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Painted surfaces – Important sources of polychlorinated biphenyls (PCBs) contamination to the urban and marine environment

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Paint from structures built during the period 1950–1970 may be the most important source of PCBs in an urban environment.

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ABSTRACT

A study of a large number of samples of flaking old paint from various buildings in Bergen, Norway ($N = 68$) suggests that paint may be the most important contemporary source of PCBs in this urban environment with concentrations of PCB_7 up to 3.39 g/kg. Twenty-three of the samples were collected from a single building, and the concentrations were found to vary over 3 orders of magnitude. In addition, 16 concrete samples from a large bridge previously coated with PCB-containing paint were collected and separated into outer- and inner samples indicating that PCBs are still present in high concentrations subsequent to renovation. PCBs were found in several categories of paint from wooden and concrete buildings, potentially introduced to the environment by natural weathering, renovation, and volatilization. Consequently, this dispersion may lead to increased levels of PCBs in urban atmospheres, soils, and harbor sediments where high concentrations have resulted in Governmental advice against consumption of certain seafood.

1. Introduction

Polychlorinated biphenyls (PCBs) have been added to a wide range of products and applications, including hydraulic oils, electrical transformers and capacitors, double framed glazing windows, concrete constructions, sealants, and paint (Sundahl et al., 1999; Hellman and Puhakka, 2001; Poland et al., 2001; Andersson et al., 2004; Herrick et al., 2004; Kohler et al., 2005; Shin and Kim, 2006). Emissions of PCBs to air from primary sources, such as the manufacturing and intentional use of PCBs, have been reduced after the ban of PCBs in various products executed during the 1970s and 1980s (Breivik et al., 2002a,b). However, PCBs are susceptible to volatilization from secondary source compartments such as soil, vegetation, water, atmospheric particles, and products containing PCBs with temperature as one of the factors controlling the dispersion of PCBs in the environment (Halsall et al., 1995, 1999; Wania et al., 1998; Breivik et al., 2004). A number of studies of PCBs in the environment have been dedicated to indicate ambient levels, sources, and atmospheric emissions on a regional or global scale (Breivik et al., 2002a,b, 2004; AMAP, 2002, 2003) followed by studies of PCBs in specific environmental media, such as soil, sediments, and water (Meijer et al., 2003; Rossi et al., 2004). Anthropogenic activities have been accredited for most of the PCB levels found in different parts of nature, although some studies have indicated that PCBs are being formed de novo in thermal processes (Schoonenboom et al., 1995; Ishikawa et al., 2007). Particle-bound PCBs, transported from unknown active sources to urban soil and runoff sediments, however, appear to be a major challenge in urban areas (Jartun et al., 2008).

A review of the history of PCB usage in Norway indicated that an estimated total of 1140 metric tonnes of PCBs were used in different applications, such as capacitors, concrete coatings, and double framed glazing windows. The Norwegian environmental authorities indicate that about 155 metric tonnes of PCBs still remain in different applications in Norway today (Fig. 1), of which PCBs in paint is estimated to constitute about 5% (7.75 metric tonnes) (BNL, 2005).

In a study of PCBs in paint from a former nuclear test reactor in Savannah, USA, PCB concentrations exceeding 6% were found in bulk samples of indoor paint (Lowry et al., 1998, 1999). In a remediation project from a former military base in Arctic Canada, concentrations of PCBs as high as 7.4% were found in samples of paint (Poland et al., 2001). The Norwegian Defence Research Establishment determined the contents of selected pollutants in paint from a naval vessel (frigate), and the highest concentration of PCB_7 in interior vessel paint reported in that study was 1.25 mg/kg (Johnsen, 2001). Andersson et al. (2004) found PCBs in samples of

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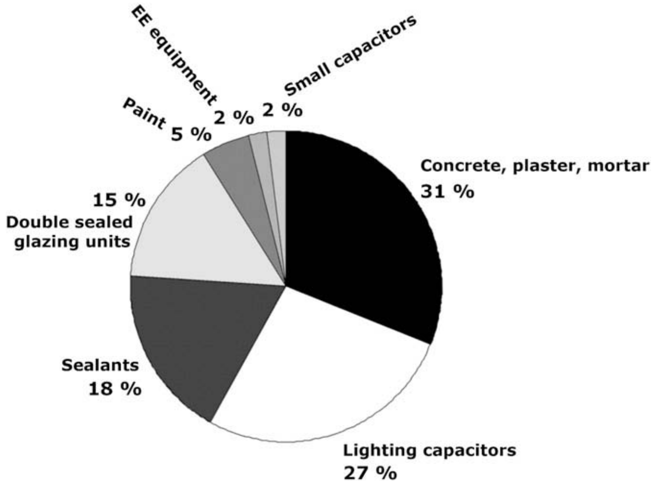

Fig. 1. Remaining polychlorinated biphenyls (PCBs) in products from Norway, shown by percentage of 155 tonnes. Numbers are from BNL (2005) and Norwegian Pollution Control Authority.

building materials, including five samples of paint, from buildings in Bergen, Norway, ranging from <0.001 to 1940 mg/kg. Ruus et al. (2006) observed high levels of PCBs in blue mussels in the Sørfjord, western Norway. The contamination source was removal of old paint and plaster from the façade of an old power station in the area, containing ≥336 mg/kg of PCB_7 in the outermost layer.

