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Active sources and dispersion mechanisms of pollutants, especially polychlorinated biphenyls (PCBs), in the urban environment

Thesis for the degree of Philosophiae Doctor

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1 Acknowledgements

Approaching the sweet relief of completing this Ph.D.-voyage I don't want to wait until the end of *Acknowledgments* to say a few words about my wife, **Hege**, and my two shining stars **Hanna Marie** and **Mia Helene**. We have been traveling on a bumpy road the last couple of years, and I have been through the two best and two worst single episodes of my life during this period. Interestingly enough, they were the same two episodes. Focusing on the Ph.D.- thesis has not been easy all the time. While at work I'm thinking of you, and when I'm at home I'm haunted by abbreviations, mostly Ph.D.s, PCBs, and NGUs. Hopefully, the clouds surrounding my occupied head will dissolve now. I love you girls, and this piece of booklet is exclusively dedicated to you.

Working under the enthusiastic guidance of **Rolf Tore Ottesen** and **Tore Volden** at the Geological Survey of Norway (NGU) is always a pleasure, and no days seem the same. The amount of projects may have been a bit much for a simple boy from Eidsvoll from time to time, but I know that I have grown with the responsibility you have given me.

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Luckily for them, my lovely **Jartun** and **Hartz** families and closest friends have had no particular idea of what I've been doing at work these four years. They all show me the importance of the life outside the office away from dusty research papers, although some of my friends have been through similar endeavors.

To wrap this up, I would like to thank my old principal at Vilberg ungdomsskole for teaching me the *Touch-technique* on an ancient typewriter. My last score was 446 strokes/89 words pr. min, which consequently would imply that this thesis theoretically (..note the alliteration..) could have been written in 6 hours and 48 minutes. It did not.

2 Preface

Several studies focusing on the environmental condition of marine sediments have been carried out in Norway. As a consequence of a general mapping performed in the Bergen harbor in 1994, an advice against the consumption of certain kinds of seafood was given. By the year 2005, 31 Norwegian fjords and harbors exhibited contaminated sediments. Something had to be done. The fact that Norwegian fjords and harbors were polluted did not correlate well with the picturesque image of Norway as a clean, undisturbed country. Norwegian county action plans were implemented and pilot projects were performed to find suitable methods for improving these environmental conditions. Several scientific institutions have joined together in interdisciplinary studies on how to manage this challenge, with the future goal of cancelling the consumption advisories.

Urban Risk is a project initiated by the Geological Survey of Norway (NGU) and the Norwegian University of Science and Technology (NTNU) to study the importance of the urban environment, the sources and possible dispersion mechanisms of pollutants as a possible main contributor to the contamination of the marine environment. The project received financial support by the County Governor of Hordaland and the Norwegian Pollution Control Authority (SFT). Other collaborators in our various Urban Risk projects have been the City of Bergen, the City of Harstad, the Governor of Svalbard, the Norwegian Public Roads Administration, Norwegian Air Research Institute (NILU), AnalyCen AS, Alcontrol AB, Tulane University in New Orleans, and the Norwegian Water Resources and Energy Directorate.

An interdisciplinary Ph.D.-thesis such as this one is not a result of one man's effort alone. I'm very grateful for being a part of this. And, as Gandalf said: *"No, the journey doesn't end here."*

"A wonderful fact to reflect upon, that every human creature is constituted to be that profound secret and mystery to every other. A solemn consideration, when I enter a great city by night, that every one of those darkly clustered houses encloses its own secret; that every room in every one of them encloses its own secret; that every beating heart in the hundreds of thousands of breasts there, is, in some of its imaginings, a secret to the heart nearest it!"

- Charles Dickens (Tale of Two Cities)

3 Abstract

The role of anthropogenic activity on the environmental condition of air, water, and soil has been a topic of research worldwide for decades. Our way of life has been the cause of a long range of local, regional, and global environmental challenges such as smog in cities, climate change, pollution of local drinking water, and the contamination of fjord and harbor sediments affecting marine organisms.

Contaminated marine sediments have been found outside areas influenced by major anthropogenic activities all over the world. These sediments receive materials in the form of suspended particles from various sources such as rivers, urban and agricultural runoff, sewage, and industrial emissions. In addition, marine sediments may act as sinks for atmospheric deposition. All the major pollutants such as heavy metals, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), dioxins, and brominated flame retardants (BFR) have been found in high concentrations in marine sediments from Norwegian fjords and harbors. In 31 locations a dietary advice against the consumption of certain seafood has been effectuated by The Norwegian Food Safety Authority and the Scientific Committee for Food Safety. Costly remediation plans, including dredging, deep-water disposal, and capping have been suggested for several areas, and pilot projects have been carried out to improve the environmental condition of the marine environment. The causes of the given contamination in each case have mostly been dedicated to industrial emissions of historical origin, which means that several remediation plans rely on the assumption that these emissions have been terminated and consequently will lead to a gradual improvement of the environmental condition when enough time passes.

Urban Risk is a project devoted to establish sustainable methods of locating and studying active sources of contamination within the urban environment. The thesis that you are holding in your hand is focusing on the urban contribution of pollutants to the marine environment by the means of stormwater runoff (Paper I) and the role of selected sources of contamination such as paint (Paper II). Main focus have been given to polychlorinated biphenyls (PCBs), which seems to be a major pollutant in several of the areas affected by contaminated marine sediments. A lot of work on PCBs has been carried out in the city of Bergen, Norway, which has been the main study area for this Ph.D.-work. However, recent developments and scientific advances brought the Urban Risk project to the Arctic, studying the role of anthropogenic activity on local PCB-contamination in the three largest settlements of Spitsbergen, Norway (Paper III).

Major achievements in this Ph.D.-study include:

- a suitable method was developed to study ongoing contamination from active sources of particle-bound pollutants in the urban environment by using stormwater sediment traps
- so far unknown sources of contamination could be revealed by using small catchments
- PCBs are transported bound to fine grained particles, and will enter the marine environment in overflow
- the main source of PCBs in the studied area is paint
- anthropogenic activity combined with decaying PCB-containing common applications (paint, small capacitors, concrete) have to a large degree contaminated soils and sediments in the Russian coal-mining settlements of Spitsbergen in the Arctic.

4 Abbreviations

AMAP	Arctic Monitoring and Assessment Programme
B(a)P	Benzo(a)pyrene, a PAH component
BFR	Brominated flame retardants
BMP	Best management practices
CV	Coefficient of variance
DDT	Dichlorodiphenyltrichloroethane
dw	Dry weight
GC-ECD	Gas chromatography with electron capture detector
GFAAS	Graphite furnace atomic absorption spectrometry
HBCD	Hexabromocyclododecane (a BFR substance)
HCB	Hexachlorobenzene
ICP-AES	Inductively coupled plasma atomic emission spectrometry
ICP-MS	Inductively coupled plasma mass spectrometry
IUPAC	International Union of Pure and Applied Chemistry
mg/kg	Milligrams per kilogram (e.g. for a pollutant in a sediment sample)
NGU	Geological Survey of Norway
NILU	Norwegian Air Research Institute
NS	Norwegian standard
NTNU	Norwegian University of Science and Technology
ppb	Parts per billion (same as µg/kg or ng/g)
ppm	Parts per million (same as mg/kg or µg/g)
PAH_{16}	Polycyclic aromatic hydrocarbons (16 US-EPA components)
PCB_7	Polychlorinated biphenyls, represented by the seven IUPAC-congeners no. 28, 52, 101, 118, 138, 153, and 180.
POPs	Persistent organic pollutants
SD	Standard deviation
SFT	Statens forurensningstilsyn (Norwegian Pollution Control Authority)
TOC	Total organic carbon
US-EPA	United States Environmental Protection Agency

5 Background

5.1 Contaminated sediments

There is currently an environmental focus on polluted sediments in harbors and fjords in Norway. Marine sediments and organisms from 32 harbors and fjords (updated 2005) in Norway contain concentration levels of pollutants such as PCBs, PAHs and heavy metals that by far exceed the guideline levels of the Norwegian Pollution Control Authority (SFT) and the Norwegian Food Safety Authority (Mattilsynet). Table 1 shows the classification of marine sediments based on the content of selected pollutants. In each of the areas (table 2, figure 1), a dietary advice against the consumption of certain seafood exists, and there may also be local restrictions on the trade of fish and shellfish (Økland, 2005). Contaminated sediments are, however, not only confined to the marine environment. Studies have also shown elevated concentrations of PCBs and BFRs in sediments and biota of Lake Mjøsa, Norway (Fjeld et al., 2004; Mariussen et al., 2008).

Table 1. Environmental classification of Norwegian marine sediments based on the content of selected pollutants, adapted from Molvær et al., (1997).

		Environmental classification				
		I	II	III	IV	V
Parameters	Units	Insignificantly polluted	Moderately polluted	Markedly polluted	Strongly polluted	Very strongly polluted
Arsenic (As)	(mg/kg)	<20	20 - 80	80 - 400	400 - 1000	>1000
Cadmium (Cd)	(mg/kg)	<0,25	0,25 - 1	1 - 5	5 - 10	>10
Chromium (Cr)	(mg/kg)	<70	70 - 300	300 - 1500	1500 - 5000	>5000
Copper (Cu)	(mg/kg)	<35	35 - 150	150 - 700	700 - 1500	>1500
Mercury (Hg)	(mg/kg)	<0,15	0,15 - 0,6	0,6 - 3	3 - 5	>5
Nickel (Ni)	(mg/kg)	<30	30 - 130	130 - 600	600 - 1500	>1500
Lead (Pb)	(mg/kg)	<30	30 - 120	120 - 600	600 - 1500	>1500
Zink (Zn)	(mg/kg)	<150	150 - 700	700 - 3000	3000 - 10000	>10000
PCB_7	(µg/kg)	<5	5 - 25	25 - 100	100 - 300	>300
PAH_{16}	(µg/kg)*	<300	300 - 2000	2000 - 6000	6000 - 20000	>20000
B(a)P	(µg/kg)	<10	10-50	50-200	200-500	>500
TBT	(µg/kg)	<1	1 - 5	5 - 20	20 - 100	>100
	*naphtalene not included					

The specific sources of sediment contamination in each case may be difficult to assess. It is almost impossible to quantify old, terminated industrial emissions. For the city of Bergen alone, over 300 companies surrounding the harbor basin have been listed as possible polluters, but only a handful of these can document their current emissions to

the harbor according to their appointed discharge permits (Kryvi et al., 2005). The industrial emissions of PCBs have assumingly ceased after the national ban on new use implemented in 1980 (Norwegian Legislation, 2004). However, several products containing PCBs are still unaccounted for, and the contribution from such sources, actively distributing PCBs to the urban environment, is the main target of the Urban Risk project.

