating from a contaminated surface along the wall. For brick, Chang et al. (2002) and Woodard and Curran (2010) found concentration levels of 1-2 ppm PCB total at a distance of 10-12.5 cm from the sealant. The worst case scenario was above this concentration level.

## CONCLUSIONS

The PCB concentration in the sealant has poor relation to the concentration of the adjacent concrete or brick. The migration of PCB showed some variation in the profiles of concentration into the materials, though in general there is a tendency of a relative higher concentration near the primary source in concrete and longer migration at lower concentrations in brick. The concentration profiles seemed to fit an exponential function for concrete and a power function for brick. A worst case scenario based on the present data sets shows that a concentration of less than 50 ppm PCB total (hazardous waste) in the material is obtained at a distance of more than 5 cm from the sealant in concrete and 3.5 cm in brick. The results for the worst case scenarios seem realistic compared with other findings of migration of PCBs into concrete and brick.

## ACKNOWLEDGEMENT

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