Jartun et al. (2008) described the dispersion mechanism of pollutants by urban runoff within small catchments in an urban environment. As a consequence, The Geological Survey of Norway (NGU) initiated a systematic follow-up survey in cooperation with the relevant county and city environmental authorities studying PCBs in exterior paint from building façades within some of these specific catchments. A total of 68 bulk paint samples were collected from the exterior surface of buildings and structures in the city of Bergen, Norway. This paper provides data demonstrating the potential of PCB-dispersion from painted surfaces. Moreover, the study discusses the potential implications of PCB-containing structures for the environmental conditions of marine sediments, the main recipient of urban runoff.

2. Methods

2.1. Sampling

Samples of flaking paint (~10–50 g) from building façades (68 samples) were collected in November 2006 using a small spatula and a knife. About half of the buildings were selected within small catchments in an urban area based on a geochemical map presenting data on PCB_7 in urban stormwater sediments (Jartun et al., 2008). The other half was selected based on criteria such as building age and the degree of surface flaking to provide an overview of the PCB-contamination challenge within the urban area. Most of the buildings were from the period 1950–1970. In addition, 16 drill core samples ($d = 60$ mm, length = 60 mm) from a concrete bridge previously coated with PCB-containing paint (16 samples) were collected and separated into exterior and interior fractions prior to analyses. The paint samples were mostly obtained from concrete structures, but some were also collected from wooden buildings ($N = 7$, sample ID 26, 29, 30, 36, 38, 39, and 51 in Table 2). The variation of PCB concentrations in one single façade was studied by collecting 23 different samples of paint from the same building. This was an attempt to establish how many samples are necessary to reproduce representative concentrations in terms of future remediation and waste disposal. Field duplicates were also collected and analyzed from five locations. The bulk paint samples were stored in small Rilsan® bags prior to analyses performed by the laboratory AnalyCen AS, Moss, Norway in collaboration with the Geological Survey of Norway.

2.2. Chemical analyses

For the determination of PCB congeners, a representative sample of 10 g homogenized material was extracted with 20 mL acetone and 1 mL internal standard of PCB no. 53 (0.10 μg/mL). The extracts were mixed with 50 mL 0.2 M sodium chloride/0.1 M phosphoric acid and 20 mL acetone/n-hexane (1:3). The water phase was removed, and sodium sulphate was subsequently added to remove excess water. The organic phase was concentrated to 1 mL and then cleaned with 1 mL 2-propanol and 1 mL tetrabutylammonium hydrogen sulphate (TBA) with sodium sulphite in excess. Finally the organic phase was cleaned using concentrated sulphuric acid. The quantification of seven PCB congeners (PCB_7: IUPAC-numbers 28, 52, 101, 118, 138, 153, 180) was carried out using gas chromatography (HP6890) with electron capture detection (GC-ECD) with a DB5 122-5062 column of 60 m × 0.25 mm inner diameter and 0.25 μm film thickness. The method is based upon the NORDTEST Technical Report No. 329 (Karstensen et al., 1997). The laboratory is accredited for specific organic analyses, including PCBs, according to European standards NS-EN ISO 17025, NS-EN ISO 9001, and NS-EN ISO 14001, including participation in ring tests and continuous analyses of blanks and lab duplicates. The precision of the analytical method is 15–20%. For quality control a certified reference material purchased from Resource Technology Corporation (RTC) was analyzed regularly. In addition to regular samples unlabelled field duplicates from five different locations were submitted for analysis.

Table 1
Results of PCB concentrations (in mg/L, $N = 3$) in certified reference material (RTC)[a]

Compound	AnalyCen (mg/L)	SD (mg/L)	CV (%)	Certified value (mg/L)	SD (mg/L)
PCB 28	39.5	7.89	20.0	44.0	5.00
PCB 52	34.4	4.51	13.1	38.0	4.00
PCB 101	40.2	1.83	4.6	44.0	4.00
PCB 118	24.3	3.82	15.7	28.0	3.00
PCB 138	36.3	5.16	14.2	27.0	4.00
PCB 153	41.0	6.79	16.5	50.0	4.00
PCB 180	18.5	2.66	14.4	22.0	2.00
PCB_7	238	36.0	15.1	253	26.0

[a] Certified values and standard deviation (SD) of samples from Resource Technology Corporation (RTC) are shown in comparison to the results obtained at the lab AnalyCen (CV = coefficient of variation).