Table 2. Norwegian fjords and harbors with contaminated sediments and dietary advice against consumption of certain seafood. The three settlements on Svalbard (Paper III) not included in the contaminated sediment study by Økland (2005) are also indicated on the map.

ID	Fjord/harbor	Contaminants	ID	Fjord/harbor	Contaminants
1	Honningsvåg	PAH	19	Karmsund	PAH, PCB
2	Hammerfest	PAH	20	Stavanger	PAH, PCB
3	Tromsø	PAH	21	Sandnes	PAH
4	Harstad	PAH, heavy metals	22	Flekkefjord	PCB
5	Ramsund	PCB	23	Fedafjord	PAH
6	Narvik	PAH, PCB	24	Farsund	PAH, PCB
7	Brønnøysund	PAH	25	Kristiansand	PCB, dioxins
8	Sandnessjøen	PAH	26	Arendal	PCB
9	Vefsnfjorden	PAH	27	Tvedestrand	PCB
10	Ranfjorden	PAH	28	Kragerø	PAH, dioxins
11	Trondheim	PAH, PCB	29	Grenlandsfjordene	Dioxins
12	Hommelvik	PAH	30	Sandefjord	PCB
13	Sunndalsfjorden	PAH	31	Tønsberg	PCB
14	Årdal	PAH	32	Oslofjorden	PCB
15	Ålesund	BFR (HBCD)	*33*	*Longyearbyen*	-
16	Sørfjorden	Heavy metals, PCB	*34*	*Barentsburg*	*PCB,HCB,DDT (Evenset et al., 2006)*
17	Bergen	PCB	*35*	*Pyramiden*	*PCB,DDT (Evenset et al., 2006)*
18	Sauda	PAH			

Actions to improve the environmental condition of marine sediments will be proposed in each of the locations comprised by consumption advisories promoted by Report no.12 to the Storting (The Norwegian Parliament) (2001-2002) – *Protecting the riches of the seas* (Ministry of Environment, 2002). A detailed action plan initiated by a joint association of scientific environmental institutions is expressed for Bergen harbor specifically (Kryvi et al., 2005). Considerations regarding the causes of contamination, risk assessments, and the effects of remediation are taken into account before deciding

on the final method to improve the conditions. Several alternatives for remediation of contaminated sediments exist, with the main approaches being

1. dredging for a subsequent disposal of the materials onshore or in a deep-water environment,
2. capping with clean materials, and
3. natural attenuation with regular monitoring

A successful design of suitable remediation plans involves hydraulic, chemical, and geotechnical approaches in order to decrease the possibility of pollutant dispersion (Mohan et al., 2000; Fredette and French, 2004; Schaanning et al., 2006; Förstner and Apitz, 2007; Eek et al., 2008). The choice, and effect, of any action initiative is dependent on the relative importance of terminated, old sources and active sources. Results from pilot remediation projects in Norway have indicated a general improvement of contamination level after dredging, but in some cases a top-layer of the bottom sediment was still highly contaminated (Voie et al., 2002). There may be several reasons for this observation such as the inevitable whirl up of fine particles during dredging resulting in highly contaminated suspended particles (Mackie et al., 2007). Voie et al. (2002) also experienced that the dissolved concentrations of PCBs in the water column remained unchanged after dredging and the PCBs were still available to sediment dwelling species accumulating the pollutants directly from the water.

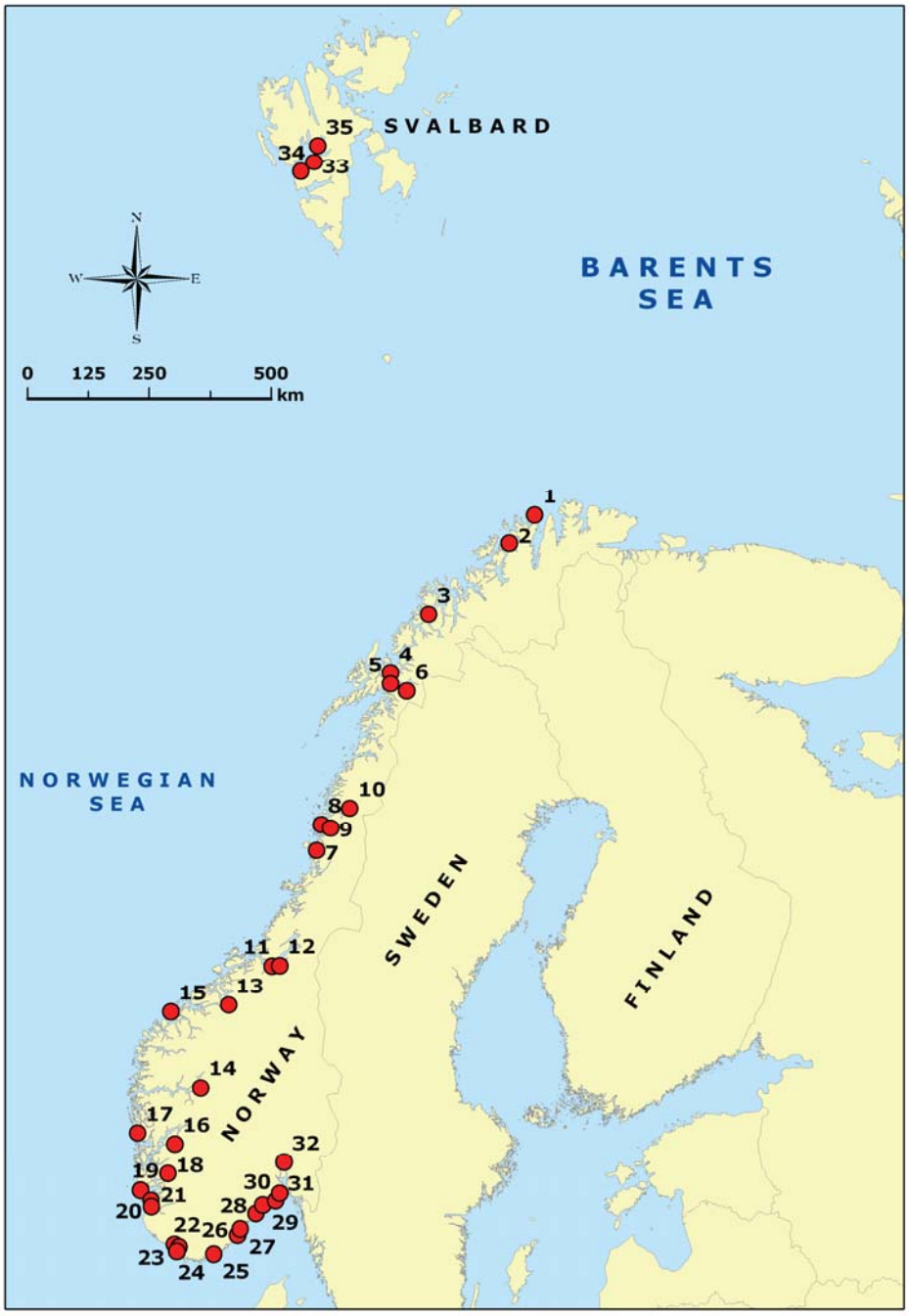

Figure 1. Map indicating Norwegian sites with contaminated sediments and dietary advice against the consumption of certain seafood (adapted in ArcMap from Økland, 2005). The three settlements on Svalbard studied in Paper III are added to the map. See table 2 for details.

Still ongoing discharges from unknown sources have been suggested to account for some of the observations of still contaminated sediments after remediation of Norwegian harbors. This was part of the basis for the Urban Risk initiative. The questions to be answered were how important the urban environment is to the dispersion of PCBs to the marine sediments, what are the driving mechanisms for the eventual dispersion, and how are we to locate the contributing specific sources with suitable methods? Figures 2 and 3 are maps showing the condition of marine sediments in two areas of Bergen harbor based on the concentration of PCBs (adapted from Kryvi et al., 2005). The colors represent the different classes of contamination presented in table 1.

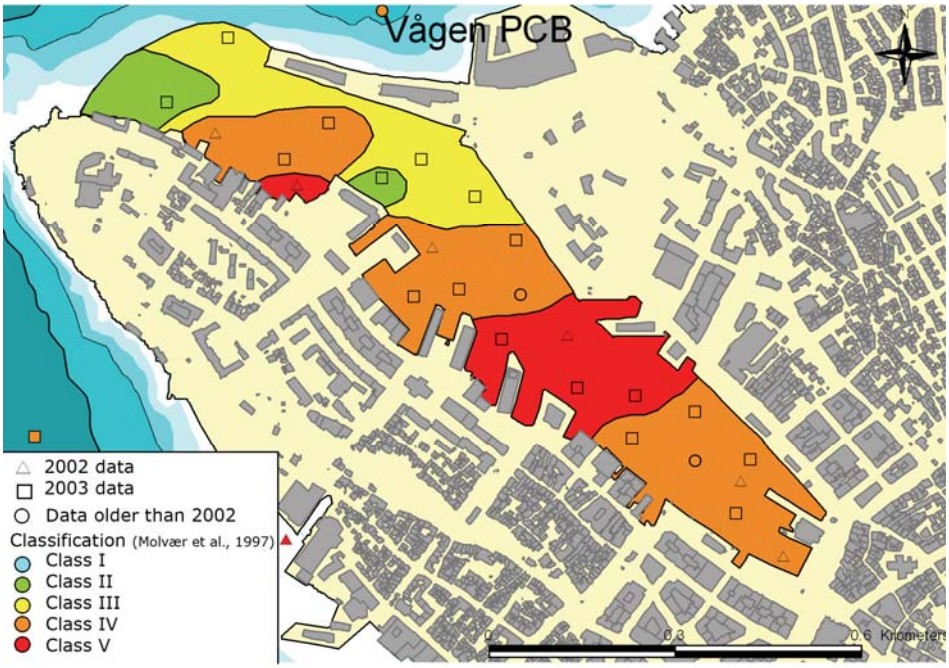

Figure 2. Environmental condition of marine sediments in Vågen, Bergen harbor, based on the concentration of PCBs (adapted from Kryvi et al., 2005).