3. Results

Table 1 presents the values of PCB_7 in replicate analyses of the reference materials (RTC) received at AnalyCen compared to the certified values. The results from the field duplicates are considered acceptable, although the reproducibility at low levels is inferior to that observed in the higher concentration range (Fig. 2).

PCB_7 concentrations in 68 samples of paint from the urban environment range from <0.001 to 3390 mg/kg. The median value for all the paint samples is 0.250 mg/kg (Table 2), and a cumulative frequency distribution (Fig. 3) shows that 18% of the collected

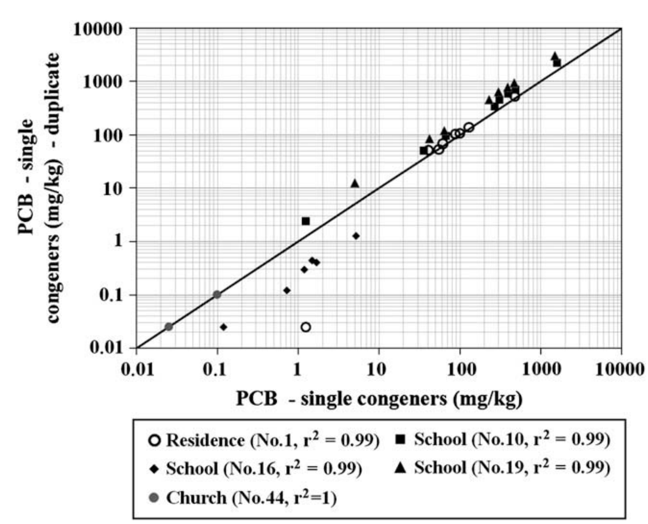

Fig. 2. Results from analysis of lab duplicates expressing the concentrations of the single congeners of PCB_7 (PCB no. 28, 52, 101, 118, 138, 153, 180) in samples of paint ($N = 5$).

Table 2
Analytical results for PCB_7 including the single congeners (PCB IUPAC no. 28, 52, 101, 118, 138, 153, and 180) in 68 samples of paint from buildings in Bergen, Norway, including 23 different samples from the same building complex (sample ID 04–23)

Sample ID	Building category	PCB_7 (mg/kg)	PCB 28 (mg/kg)	PCB 52 (mg/kg)	PCB 101 (mg/kg)	PCB 118 (mg/kg)	PCB 138 (mg/kg)	PCB 153 (mg/kg)	PCB 180 (mg/kg)
01	Residential building	480	<2.5	41	88	62	130	100	55
01-D	Residential building	522	<0.05	51.6	103	68.4	139	106	53.7
02	Industrial building	0.28	<0.05	<0.05	<0.05	<0.05	0.13	0.08	0.07
03	Storage	0.5	<0.05	<0.05	0.07	<0.05	0.18	0.15	0.1
04	School	5.03	0.44	<0.05	0.58	0.11	1.5	1.4	1
05	School	7.6	<0.05	<0.05	0.2	<0.05	0.65	0.52	0.43
06	School	1.27	<0.05	<0.05	0.1	<0.05	0.47	0.38	0.32
07	School	3.6	<0.05	<0.05	0.42	<0.05	1.3	1.1	0.81
08	School	4.4	<0.05	<0.05	0.47	0.11	1.5	1.3	1
09	School	12	<0.05	<0.05	0.55	0.14	1.5	1.3	8.4
10	School	1600	<2.5	36	270	67	490	400	310
10-D	School	2200	2.38	50.1	338	94.8	704	577	445
11	School	1800	<2.5	30	250	66	580	490	420
12	School	440	<2.5	9	68	12	140	120	89
13	School	2.6	<0.05	<0.05	0.39	<0.05	0.86	0.74	0.56
14	School	0.73	<0.05	<0.05	<0.05	<0.05	0.3	0.24	0.19
15	School	0.84	<0.05	<0.05	<0.05	0.21	0.28	0.23	0.21
16	School	5.25	<0.05	<0.05	0.73	0.12	1.7	1.5	1.2
16-D	School	1.26	<0.05	<0.05	0.121	<0.05	0.402	0.439	0.296
17	School	0.71	<0.05	<0.05	0.11	<0.05	0.25	0.21	0.14
18	School	620	<2.5	18	110	34	200	160	95
19	School	1500	5	42	230	64	470	390	300
19-D	School	3030	12.7	85.2	456	120	939	779	640
20	School	60	<2.5	<2.5	9	<2.5	20	18	12
21	School	0.13	<0.05	<0.05	<0.05	<0.05	<0.05	0.13	<0.05
22	School	400	<2.5	9	63	21	130	100	62
23	School	1700	<2.5	31	240	75	550	450	320
24	Industrial	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
25	Industrial	0.388	0.075	0.143	0.076	0.094	<0.05	<0.05	<0.05
26	Industrial	<10	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5
27	Residential	<0.05	<0.05	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05
28	Industrial	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
29	Residential	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
30	Residential	0.299	<0.05	<0.05	<0.05	<0.05	0.125	0.104	0.07
31	Church, NW side	0.212	<0.05	<0.05	0.065	0.065	0.082	<0.05	<0.05
32	Church, S side	20.9	<0.05	3.65	5.01	4.91	4.07	2.8	0.481
33	Industrial	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
34	Industrial	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
35	Industrial	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
36	Residential	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
37	Residential	<10	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5
38	Residential	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
39	Residential	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
40	Residential	<10	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5
41	Residential	0.598	<0.05	<0.05	0.079	<0.05	0.224	0.19	0.105
42	Residential	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
43	Residential	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
44	Church	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
44-D	Church	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
45	Fire station, Building A	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
46	Fire station, Garage	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
47	Old prison building	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
48	Public building	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
49	Wall	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
50	Fire station, Building B	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
51	Fire station, Building C	0.323	<0.05	<0.05	0.066	<0.05	0.133	0.124	<0.05
52	Fire station, Building D	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
53	Fire station, Building E	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
54	Residential	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
55	Industrial, Renovation	<10	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5
56	Industrial, Renovation	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
57	Industrial, Renovation	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
58	Day care center	3390	1.76	553	881	782	681	399	92.3
59	Residential	<10	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5	<2.5
60	Old storage	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
61	Old storage	<0.20	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
62	Storage	0.281	0.229	<0.05	<0.05	<0.05	0.052	<0.05	<0.05
63	Marine dock	12.8	1.42	3.02	2.45	3.63	1.00	0.865	0.404