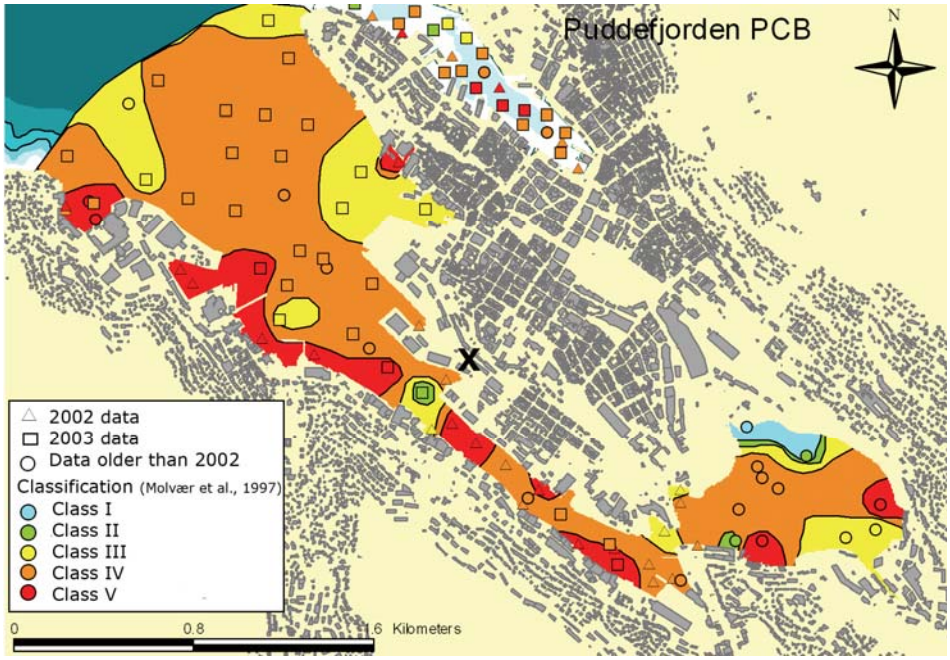

Figure 3. Environmental condition of marine sediments in Puddefjorden, Bergen harbor, based on the concentration of PCBs (adapted from Kryvi et al., 2005). The X marks the location of one particular sampling site discussed in chapter 6.

5.2 The Urban Environment

By the end of 2008 more than half of the World's population will be living in urban areas; towns and cities (UNFPA, 2007). Urbanization has been stabilized in America and Europe, with 3 out of 4 people living in urban areas. In Africa and Asia the number of urban dwellers is rapidly increasing, and will exceed 50 % within few years. Although most cities constitute the foundation of innovation and wealth, local urban governments are faced with complicated challenges such as slum development, unemployment, and the risk of terror. Sustainable advances in water and food supply, waste treatment, and sewer systems are needed in every expanding city. In addition, there are important challenges regarding air, water, and soil pollution, and the associated health implications (Genske, 2003; Starke, 2007).

5.2.1 Urban soil pollution in Norway

Geological Survey of Norway (NGU) has carried out studies of urban soil pollution in several Norwegian cities focusing on both inorganic and organic pollutants (Ottesen and Volden, 1999; Ottesen and Langedal, 2001; Jartun et al., 2002; Jartun et al., 2003; Jartun and Volden, 2006; Jartun et al., 2006). Geochemical maps describing the geographical distribution of pollutants within the city limits indicate that the older, central parts of these urban areas are generally polluted with various heavy metals such as Pb, Cd, Zn, and Hg in addition to PAHs. Studies have also revealed that the surface soils within children's playgrounds are often polluted with As originating from the extensive use of CCA (copper, chromium, arsenic) impregnated wood used in playground equipment (e.g. Jartun et al., 2003). PCBs have been found in high concentrations in soils of specific areas, which in most cases reflect one or several local sources such as masonry coatings (Andersson et al., 2004).

5.2.2 Tracing active sources of contamination – introducing the stormwater sediment method

Even though urban soil has proven to be a suitable sample medium to establish the environmental impact of an urban area, the specific sources of contamination may remain unidentified for a given location. First of all, urban soil is a dynamic sample medium, meaning that it has been turned and used over and over again following the beat of the city since people first set their foot in that particular area. Excavation, relocation, and burial through several decades have generally made urban soils into inhomogeneous signals of anthropogenic activities (e.g. Ottesen and Langedal, 2001). For this reason, urban soil may be unsuitable for the concept of detecting ongoing contamination to e.g. the marine environment. Furthermore, the current model for calculation of urban runoff from impervious surfaces is based on dissolved pollutants and do not focus on the particle-bound dispersion (Lindholm, 2004).

Based on the knowledge from several studies of urban soil pollution, the need for a suitable and practical method appropriate for studying the role of active sources of

contamination to the marine environment became evident. Urban runoff sediments entrapped in small stormwater traps suited most of the intentions for the Urban Risk project presented in chapter 2. The method is described in detail in Paper I (Jartun et al., 2008). The main motive behind choosing stormwater sediments as a sample medium for ongoing contamination in an urban environment was that these materials are "young", given that they are removed more or less regularly each year by the proper authorities dependent on where the traps are located. In most cases the removal responsibilities apply to the Norwegian Public Roads Administration or the Municipality. Furthermore, each stormwater trap has a limited catchment area from which it receives materials. If a high concentration of any given pollutant is found in the runoff sediments, the source of contamination is confined to a highly specific area of no more than 5-600 m^2.

5.2.3 The impervious surfaces of the urban environment

One of the most apparent impacts of urbanization is the complete loss or burial of surface soil as a result of anthropogenic activities such as building and road construction. Any urban area may consist of between 50-100 % of impervious surfaces represented by roofs, asphalt roads, and concrete (Lindholm, 2004 ; Lu and Weng, 2006), which represents a considerable volume of buried or displaced soil in addition to an important change in the hydrological distribution of urban surface water. Figure 4 illustrates the impact of urbanization, allowing only a minimum amount of stormwater to infiltrate the ground, evaporate, or be retained by vegetation (Mays, 2001). The water balance will change, resulting in increased runoff rate, lowering of the groundwater table, and deterioration of herbal growth. Development of impervious surfaces may consequently cause several episodes of high-volume runoff during a normal precipitation year, especially in a coastal environment such as Bergen with an annual amount of rainfall averaging 2250 mm/yr. Eventually, large storm episodes may be followed by challenges regarding the hydraulic efficiency of old sewer systems and consequently a direct discharge to a downstream recipient in overflow (Thorolfsson, 1999). Figure 5 presents some possible solution of Best Managing Practices (BMP) to manage the expected increase in rainfall and runoff occurrences (Lindholm et al., 2005; IPCC, 2007).

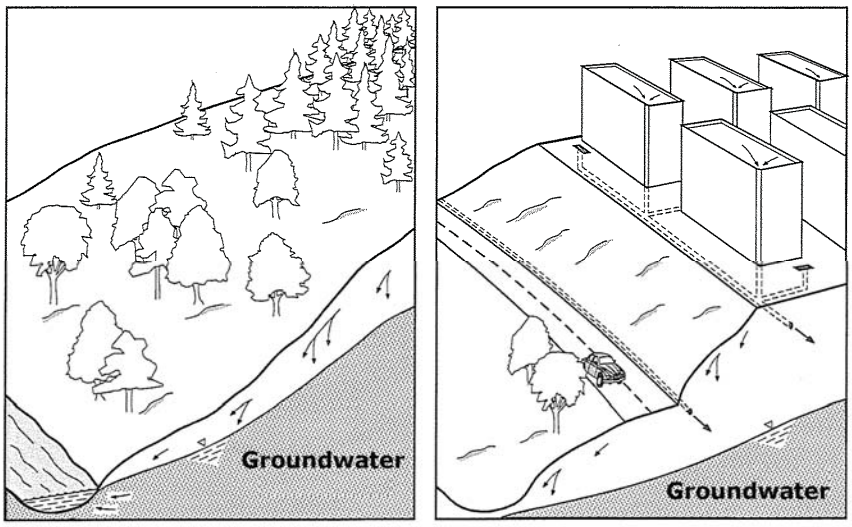

Figure 4. The effect of urbanization upon runoff.

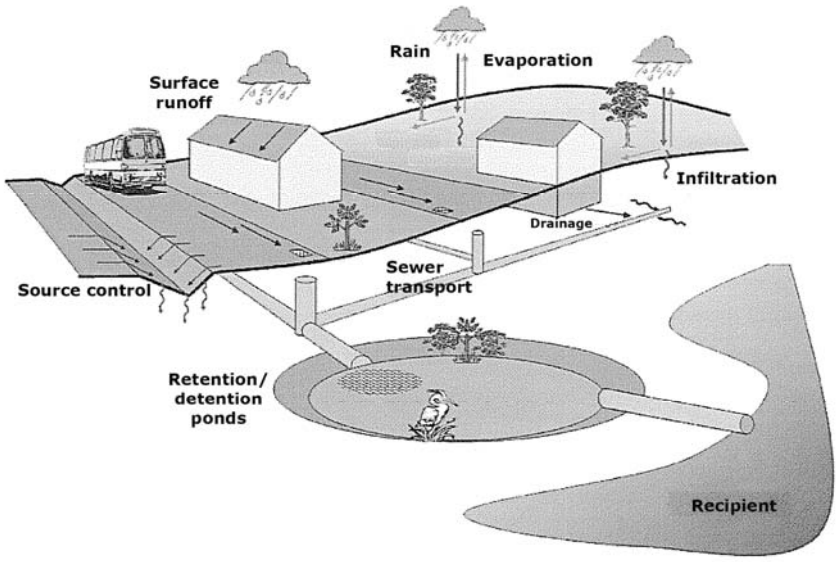

Figure 5. Composite stormwater system; examples of solutions to reduce a high-volume runoff peak facilitated by impervious surfaces.

5.2.4 Main study area – around Bergen city harbor

As a main principle, set forth by the city of Bergen, it is not desirable to direct stormwater into the sewage system unless the stormwater is highly contaminated. This is a somewhat vague principle, because there are several considerations to take into account such as the age and capacity of the pipeline network, the cost of treatment, and the type and quantity of a potential contamination. In addition, how do you know if the stormwater is contaminated at a certain point? The main development of the water-and-sewage network in Bergen began around 1854 when the first municipal waterworks were established from the main freshwater source Svartediket to the city centre. At the same time the first sewage pipelines were constructed, and the oldest pipeline still in service in Bergen today dates back to 1880. In 2003 a total length of 1150 km public sewage pipelines existed underneath the feet of the around 247 000 inhabitants, including 400 km of combined system, 400 km of separate waste water, and 350 km of stormwater pipelines. Generally, there is a combined system in the old and central part of the city, where untreated household sewage has been discharged directly into the harbor for many years. However, during recent years the wastewater and stormwater are split into separate systems in several parts of the city leading the former to treatment facilities and the latter still often directly to the harbor to avoid a constraint of the network capacity. In areas still having a combined system there are pumping stations installed to lead the water to a treatment facility. Unfortunately, pipelines with low capacity combined with large amounts of rain (2250 mm on average annually for Bergen city centre) cause the system to go into overflow approximately 30 % of the time, consequently directing (contaminated) stormwater directly to the recipient.