samples of paint have a PCB_7 concentration of 50 mg/kg or more, which is the hazardous waste concentration limit in Norway (Norwegian Legislation, 2004). The congener profiles in samples with detectable amounts of the individual PCB_7 congeners vary, but with a dominant contribution of mid- to high-chlorinated congeners probably reflecting different types of PCB mixtures originally added to the paint. When compared with standard profiles from different technical mixtures of PCBs derived from Konieczny and

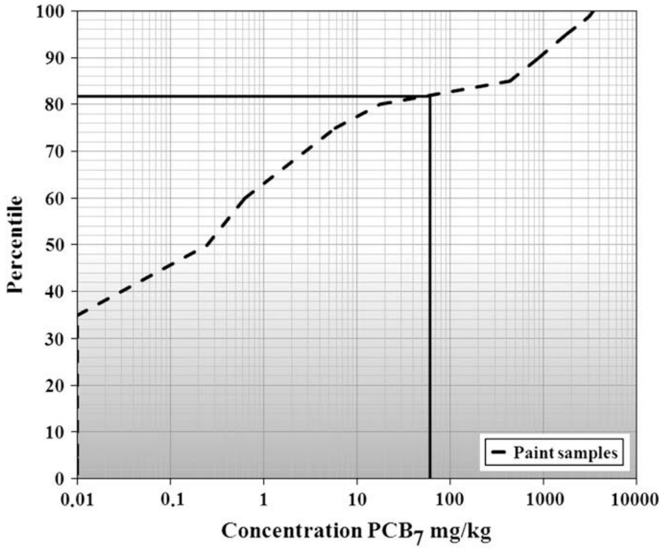

Fig. 3. Cumulative frequency distribution of PCB_7-concentration in 68 samples of paint from buildings and structures within an urban environment. Note the logarithmic scale on the abscissa. Bold line indicates the concentration of 50 mg/kg PCB_7, representing the lower limit of hazardous waste in Norway.

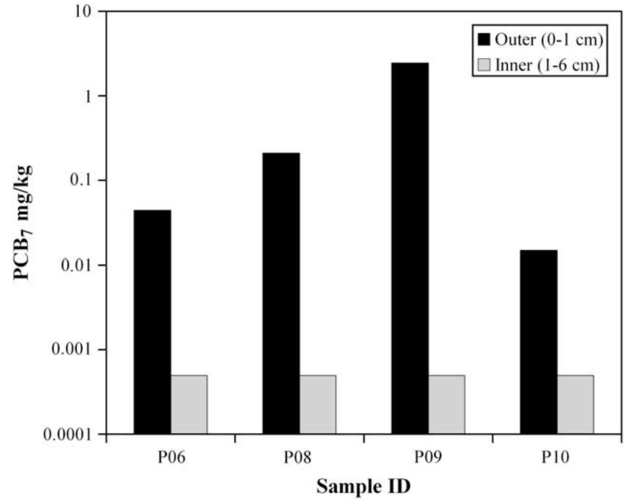

Fig. 4. Variation of surface (0–1 cm) and inner core (1–6 cm) measurements of PCBs in four samples of concrete from a bridge previously coated with a PCB-containing paint.