5.3 Human and environmental health implications of PCBs

The Stockholm Convention on Persistent Organic Pollutants (POPs) call on the acknowledging parties to recognize the toxic and persistent properties of compounds such as PCBs, DDT, HCB, furans, and dioxins with the objective of protecting human and environmental health (UNEP, 2001). Obligations to reduce or eliminate emissions

of POPs include restriction or prohibition of intentional production and use in addition to regulations on import/export. The main exposure of PCBs to humans is by ingesting contaminated fish or other seafood (e.g. Sandau et al., 2000). In addition, occupational exposure may be significant e.g. during management of old electrical transformers or capacitors, and renovation of contaminated buildings. Children may be exposed to PCBs prenatally and from breast milk since the PCBs are stored in fat tissue in the mother's body and subsequently released during pregnancy, cross the placenta, and finally enter fetal tissues. Human health effects associated with PCB exposure may include liver, thyroid, dermal and ocular alterations, immunological alterations, neurodevelopmental changes, reduced birth weight, reproductive toxicity, and cancer (ATSDR, 2000).

Dispersion and mobilization of pollutants within environmental compartments may give rise to both human health problems and environmental challenges. The Urban Risk project is dedicated to study specific contaminants such as the PCBs, PAHs, and heavy metals in the urban environment and their significance to a downstream recipient, consequently the marine sediments. As mentioned in chapter 5.1, specific advice against the consumption of seafood exists in more than 30 fjords and harbors in Norway alone (Økland, 2005). High concentrations of pollutants in sediments, both as bottom sediments and suspended, will subsequently lead to a biomagnification through food chains depending on specific properties of a given pollutant such as lipophilicity and resistance to degradation (Ruus et al., 2005). The presence of pollutants such as PCBs in air, water, soil, or sediment becomes especially important to address in Arctic areas, where the accumulation and biomagnification is enhanced by a number of factors such as several trophic levels and a general high fat content in animals. Consequently, the levels of pollutants in arctic areas are causing high concentrations in top predators such as the polar bear (Skaare et al., 2000; Haave et al., 2003).

6 Urban Risk – discussion of main results

The major findings in the Urban Risk project are presented and discussed in detail throughout Papers I-III. There are however a few approaches that deserve mentioning here in a main chapter discussing the achievements from the project. And, as mentioned in chapter 2, the journey is not over with the completion of this Ph.D.-thesis. There are several paths to follow from this point forward as mentioned in chapter 7.

6.1 Small catchment stormwater traps

Paper I provides concentrations of several pollutants in samples of stormwater sediments originating from small catchments within the impervious urban environment. Figure 6 outlines the main idea behind choosing these technical installations as a tool to reveal active sources of pollutants that may in the long run contaminate the recipient. A GIS-application of the sewage system in Bergen, provided by the Agency for Water and Sewerage Works at Bergen Municipality, was used as a basis for understanding how the urban dispersion of pollutant via the stormwater system may function. This is a complicated, but suitable tool also for predicting marine areas potentially susceptible for contamination by the active sources. In this particular case, concentrations of PCB_7 up to 650 µg/kg were found in duplicate samples of stormwater sediment (Paper I). Additional studies presented in Paper II indicated that the source of PCB contamination arose from flaking paint from the building marked in red color in figure 6, draining to this specific stormwater trap. The concentration in paint was about three orders of magnitude higher than what was found in the stormwater trap, indicating a dilution from other sediment sources such as asphalt wear and tear, and from the sanding of icy roads in winter. Neighboring sediment traps not receiving materials from this specific building, but still from the same possible additional sources, had low concentrations of PCB.

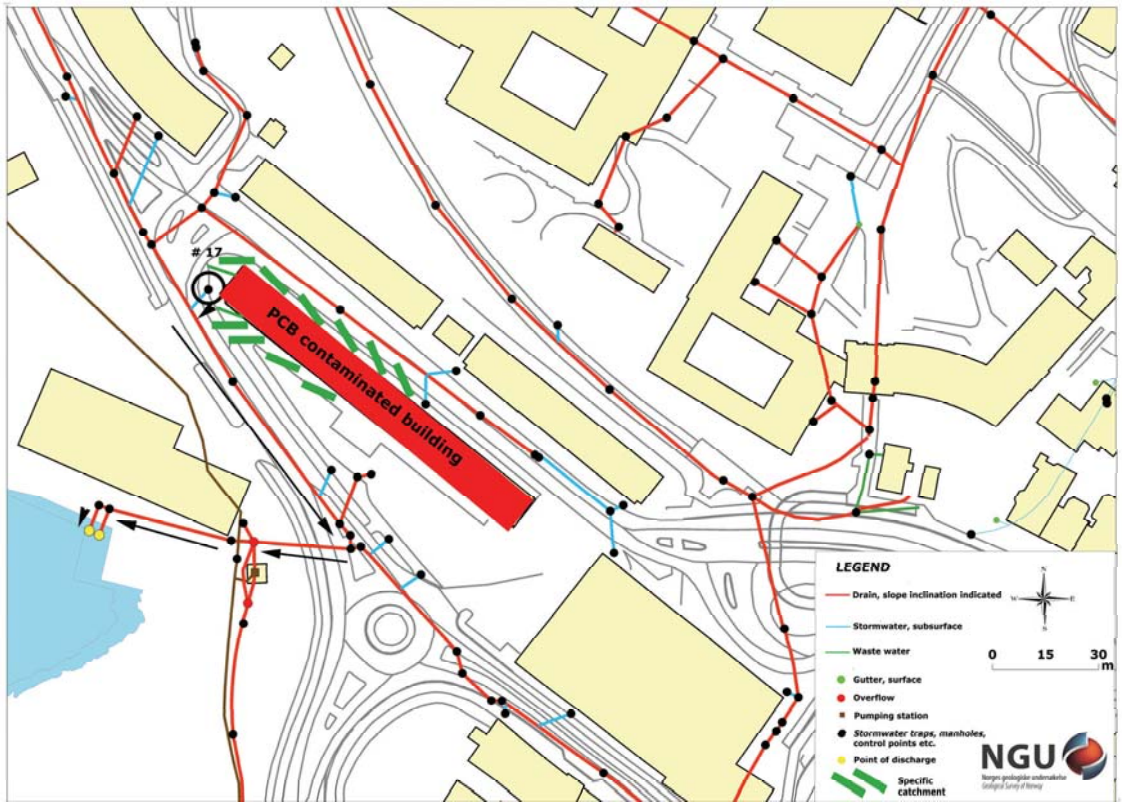

Figure 6. Principles for the small catchment idea. High concentrations of PCBs found in the stormwater trap (sample # 17, encircled) must derive from the marked green-striped area. In this specific case, the source of PCB contamination is the surface paint layer of the red shaded building. Arrows indicate the pipe inclination towards a point of overflow and discharge to the harbor.

Natural weathering and erosion of building surfaces is believed to cause the mobilization of contaminants to the immediate environment in particle bound form. Flaking paint may be a key contemporary source of PCBs (Paper II), see figure 7. Renovation of contaminated building surfaces may further enhance the dispersion, as is outlined in figure 8.

Figure 7. Flaking, PCB-containing paint from the surface of the indicated building in figure 6 (Photo: R.T.Ottesen).

Figure 8. Renovation of contaminated building surfaces will enhance the mobilization of e.g. PCBs to the stormwater system (Photo: M.Jartun).

Studying the PCB_7 profile represented by the relative contribution from the individual congeners, conformity between the two sample media (stormwater sediment and paint from building surface) is indicated (figure 9). Further indication of an active contribution of PCBs downstream to the marine environment can be observed by studying the PCB_7-profiles from the stormwater trap (sample ID # 17) and three marine sediment locations in Puddefjorden close to the discharge point downstream, adapted from Kryvi et al., 2005 (figure 10). Caution should be made when comparing the concentrations and congener profiles in the samples of stormwater sediment and marine sediment in this case because these samples were not from the same project, not sampled by the same team, not analyzed by the same laboratory, and not subject to the same analytical procedure. In addition, considerations on the extent of chemical, physical or biological degradation are not taken into account. Furthermore, sediments outside the discharge point indicated in figure 6 may consist of stormwater sediments deriving from other small catchments in addition to the one draining directly to location # 17 on the map. If we nevertheless humbly allow ourselves to compare the profiles under these premises, it is indicated that PCB-sources found onshore, in this case a

contaminated building surface, will have an effect on the environmental condition of the downstream recipient.

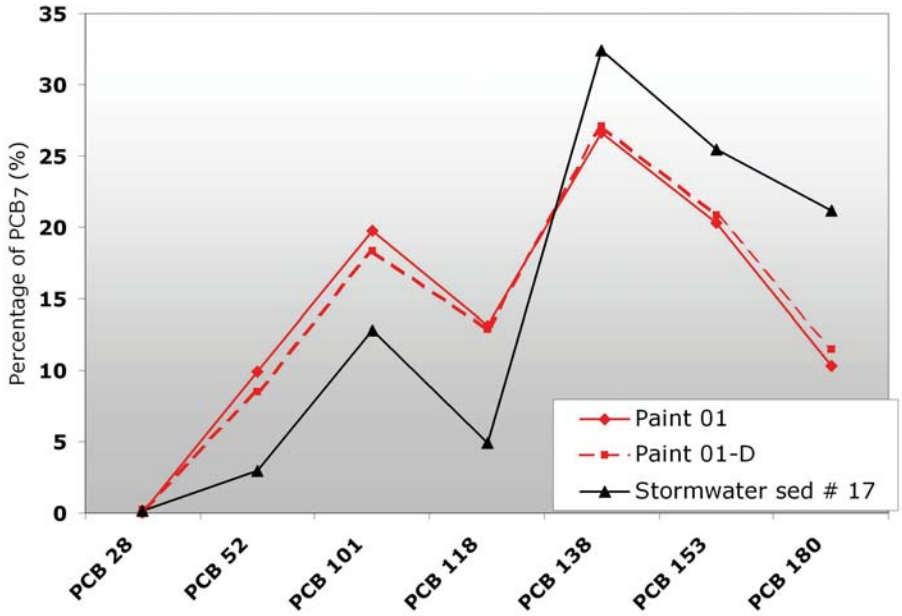

Figure 9. Comparison between PCB_7 profiles in stormwater sediment sample ID # 17 (Paper I), and duplicate paint samples # 01 and # 01-D (Paper II).

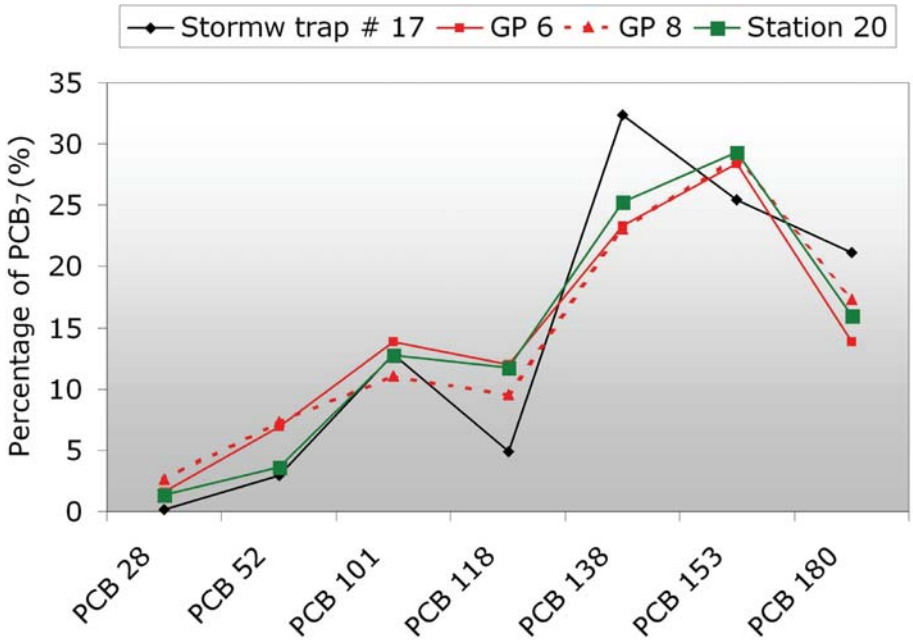

Figure 10. Comparison between PCB_7 profiles in stormwater sediment sample ID # 17 (Paper I), and samples of marine sediment in Puddefjorden close to this location (Kryvi et al., 2005).

6.2 The characterization of stormwater sediments – additional insight

The grain size distribution of the stormwater sediments in 21 selected samples is discussed in Paper I, focusing on the possibility for dispersion in suspended form. Generally, the sediments were fine grained, in some samples up to 90 % of the sample volume consisted of particles with a diameter less than 250 μm. The finest and the coarsest sample observed in addition to the average is indicated in the distribution figure in Paper I. The results further indicate a relationship between the topography of Bergen city and the grain size distribution found in stormwater sediments, possibly reflecting a more effective flush out of fine grained sediment in the sewage system of steeper areas.

In some of these samples enough material was available to carry out PCB-determination within different size fractions (not discussed in Paper I). In eight selected samples the dry, original sample material (> 2 mm) were sieved into five new fractions (i-v);

i. < 60 µm
ii. 60 – 150 µm
iii. 150 – 250 µm
iv. 250 - 500 µm
v. 500 - 2000 µm

The PCB_7-concentration was determined following the analytical procedure presented in Paper I. The analyses were performed on stored, dry material, consequently the possibility of alteration to the material in the process from wet, raw sample to a completely dry material should be considered. Illustrations describing the concentrations of PCB_7 in the five specific size fractions are shown for two individual samples in the stormwater sediment study (Paper I), including sample # 17 discussed previously (figure 11). Sample ID 51 was collected close to the main square, Torgallmenningen, in the city center of Bergen. The black curve indicate the percentage of each size fraction compared to the total sample volume, i.e. for sample ID 17, the i-fraction constitutes 57 % of the total sample. The bars indicate the concentration of PCB_7 in each size fraction, i.e. the measured concentration of PCB_7 in the i-fraction of sample ID 17 is 0.39 mg/kg.

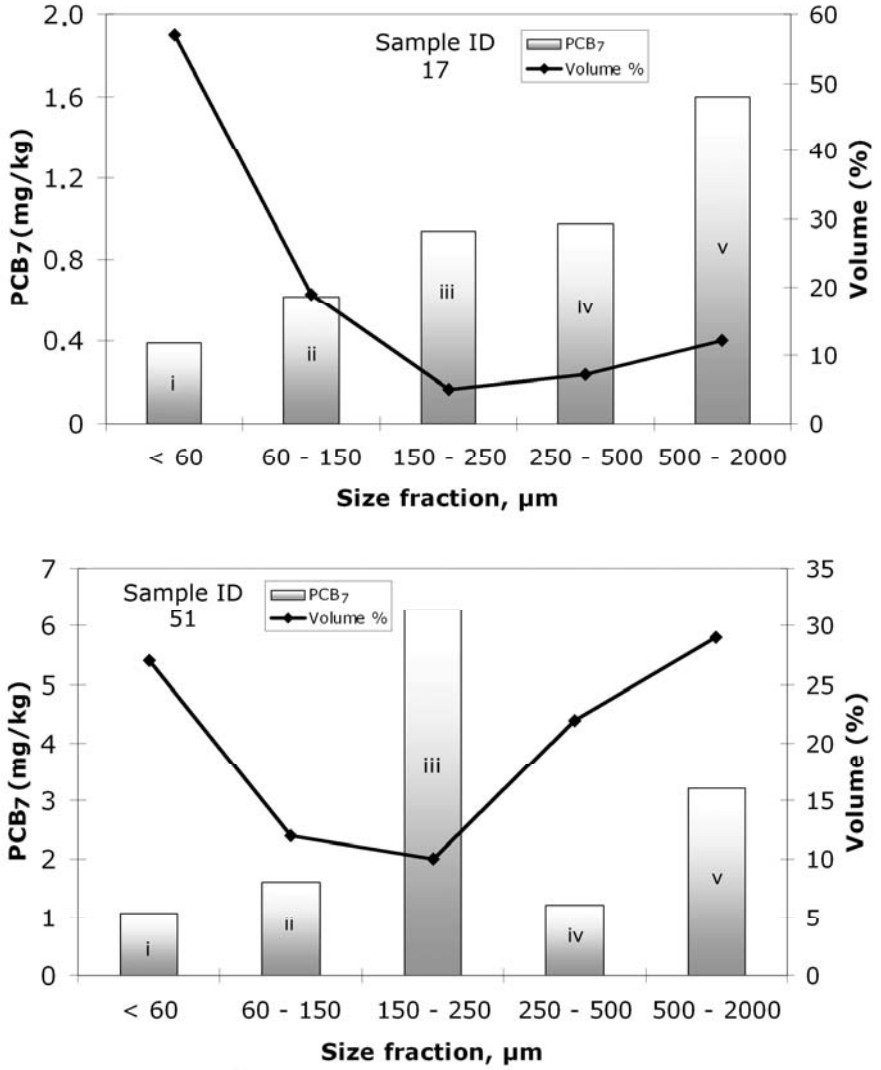

Figure 11. Concentration of PCB_7 (mg/kg) in five size fractions from sample ID 17 (top) and 51 (bottom) in the stormwater sediment study (Paper I). The black curve indicates the volume of each fraction compared to the total sample.

The two examples shown in figure 11 indicate no direct relation between particle size and concentration of PCB_7 in the studied size fractions. The same is observed in the other six samples not shown here. However, when studying the material more closely there seems to be a relation between the PCB_7 distribution for the five different fractions and the main type of PCB-source for each sample. As an example, the highest

concentration of PCB_7 in sample # 17 is found in the coarser v-fraction. The main source at this location is the surface of a contaminated building as shown in figure 6. Considering the sampling locations and potential sources within each given catchment, it appears that stormwater traps receiving materials from a building surface (e.g. sample ID 17), the highest PCB_7 concentrations are found in the coarser percentage of the material. A suggested explanation for this is the weathering and mobilization of relatively large pieces of paint and plaster/concrete from the surface of buildings. Some of the samples have the highest concentration of PCB_7 in one or more of the smaller size fractions, including one location close to a local recycling station (see Paper I). The abundance of potential sources within this particular catchment is probably reflected in the size fractions, exhibiting a more or less similar distribution in all five fractions. This is also seen for some of the other locations where the concentrations of PCB in the stormwater sediments derive from several potential sources.

6.3 Estimations of "background" levels of PCBs – a dead-end-story?

When studying the stormwater sediments from small catchments, such as the one shown in figure 6, there is always a question of the "background" or "divine" contribution, i.e. PCBs falling from the sky in the form of particles washed out of the urban atmosphere by precipitation, or PCBs dissolved in the water phase. To estimate this contribution a series of bulk precipitation samplers adapted from Reimann et al. (1997) and Chekushin et al. (1998) were placed within the central city of Bergen and in more rural parts of the surrounding area. The main objective of this part of the Urban Risk project was to provide an estimate of the wet deposition of polychlorinated biphenyls (PCBs) within an urban region affected by elevated precipitation. The designated locations were selected to cover both remote areas and regions possibly influenced by local sources of PCBs, i.e. central, urban Bergen and the area close to a local municipal solid waste incinerator (MSWI). In addition to precipitation samples, six samples of cleansed stack gas from the MSWI were analyzed. The concentrations of PCBs found in our precipitation samples were remarkably low compared both to Norwegian background studies (Aas et al. 2006) and studies carried out elsewhere, e.g. in Eastern

Mediterranean (Mandalakis and Stephanou, 2004), in New Jersey (Van Ry et al., 2002), in Chesapeake Bay (Dickhut and Gustafson, 1995), in Southern Sweden (Backe et al., 2002), and in the Baltic Sea region (Agrell et al., 2002). The median value of PCB_7 found in the precipitation samples was 47.8 pg/L. All samples but one (PCB_7 = 1080 pg/L) exhibited concentrations close to the median value. The suggested source of the elevated concentration of PCB_7 found in the one single sample was emissions from the MSWI, located only 2-3 kilometers away. Results from PCB-measurements of the stack gas confirmed that PCBs enter the atmosphere from this waste treatment plant in concentrations up to 275 times the average air concentration in Norway (Aas et al., 2006). The PCB congener profiles in the stack gas complied with the profiles in our precipitation sample close by.

Based on the results of PCBs in precipitation, calculations of potential annual wet deposition for the sampling areas were made. However, in addition to real-world precipitation samples we included six samples of doubly distilled water and pieces of the sampling devices embedded in distilled water as “blanks” to the laboratory. Subsequently, it became evident that the concentrations of PCBs in these “blank” samples were as high (or low) as those of the real precipitation samples, consequently leading to a temporarily rejection of the results. This time-consuming part of Urban Risk may not have led us closer to a well-defined estimate of the wet deposition contribution to the environmental condition of e.g. stormwater sediments or subsequently marine sediments, but it was a lesson well learned. We cannot rule out the possibility that the plastic materials constituting the precipitation sampler may contain PCBs in small amounts, as indicated in table 3. Similar challenges were not observed with the stormwater sediment samples.

Table 3. PCB_7-concentrations in "blank" samples of doubly distilled water (DDW) alone (A+B) and including pieces of the sampling equipment (C+D) in addition to the average concentration found in 14 samples of precipitation from Bergen. Analyses performed at NILU.