Mouland (1997), the profiles from PCBs in paint exhibit patterns similar to the common PCB mixtures Aroclor 1254, Aroclor 1260, Clophen A60, or Kanechlor KC-500. The origin of the PCB added to various formulas of paint is, however, difficult to identify as the mentioned technical mixtures derive from USA, Germany, and Japan, respectively. In addition, the degree of degradation of individual congeners remains unknown in this study.

Results for PCBs in samples from the façade of Puddefjordsbroen bridge are given in Table 3. The concentrations of PCB_7 in the samples of concrete ranged from <0.001 to 53.3 mg/kg, with a median value of 0.010 mg/kg. The amounts of PCB_7 found in the samples from the bridge ($N = 16$) may be divided into three categories; (a) not detectable (<0.001 mg/kg, $N = 5$), (b) traces (0.006–0.044 mg/kg, $N = 5$) and (c) high (0.210–53.3 mg/kg, $N = 6$). Fig. 4 indicates the variation between the inner core (concrete) and outer core (paint and plaster) in terms of PCB_7 concentration.

The variation of results from the school building sampled in detail (sample IDs 04–23, Table 2) very clearly demonstrates the need for collecting more than one sample to classify building materials into different categories of waste. The concentration range of PCB_7 in all samples of paint from this particular school is <0.001–3032 mg/kg, with a median value of 7.6 mg/kg. A cumulative frequency distribution of these samples from the school is given in Fig. 5. The curve displays two distinct regions with steeper slope than the rest, each reflecting a narrow range of PCB_7 concentrations for a relatively high percentage of samples. The first steep region reflects PCB_7-concentrations approaching the median (50 percentile) value of 7.6 mg/kg. Another steep region is indicated from PCB_7-concentrations of about 1000 mg/kg (70th percentile) to the maximum concentration of 3032 mg/kg. This distribution pattern indicates two separate sources of PCB from this location, as confirmed by visual observation of two different paint types during sampling. The coefficient of variation (CV) calculated from all samples ($N = 23$) collected from the school is 151%, demonstrating a large variation in the data set. CV from samples of the grey paint ($N = 13$) and yellow paint ($N = 10$) were 98% and 70%, respectively, indicating smaller variation within these individual sources. The relative contribution of the single PCB_7 congeners is similar for these two sources, indicating that the same type of PCB product was used on the façade, although one of the sources contains much higher amounts of PCB than the other. Whether this is a result of dilution or contamination of one painted surface area by the other in this particular case is beyond the scope of this study.

Table 3
Concentrations of PCB_7 and individual congeners in 12 core samples from the Puddefjordsbroen bridge case study (samples no. P01–P12), including four outer-inner core studies

Sample no.	PCB_7 (mg/kg)	PCB 28 (mg/kg)	PCB 52 (mg/kg)	PCB 101 (mg/kg)	PCB 118 (mg/kg)	PCB 138 (mg/kg)	PCB 153 (mg/kg)	PCB 180 (mg/kg)
P-01	0.452	<0.001	0.06	0.103	0.114	0.105	0.059	0.011
P-02	0.363	<0.001	0.036	0.076	0.087	0.099	0.054	0.011
P-03	0.007	<0.001	<0.001	0.002	0.002	0.002	<0.001	<0.001
P-04	0.006	<0.001	0.003	0.002	<0.001	<0.001	<0.001	<0.001
P-05	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
P-06_outer	0.044	<0.001	0.007	0.011	0.01	0.01	0.006	<0.001
P-06_inner	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
P-07	0.008	<0.001	<0.001	0.002	0.002	0.002	<0.001	<0.001
P-08_outer	0.21	<0.001	0.03	0.049	0.051	0.05	0.026	0.004
P-08_inner	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
P-09_outer	2.46	<0.001	0.338	0.575	0.599	0.592	0.301	0.055
P-09_inner	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
P-10_outer	0.015	<0.001	0.002	0.004	0.003	0.004	0.002	<0.001
P-10_inner	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
P-11	3.88	<0.001	0.39	0.864	0.94	1.07	0.522	0.097
P-12	53.3	0.155	8.23	13.5	12.6	11.2	6.15	1.11