PCB congener	A DDW* water A pg/L	B DDW water B pg/L	C DDW + plastic can and screw cap pg/L	D DDW + pieces of plastic bag pg/L	E Median, precipitation (N=14) pg/L
28	4.27	7.21	70.8	19.5	4.30
52	5.16	12.5	75.1	18.6	6.12
101	13.6	19.7	109	42.2	7.11
118	5.27	12.0	86.3	33.8	8.03
138	5.91	7.45	78.9	43.4	5.98
153	11.7	16.0	143	66.9	9.59
180	3.85	3.73	19.9	18.7	2.21
PCB_7	**49.7**	**78.6**	**583**	**243**	**47.8**

Analytical challenges when approaching concentrations down to pico- and femtograms per liter are also possible. Meteorological data showed a continuous period of rain during the sampling period, which means that the potential urban signal of PCBs in the atmosphere caused by e.g. whirled-up dust originating from sources within the urban area, was probably non-existent. The contribution of PCBs from wet deposition represented by precipitation is probably small compared to local sources providing the stormwater system with particle-bound PCBs (Paper I), but due to the analytical and technical challenges, this was impossible to quantify in this phase of the Urban Risk project.

7 Outlook - Relevance and further directions

Even though extensive research on pollutants such as PCBs, PAHs, and heavy metals have been conducted all over the world for decades, further understanding of their environmental influence is required on both the local and global scale. The request from the international environmental community for an increased effort from the "authorities" to continue environmental research and development is expressed in the Stockholm Convention on POPs (UNEP, 2001):

"The Parties shall, within their capabilities, at the national and international levels, encourage and/or undertake appropriate research, development, monitoring and cooperation pertaining to persistent organic pollutants and, where relevant, to their alternatives and to candidate persistent organic pollutants, including on their:

a) Sources and releases into the environment;
b) Presence, levels and trends in humans and the environment;
c) Environmental transport, fate and transformation;
d) Effects on human health and the environment;
e) Socio-economic and cultural impacts;
f) Release reduction and/or elimination; and
g) Harmonized methodologies for making inventories of generating sources and analytical techniques for the measurement of releases."

Paper I accompanying this thesis includes discussions involving items a, b, c, and g from the list above. Paper II follows up on one specific source of PCBs, involving items a, c, f, and g. Finally, paper III from the arctic environment on Spitsbergen discusses the sources, dispersion mechanisms, and possible effects of PCBs involving items a, b, c, d, e, f, and g in the proposed list from UNEP (2001) cited above. The results and discussions obtained by the work on these papers and the current thesis are but modest contributions to the ongoing environmental debate that prevails in Norway today. Nevertheless, some of the approaches, such as;

1) the method of using small urban stormwater catchments to reveal real-time contamination from active sources presented in Paper I,
2) the disclosure of exterior paint as a major source of PCBs in Norway presented in Paper II, and

3) the discovery of heavily polluted surface soil caused by large amounts of discarded waste and consequently the active erosion by local rivers in the Arctic illustrated in Paper III,

represent potentials to be developed into new scientific projects.

7.1 Paper I

Currently, the Bergen Harbor project, working on suitable solutions for the remediation of the marine sediments outside the urban, impervious Bergen area, is postponed partly because of the indications of active dispersion of pollutants presented in the Urban Risk project (Paper I). As a consequence, the Urban Risk project has, in collaboration with the Norwegian Water Resources and Energy Directorate (NVE) and the City of Bergen (Agency for Water & Sewerage Works), established a monitoring station measuring water and suspended sediment transport from a well-defined urban catchment and the subsequent discharge to the main recipient, the marine sediments of Bergen harbor. The station was activated in April/May 2008 and will be in operation at least throughout 2008, unfortunately a little too late for presentation in the present thesis. Furthermore, the stormwater sediment method presented in Paper I has later on been tested in two other Norwegian cities affected by contaminated sediments. In Harstad, sediments from 34 stormwater traps with limited catchment areas were collected and the concentrations of PCB_7, PAH_{16}, TOC, and 33 chemical elements were determined. No significant dispersion of pollutants via the stormwater system was discovered. However, the results of 186 samples of surface soil in the same project indicated high concentrations of Cd, Cu, Pb, Sn, Zn, and PAH_{16} close to the harbor. In this city, no stormwater traps were found in the immediate vicinity to the harbor (Jartun and Volden, 2006). In Trondheim, Bjervamoen et al. (2006) collected 71 samples of stormwater sediments from 53 locations. Major findings in this study included the discovery of several unknown active contamination sources for PCBs, PAHs, and specific elements. Urban runoff sediments have also been studied by others in Norway, however with a different approach in methodology (Cornelissen et al., 2008).

7.2 Paper II

Tracing the contributing sources of contamination is important in most environmental surveys. Based on the method described in Paper I and the main results of PCB in painted surfaces illustrated in Paper II, the environmental authorities in Norway and the other Nordic countries will pay more attention to exterior paint as a specific source of PCBs. When NGU and SFT chaired a PCB symposium in Trondheim in January 2008, delegates from the Swedish and Danish EPA presented their strategies for PCB regulations. Most attention regarding PCB in the environment in these two countries involves electrical equipment and sealants, and not paint in particular (Danish EPA; Swedish EPA). The relative gross weight of paint at each location may not be so high, but considering the surface area in each case, the amount of possible release of PCBs (if the paint contains PCBs) may be substantial. Other studies have discussed the content of metals in paints and dust from renovation projects, and the subsequent health effects of removal by e.g. power sanding (Mielke et al., 2001). Crude estimates of the possible release of PCBs from painted structures are presented in Paper II. The surface area of the school studied in detail in Paper II is approximately 2000 m^2, and a rough estimate based on the max-concentration of PCB_7 indicates that this building alone may contribute to 19 kg PCB_7 if all is distributed to the environment through flaking or active remediation. Similar, if we assume that the bridge Puddefjordsbroen studied in detail in Paper II was covered with PCB-paint until sandblasted in the mid-80s, the contribution from this structure alone may have been as much as 1000 kg PCB to the surrounding area in a worst-case scenario. Flaking paint is an effective way of PCB-dispersion, and these are only two cases studied in detail from the city of Bergen alone. In addition, Ruus et al. (2006) found high concentrations of PCBs in cod and blue mussels outside an old, renovated power station in Sørfjorden, Norway. High concentrations in the surface building materials, such as paint and plaster, were found to be the main source. The three case studies mentioned above, combined with the other results of PCB in paint presented in Paper II, indicate that there are reasons to believe that the potential of PCB-dispersion from painted surfaces may be large, especially in the Bergen area. NGU in collaboration with SFT have initiated (April 2008) a new

project based partly on the results presented in Paper II, to provide a national estimate on PCBs in building surfaces. In 5 Norwegian cities a total of 100 buildings, selected by age (constructed between 1950 and 1980), will be included in the study.

7.3 Paper III

Studies of PCBs in Arctic areas have been carried out to a great extent since the early 1990s, focusing especially on the effects on animal health (e.g. Dietz et al., 2000; Verreault et al., 2005). Traditionally, levels of PCBs in Arctic areas have been accredited to long-range transport (Wania and Mackay, 1995; Kallenborn et al., 2007), however local sources such as mining activities, sewage, and waste dumps have been mentioned as possible contributors to the contamination of arctic marine sediments outside settlements on Svalbard (Hop et al., 2001; Evenset et al., 2006). Detailed studies of active PCB-sources and their effect on the environmental condition of surface soil within areas heavily affected by anthropogenic activity was initiated by NGU, SFT, and the Governor of Svalbard in 2007, resulting in the major findings presented in Paper III. The results of this study indicated that the concentration of PCB_7 found in surface soils in the Russian settlements of Barentsburg and Pyramiden by far exceed what is previously found in studies from the Norwegian mainland (Andersson et al., 2004; Jartun et al., 2008). Svalbard is to be one of the best managed wilderness areas in the world (Lundkvist et al., 2008). Directions for further work indicated in Paper III include controlled management of all the waste (electrical, building materials) lying in open terrain in Barentsburg and Pyramiden. Several new projects have been proposed based on the results presented in Paper III, including a) studies of PCB sources in other settlements and mining areas, b) studies of the contribution of local sources of PCBs to air, c) studies of the direct flux of pollutants from settlements via rivers and snowmelt, and d) studies of the effect of local sources upon nesting kittiwakes and glaucous gulls in the Barentsburg area.

8 Summary of papers included in the Thesis

8.1 Paper I

Jartun, M., Ottesen, R.T., Steinnes, E. and Volden, T., 2008. Runoff of particle bound pollutants from urban impervious surfaces studied by analysis of sediments from stormwater traps. Sci. Total Environ. 396: 147-163.

Sediments from 68 stormwater traps collecting materials from limited catchments were analyzed, and the content of 31 inorganic substances (including heavy metals), PCB_7, PAHs, TOC, and grain size were determined. All locations were selected in the interface between the urban impervious environment of Bergen, Norway, and the adjacent harbor. The main objectives were a) to develop a suitable method for investigating the environmental impact of urban contamination in the direction of the marine recipient, and b) to describe the ongoing dispersion of pollutants from active sources within the catchments represented by elevated concentrations in the urban runoff sediments from stormwater traps.

The results indicated that pollutants such as PCBs, Pb, Cd, and Zn are actively dispersed in the impervious urban environment facilitated by the stormwater system. High concentrations of PCBs were found in over 20 % of the samples indicating several active sources of PCB contamination within the sampling area. Analysis of grain sizes indicated in general that up to 90 % of the sediment consists of particles of a size fraction suitable for suspended transport via the stormwater system. The method of studying active dispersion of pollutants in the urban environment described in this paper will provide significant information that may be used to reveal upstream contamination sources, and further give an indication on the real time dispersion to a downstream recipient such as the harbor sediments.

8.2 Paper II

Jartun, M., Ottesen, R.T., Steinnes, E. and Volden, T. Painted surfaces – important contamination sources of polychlorinated biphenyls (PCBs) to the urban and marine environment. Accepted for publication in Environmental Pollution, June 2008.

Based on the results presented in Paper I (Jartun et al., 2008), 68 samples of flaking paint were collected from buildings within specific catchments corresponding to high concentrations of PCB_7 found in sediments from stormwater traps. The main objective of this study was to discuss the environmental impact of painted surfaces as one possible source of PCBs to other compartments such as surface soil, urban runoff, and consequently the marine sediments. We divided the study into three single approaches,

1) a general study of flaking paint from both random and selected buildings within the urban environment,
2) a detailed study of 23 single samples from one specific building complex previously found to contain PCBs, and
3) a case study of the PCB content in both surface and inner concrete samples from a major bridge constituting a significant surface area.

Painted surfaces as a major source of PCBs have not previously been studied in detail. The results of our study indicate that more attention should be paid to this specific application because of the potential of dispersion facilitated by weathering, flaking, and stormwater transport. Concentrations of PCB_7 up to 3390 mg/kg were found in the exterior paint of a day care centre in Bergen. Results from the second approach indicate that a building complex may consist of several types of paint exhibiting different patterns of PCB contamination. This is an important aspect in regards to the treatment of future renovation waste from such buildings. One sample is probably not enough to provide a sufficient status of the environmental condition of one particular building. Results from the third approach mentioned above show the concentration of PCB_7 in the surface and inner concrete of a bridge previously covered with a white layer of paint that apparently contained high concentrations of PCB. The bridge was sandblasted in the mid 1980s, and high concentrations of PCB_7 have been found in samples of surface soil and marine sediments in the immediate vicinity. There were still high concentrations of PCBs in the surface material (plaster, remains of paint) of the bridge, whereas the inner concrete was proven clean. Still, in the worst case scenario this bridge may have contributed to the dispersion of more than 1000 kg of PCB_7 alone because of the surface area of over 11 000 m^2.

8.3 Paper III

Jartun, M., Ottesen, R.T., Volden, T. and Lundkvist, Q. Local sources of polychlorinated biphenyls (PCBs) in Russian and Norwegian settlements on Spitsbergen Island, Norway. Submitted to Journal of Toxicology and Environmental Health, May 4th 2008.

The third paper accompanying this thesis brings the Urban Risk philosophy to the Arctic, studying the effects of possible local sources of PCBs within areas affected by anthropogenic activity. Traditionally, long-range transport, represented by dispersion via air or ocean currents, has been proposed as the main cause of PCB levels found in samples of air, soil, or biota on Svalbard. In our study, samples of surface soil and various products such as paint, concrete, small capacitors, and transformer oils were collected from Barentsburg, the abandoned Pyramiden, and Longyearbyen, the three largest settlements on Spitsbergen Island, Svalbard, Norway. Surface soils in the two Russian settlements are highly contaminated with PCB_7 compared to both the Norwegian settlement of Longyearbyen and other Norwegian cities. Main sources include flaking paint, as previously mentioned in Paper II, and oil from small capacitors. Large amounts of electrical waste and abandoned, decaying buildings have contaminated large areas of soil, which subsequently is readily available for fluvial erosion during snowmelt and summer season.

9 Additional work performed during the Ph.D.-study

During the four years of my Ph.D.-study I have been most privileged to gain further experience and knowledge from several projects not directly (but still interconnected somehow…) associated with the thesis or Papers I-III presented here. The list completes the professional work carried out between 2004 and 2008.

9.1 Publications (alphabetical)

Andersson, M., Jartun, M. and Volden, T., 2004. Miljøundersøkelse av spredning av miljøgifter fra snødeponiet i Ilabekken *(Environmental survey of the dispersion of pollutants from a snow dumping site draining to Ilabekken creek)*. NGU-report 2004.041 (in Norwegian).

Andersson, M., Volden, T. og Jartun, M., 2005. PCB i asfalt i Trondheim *(PCB in asphalt from Trondheim)*. NGU-report 2005.045 (in Norwegian).

Andersson, M., Haugland, T., Jartun, M. and Jensen, H., 2006. Kartlegging av jordforurensning i 46 barnehager i bydel Alna (*Soil pollution in 46 day-care centers from the district of Alna, Oslo)*. NGU-report 2006.086 (in Norwegian).

Andersson, M., Haugland, T., Ottesen, R.T., Volden, T. and Jartun, M., 2006. Kartlegging av jordforurensning i 18 barnehager i bydel Grünerløkka (utenfor Ring 2) *(Soil pollution in 18 day-care centers from the district of Grünerløkka, Oslo)*. NGU-report 2006.064 (in Norwegian).

Bølviken, B., Bogen, J., Jartun, M., Langedal, M., Ottesen, R.T. and Volden, T., 2004. Overbank sediments: a natural bed blending sampling medium for large-scale geochemical mapping. Chemom. Intell. Lab. Syst. 74: 183 – 199.

Eggen, O.A., Haugland, T., Finne, T.E. and Jartun, M., 2006. Kartlegging av jordforurensning i 58 barnehager i bydel Østensjø *(Soil pollution in 58 day-care centers from the district of Østensjø, Oslo)*. NGU-report 2007.016 (in Norwegian).

Haugland, T., Ottesen, R.T., Volden, T. and Jartun, M., 2005. Jordforurensning i OBY-barnehager innenfor Ring 2 *(Soil pollution in Oslo day-care centers within city limit 2)*. NGU-report 2005.064 (in Norwegian).

Haugland, T., Andersson, M., Volden, T. and Jartun, M., 2006. Kartlegging av jordforurensning i 25 barnehager i bydel Gamle Oslo (utenfor Ring 2) *(Soil pollution in 25 day-care centers from Gamle (Old) Oslo).* NGU-report 2006.063 (in Norwegian).

Jartun, M., 2005. PCB – kilder og spredning fra urbane områder *(PCB – sources and dispersion from urban areas)*. Miljø og helse 01/05: 20 – 21 (in Norwegian).

Jartun, M. and Volden, T., 2005. Miljøtilstanden ved opplagsplasser og pussesteder ved 11 småbåthavner i Trøndelag *(The environmental condition at 11 storage yards and small boat harbors in Trøndelag)*. NGU-report 2005.012 (in Norwegian).

Jartun, M., Ottesen, R.T. and Volden, T., 2005. Spredning av miljøgifter fra tette flater i Bergen *(Dispersion of pollutants from impervious surfaces in Bergen)*. NGU-report 2005.051 (in Norwegian).

Jartun, M., Volden, T. and Ottesen, R.T., 2005. PCB-innhold i sandfangsmasser i Bergen – foreløpige resultater *(PCB content in urban runoff sediments from Bergen – preliminary results)*. VANN 1/05: 29 – 34 (in Norwegian).

Jartun, M. and Volden, T., 2006. Jordforurensning i Harstad *(Soil pollution in Harstad)*. NGU-report 2006.014 (in Norwegian).

Jartun, M., Volden, T. and Alexander, J., 2006. Jordforurensning i Odda *(Soil pollution in Odda)*. NGU-report 2006.023 (in Norwegian).

Jartun, M. and Jørgensen, T., 2006. Kartlegging av PCB, PAH og tungmetaller i asfaltdekker fra områdene Kristiansand, Oslo og Bergen *(PCB, PAH, and heavy metals in asphalt coatings from Kristiansand, Oslo, and Bergen)*. NGU-report 2006.029 (in Norwegian).

Jartun, M., 2006. Datarapport fra oppfølgende undersøkelser av PAH (16)-konsentrasjoner i 3 asfaltkjerner fra Kristiansand og Oslo *(Data report from a follow-up study of asphalt in Kristiansand and Oslo)*. NGU-report 2006.065 (in Norwegian).

Jartun, M. and Volden, T., 2007. PCB i nedbør fra Bergensområdet *(PCB in precipitation from the Bergen area)*. NGU-report 2007.074 (in Norwegian).

Jartun, M., Volden, T. and Ottesen, R.T., 2007. PCB fra lokale kilder i Barentsburg, Pyramiden og Longyearbyen på Svalbard (PCB from local sources in Barentsburg, Pyramiden, and Longyearbyen in Svalbard). NGU-report 2007.075 (in Norwegian).

Lundkvist, Q., Pedersen, H.R., Ottesen, R.T., Volden, T., Jartun, M., Gabrielsen, G.W., Skåre, J.U., Kallenborn, R., Ruus, A., Dahle, S., Evenset, A., Vongraven, D., Jenssen, B.M., Ekker, M. and Hindrum, R., 2008. PCB on Svalbard, status of knowledge and management, April 2008. Governor of Svalbard Report 1/2008. Also available in Norwegian and Russian.

Mielke, H.W., Gonzales, C.R., Powell, E., Jartun, M. and Mielke, P.W., 2007. Nonlinear association between soil lead and blood lead of children in metropolitan New Orleans, Louisiana: 2000-2005. Sci. Total Environ. 388: 43-53.

9.2 Lectures (chronological)

The Norwegian Society for Soil and Sediment Network (Miljøringen). Seminar: Undersøkelser av forurenset grunn og forurensede sedimenter *(Studies of polluted soil and sediments)*, Sandefjord June 7-8 2004. Title: Er beslutningsgrunnlaget godt nok i dagens miljøtekniske undersøkelser *(Is the decision basis well-founded in todays environmental surveys?)*

Environmental symposium in Harstad, January 2005. A two hour lecture: Forurenset jord – forurensede sedimenter *(Polluted soil – polluted sediments)*.

The 14th national seminar on hydrogeology and environmental geochemistry at NGU, February 8-9 2005. Title: *News from NGU.*

Norwegian-Swedish environmental meeting, Strømstad, September 4-7 2005. Title: Urban Risk – spredning av miljøgifter fra by til havnebasseng *(Urban Risk – dispersion of pollutants from city to harbor)*

The Norwegian Society for Soil and Sediment Network (Miljøringen) seminar on Sources of pollutants, Bergen, November 9-10 2005. Title: Spredning av miljøgifter fra tette flater i Bergen *(Dispersion of pollutants from impervious surfaces in Bergen)*.

The 15th national seminar on hydrogeology and environmental geochemistry at NGU, February 7-8 2006. Title: Kartlegging av PCB i asfaltdekker *(Mapping of PCBs in asphalt)*.

Dioxin 2006 – international conference, Oslo August 2006. Title: *Sources and dispersion mechanisms of PCBs in an urban environment*.

Seminar on educational science, NTNU, Trondheim November 24th 2006. Title: Gifter i våre omgivelser – utfordringer for miljø og helse *(Pollutants in our surroundings – implications for environment and human health)*.

Tulane University, New Orleans KERRN and CBR seminar series, May 15th 2007. *How safe is your playground? – A look into the health of soils in playgrounds*.

Interdisciplinary workshop – PCB on Svalbard, NGU January 9-11 2008. Title: PCB fra lokale kilder på Svalbard *(PCBs from local sources on Svalbard)*.

Nordic seminar on PCBs – Quo vadis? NGU January 30-31 2008. Title: PCB - spredning fra land til det marine miljø *(PCB – dispersion from urban areas to the marine environment)*.

Lecture in a short-course on PCBs in building materials. Glassfabrikken, Larvik, February 6th 2008. Title: PCB – vidundermiddel og miljøgift *(PCB – from wonder compound to pollutant)*.

2nd Norwegian Environmental Toxicology Symposium, NTNU Trondheim April 2-4 2008. Title: *Local sources of PCB contamination in Svalbard*. Awarded best presentation.

9.3 Posters

4th European Meeting on Environmental Chemistry (EMEC 4), Plymouth, England, December 10 – 13 2003. Title: *Arsenic contamination of Norwegian playfields*.

The 5th SEDNET conference, Oslo, May 27-29 2008. Title: Urban Risk – Runoff sediments from small urban catchments – tracing active sources of toxic pollutants.

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Project manager for 307300 Urban Risk (2004 – this day).

Project manager for 323900 PCBs from building surfaces in Norway (April 2008 – this day).