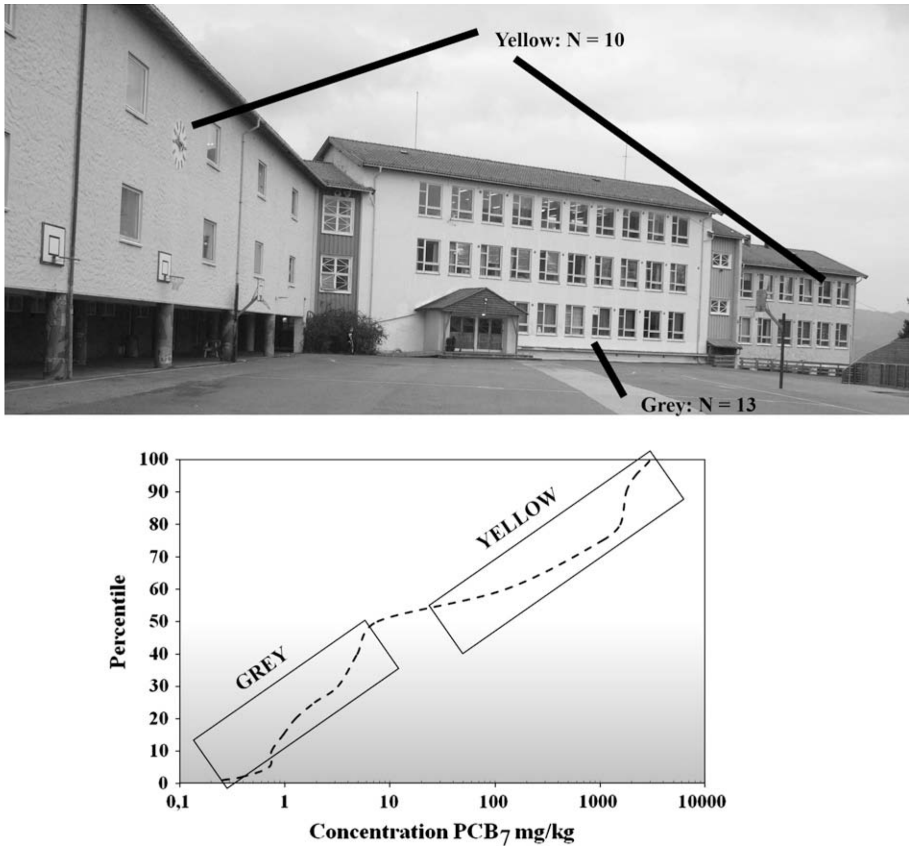

Fig. 5. PCBs in 23 samples of paint from a single school building façade. Two different types of paint show different ranges of PCB_7-concentration given in the cumulative frequency distribution. Note the logarithmic scale on the abscissa.

4. Discussion

4.1. PCB in paint

Building materials contain a variety of chemical compounds (Andersson et al., 2004; Herrick et al., 2004). Polychlorinated biphenyls (PCB) are potentially harmful to both man and the environment, and because of their intense use in a variety of products worldwide there is a need for detailed studies to prevent human exposure and further dispersion to the environment. Exterior walls constitute large surface areas, and are exposed to extreme weather conditions especially in regions heavily influenced by intense rain and alternating temperature. Empirical data on possible sources and the release of harmful substances from building materials to the exterior and interior environment is scarce, and has often been neglected in environmental assessment methods. Rehabilitation or demolition of buildings has become a major environmental challenge.

Our work to establish an overview of the challenge concerning PCB-containing paint within urban areas has documented extensive use of PCB applied to wooden and concrete buildings, and to other concrete constructions as well. Paints, or surface coatings in general, are mixtures mainly consisting of the following ingredients: binder, liquid, pigment, and additives. The binder is a polymer or a resin, which provides the basis of the continuous paint film by adhering the pigment particles together (Lambourne and Strivens, 1999). Because of their protective purposes, paints are required to resist attacks by acids, alkalies, and oxidation, and to have a low permeability to water in order to resist biological, physical, and chemical decomposition (Martens, 1968). The protective requirements of paints resulted in extensive use of additives that may be potentially toxic, such as PCB, tributyltin (TBT), lead, mercury, and cadmium (van Alphen, 1998). Plasticizers were added to different formulas of paint to prevent brittleness by enhancing the physical and chemical resistance of the paint. The plasticizer would increase the durability, and give the paint formula an improved chemical and thermal resistance. Chlorinated paraffins and chlorinated diphenyls were the most popular plasticizers in the 1960s (Martens, 1968). A content of up to 25% pure PCB was added to some formulas, and the most important use was for military, maritime, and industrial purposes, such as airplane paint and iron pipe coatings, mostly as chlorinated rubber paint. Other applications of paint with chlorinated plasticizers were road line paints, corrosion resistant coatings, and mold resistant coatings. PCBs were also added in large amounts to swimming pool paints (7.3%), polyurethane coatings (11%) and ethyl cellulose lacquers (3.2%) (Bennet, 1941, 1967). The addition of chlorinated diphenyls as plasticizers to paint was already reported over 60 years ago (Hadert, 1940), and was probably used on a global scale until prohibited in most countries from the end of the 1970s to the beginning of the 1980s (e.g. Norwegian Legislation, 2004). According to the paint industry, PCBs were exclusively added to chlorinated rubber paint in Norway. However, in our study PCBs were found in paints with an organic binder (pliolite type, acryl/pliolite type), single component silica paint (sodium or potassium silicate), and cement based paint. This indicates that it is impossible to know in advance what kind of paint may contain PCBs. Classification of paint that was applied on a surface over 30 years ago is a difficult task. Thus it was not possible in this study to directly relate the high concentrations of PCBs in samples of paint to any specific type of binding material. The paint itself does not constitute a large volume of contaminated waste. However, large surface areas should be a major concern in terms of contamination of soil, water, and air.

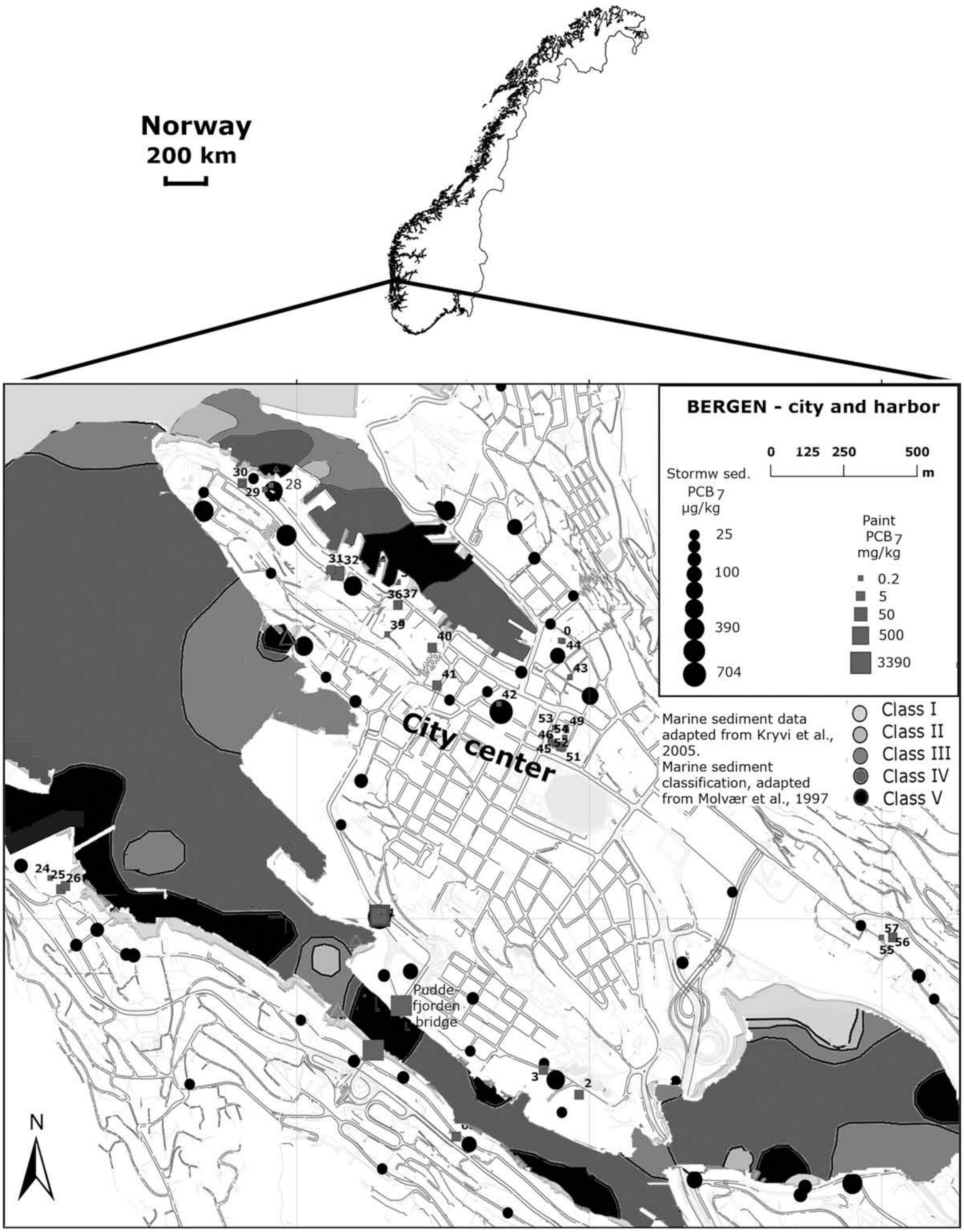

Fig. 6. Map of the study area showing the environmental condition of marine sediments based on the concentration of PCB_7 (Molvær et al., 1997; Kryvi et al., 2005), the locations of stormwater sediments studied by Jartun et al. (2008), and locations of PCB in paint (this study). The location of the school (Fig. 5) is slightly outside this map projection and not included here. For detailed description of the study area, see Jartun et al. (2008).

This appears to be especially important in areas burdened with a wet coastal climate facilitating continuous weathering of surface materials.

4.2. PCB-containing paint – important implications

Flaking paint may constitute a major source of PCBs in an urban environment, especially in areas with a coastal climate such as Bergen, Norway. Chips of paint may easily scour off from surfaces during heavy rainfall and wind, and, subsequently, impervious surfaces will facilitate a particle-bound dispersion through the urban environment via the stormwater sewage system. It may consequently be a significant source of marine sediment contamination. High concentrations of PCB_7 found in stormwater sediments indicated an active and ongoing dispersion of PCBs from the contaminated surface to the stormwater system (Jartun et al., 2008).

Fig. 6 is a map showing the Bergen harbor area with the environmental classification of marine sediments adapted from Molvær et al. (1997) and Kryvi et al. (2005). In this classification class I is defined as insignificantly polluted ($PCB_7 < 5$ μg/kg), and class V as very strongly polluted ($PCB_7 > 300$ μg/kg). Generally, the concentration of PCB_7 in the marine sediments is high, but there are several hotspots indicating a local contamination source, which may be paint. The dispersion is facilitated by stormwater runoff (Jartun et al., 2008). Renovation of buildings by sandblasting or power washing will bring about an additional dispersion of PCBs from contaminated façades in an urban area, which may consequently affect remediation of contaminated marine sediments (Kryvi et al., 2005).

Concentrations of PCB_7 in the 16 surface layer samples from Puddefjordsbroen bridge varied from <0.001 to 53.3 mg/kg, with large variations between outer- and inner-fractions (Table 3). Old information on the maintenance of the bridge indicated that it was covered with a white layer of paint when built in 1956, and then left untreated until completely sandblasted in the mid-1980s. There are no reliable records available on the composition of this paint, but it was probably chlorinated rubber paint with good adhesive properties. Old formulas indicate that such paints could have a PCB concentration of at least 8–9% (Martens, 1968; Bennet, 1941, 1967). The sandblasting carried out in the mid-1980s is probably why the concentration of PCBs was high on certain parts of the bridge and low on others. The total surface area of the bridge covered with paint is 11 000 m^2. Assuming a 2 mm thick layer of paint, a total of 22 000 l of paint would have been added to the bridge surface. Furthermore, a PCB concentration of 5% in the old paint and a general density of chlorinated rubber paint of 1.5 g/cm^3, would imply a total amount of 1650 kg PCBs added to the surface of this bridge. These are just assumptions, but they indicate that a large amount of PCBs has been mobilized to the local environment. Based on the results in Table 3, the estimated remaining amount of PCBs in the bridge surface is about 0.2–0.3 kg. Accordingly, marine sediments below the bridge are heavily contaminated with PCBs, both in surface sediment and in deeper parts down to about 40 cm (Kryvi et al., 2005). However, other factors such as tidal currents, whirl up from boat traffic, and sedimentation from various sources makes it difficult to quantify the actual contribution of PCBs from the renovation of the bridge to the underlying marine sediments. This is just an example of how important paints may be in terms of releasing pollutants to the local environment. Most probably there are numerous other concrete structures that have been, or still are, covered with PCB-containing paint. This is an important issue to address for the future in terms of pollution control.

It is also evident that a single sample from a specific building is not enough to fully identify the extent of possible PCB contamination. The school façade that was studied in detail ($N = 23$), had different layers of paint at different heights and depths, all contributing to a mixture of paint and plaster layers. One of these layers contained up to 3000 mg/kg of PCB_7, which obviously qualifies as hazardous waste. This implies that when a building is up for renovation in the future, contractors and owners must be aware of the actual situation before a proper waste plan can be executed. Crude estimates based on the assumptions for the bridge mentioned above, and the results presented in Table 2, about 3.0 kg PCB_7 still remains in the surface of this building.

The importance of PCBs in paint becomes even more comprehensible when considering the indoor environment and exposure to humans by gases and dust. Other studies have shown that humans can be exposed to relatively high concentrations of PCBs from indoor sources (Currado and Harrad, 1998; Rudel et al., 2003; Harrad et al., 2006; Kuusisto et al., 2007). The contribution of PCBs to air from painted surfaces, both exterior and interior, should be further studied in known hotspots, such as the residential or school buildings found in our studies from Bergen, Norway.

5. Conclusions

We have shown that regular exterior paint may constitute a key contemporary source of PCBs to the urban environment, consequently with potential important implications for a variety of subjects, including

- management strategies on rehabilitation/demolition and waste disposal;
- contribution to potential contemporary source inventories;
- local marine sediment remediation strategies; and
- possible human exposure.

In order to safely identify building materials containing PCBs it should be mandatory to analyze a certain number of samples of building materials (paint, mortar, plaster and sealants) before a particular building is up for renovation or demolition. This appears especially important for buildings erected or renovated between 1950 and 1970.

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