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Paper I

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Runoff of particle bound pollutants from urban impervious surfaces studied by analysis of sediments from stormwater traps

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ABSTRACT

Runoff sediments from 68 small stormwater traps around the harbor of urban Bergen, Norway, were sampled and the concentrations of polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), heavy metals, and total organic carbon (TOC) were determined in addition to grain size. Our study provides empirical data from a large area in the interface between the urban and marine environment, studying the active transport of pollutants from land-based sources. The results of the analyses clearly demonstrate the importance of the urban environment representing a variety of contamination sources, and that stormwater runoff is an important dispersion mechanism of toxic pollutants. The concentrations of different pollutants in urban runoff sediments show that there are several active pollution sources supplying the sewage systems with PCBs, PAHs and heavy metals such as lead (Pb), zinc (Zn) and cadmium (Cd). The concentration of PCB_7 in the urban runoff sediments ranged between <0.0004 and 0.704 mg/kg. For PAH_{16}, the concentration range was <0.2–80 mg/kg, whereas the concentration ranges of Pb, Zn and Cd were 9–675, 51.3–4670 and 0.02–11.1 mg/kg respectively. Grain size distribution in 21 selected samples varied from a median particle diameter of 13 to 646 μm. However, several samples had very fine-grained particles even up to the 90 percentile of the samples, making them available for stormwater dispersion in suspended form. The sampling approach proposed in this paper will provide environmental authorities with a useful tool to examine ongoing urban contamination of harbors and similar recipients.

1. Introduction

The urban environment is affected by a wide variety of local, anthropogenic activities, and this contamination is a continuous and diffuse process. Road networks, housing, and industrial activities will tend to increase the contents of heavy metals and organic pollutants in urban soils (Kelly et al., 1996; Mielke et al., 1999; Ottesen and Langedal, 2001; Mielke et al., 2004). Various types of soil may be present within the urban environment, ranging from relatively undisturbed soils, similar in some respects to their rural counterparts (Bridges, 1991; Hollis, 1991), to completely man-made soils (Ottesen and Langedal, 2001). The most significant impact of urbanization on soil is the complete loss or burial as a result of construction activities. Assuming that an average proportion of between 50 and 100% of the urban area has been built upon, this represents a considerable volume of buried or displaced soil in addition to an important change in the hydrological

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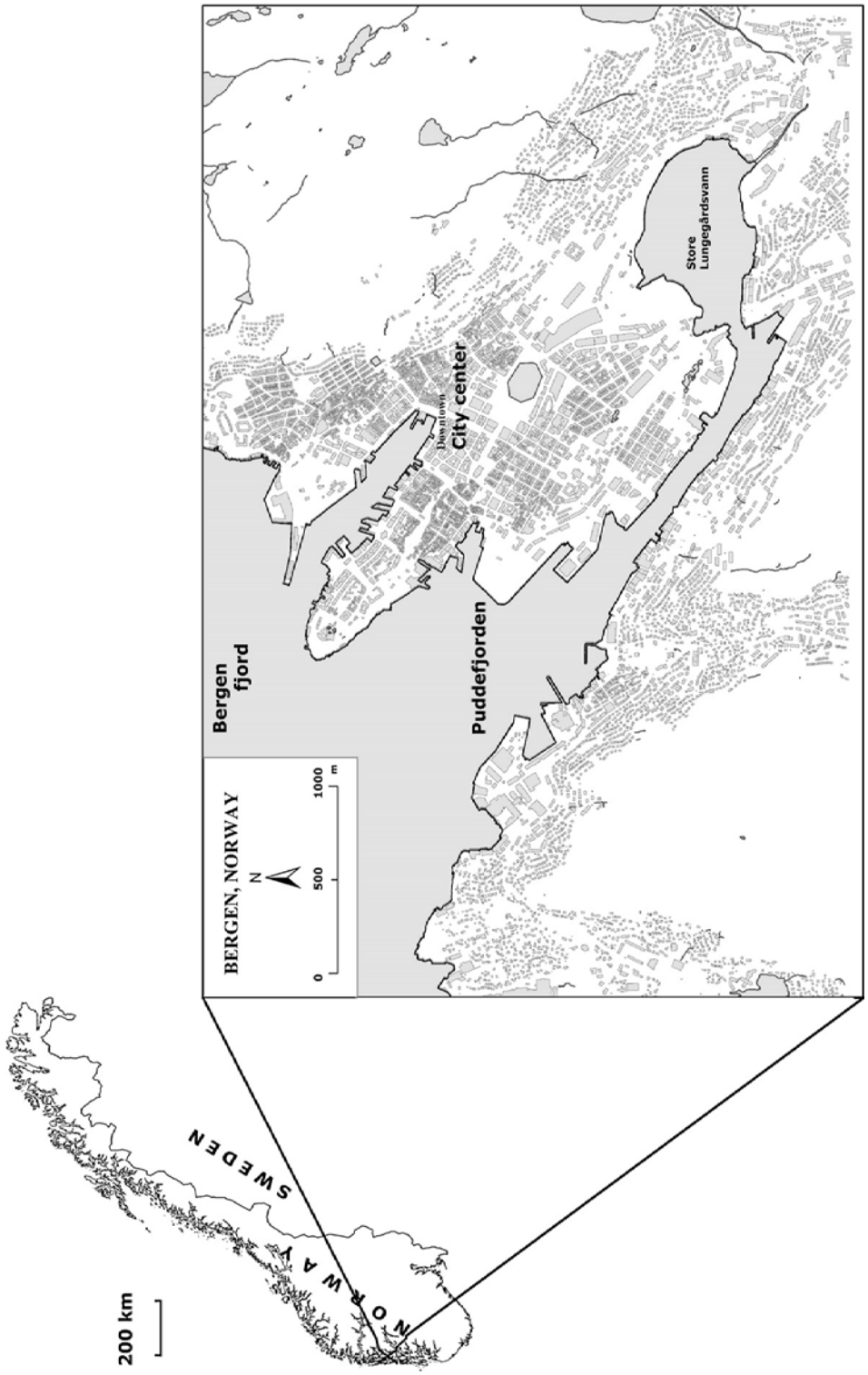


Fig. 1 – Survey area around Bergen harbor, located in the western part of Norway.

distribution of urban surface water. Impervious areas such as roofs, asphalt roads, and concrete will allow only a small amount of stormwater to be infiltrated, evaporated, detained, or retained by vegetation, which will cause episodes of high-volume runoff followed by problems with the hydraulic efficiency of old sewer systems and consequently a direct discharge to a downstream recipient (Mays, 2001). The chemical properties of stormwater are dependent on factors such as the nature of surfaces in the urban environment, anthropogenic activities, and natural processes within each catchment (Eriksson et al., 2007). The impervious areas of a typical urban environment will often constitute 60–100% of the total area (Lindholm, 2004; Lu and Weng, 2006).

There is currently an environmental focus on polluted sediments in harbors and fjords in Norway. Marine sediments and organisms from 31 harbors and fjords in Norway show concentration levels of pollutants such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and heavy metals that by far exceed the guideline levels of the Norwegian Pollution Control Authority (SFT) and the Norwegian Food Safety Authority (Molvær et al., 1997; Kryvi et al., 2005; Økland, 2005). This is also the situation in Bergen harbor, around which our survey of stormwater sediments was carried out. In each of the affected areas, a dietary advisory against the consumption of certain fish and crustaceans exists, and there may also be local restrictions on sale of fish and shellfish because of the bioaccumulation in marine benthic organisms (Ruus et al., 2005; Økland, 2005). It is of importance to reduce or stop the input of pollutants from land-based sources to the marine environment. Projects all over the world, including Norway, are being conducted to assess the remediation of marine sediments (Hauge et al., 1998; Kryvi et al., 2005; Förstner and Apitz, 2007). So far the most important sources of contaminated sediments are believed to be industrial activities or direct emissions to the harbor areas. The significance of the urban environment and runoff from impervious surfaces has until now only been recognized as one of many diffuse sources with few scientific studies backing up the actual contribution to the contamination of harbor sediments. In 2004, a project called "Urban Risk" was initiated by the Geological Survey of Norway to provide more information on:

- the concentration of arsenic, heavy metals, PAHs and PCBs in sediments from small stormwater sediment traps
- the identification of active contamination sources in the urban environment
- the sediment transport in runoff from impervious surfaces

A detailed chemical characterization of the materials accumulated on top of impervious surfaces and potentially available for transport with urban stormwater is important when assessing the risk of contamination of nearby water bodies, such as the city harbor. The contribution of different pollution sources in the local urban environment to receiving waters has been discussed by Davis et al. (2001) and Chebbo et al. (2001). Contamination from traffic (exhaust, oil spills, wear and tear of tires, vehicles), buildings (weathering, renovation, demolition, paint, plaster), and city fires may be stored in urban soils and sediments of urban water basins (Davis and Burns, 1999; Ottesen and Langedal, 2001; Jartun et al., 2003; Andersson et al., 2004). Large-scale surveys to describe urban soil pollution in Norway have been carried out by the Geological Survey of Norway in cooperation with the major cities (Ottesen et al., 1995; Ottesen and Volden, 1999; Ottesen et al., 2000a; Ottesen and Langedal, 2001; Langedal and Ottesen, 2001; Jartun et al., 2003; Andersson et al., 2004). A number of urban runoff studies carried out during the 1970s and 80s focused on physical, chemical and biological characteristics of particular storm events (Field, 1975; Field and Lager, 1975; Zanoni, 1986). Xanthopoulos and Hahn (1990) found high concentrations of pollutants in urban storm water in both dissolved and particulate form. Sutherland and Tolosa (2000) described the contaminant load of road-deposited sediment (RDS) and the remobilization and transport of especially Pb to receiving water bodies. Another study of bed sediments from an urban stream showed a strong contamination signal for Pb, and the pollution was associated with traffic as the main source (Sutherland, 2000). Later studies have measured the concentrations of different components such as total and dissolved metals, polyaromatic hydrocarbons (PAHs), pesticides, and other organic pollutants in stormwater runoff (Soller et al., 2005). The mobilization of surface materials through urban storm events may result in significant discharge of pollutants into downstream recipients (Lee et al., 2002; Soller et al., 2005). The main objective of our study as a basis for possible identification of contamination sources in the urban environment was to collect empirical data on the contribution of different pollutants from an entire urban area and their potential of dispersion via the stormwater system to the marine environment.

2. Materials and methods

2.1. *Sample sites and sampling*

Bergen is the second largest city in Norway with 235,000 inhabitants, conveniently located at the base of seven mountains on the western coast of Norway (Fig. 1). It was founded as early as year 1070 around a harbor area in the inner Bergen fjord, exhibiting a coastal climate with an average of 2250 mm precipitation each year (Table 1). The rain does not necessarily fall evenly throughout the year. During a specific storm event in September 2005 the recorded amount of daily rainfall was 156 mm (the total annual amount of rain in 2005 was

Table 1 – Monthly average temperatures (°C) and precipitation (mm) from the inner city of Bergen, Norway

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
Temp °C	1.3	1.5	3.3	5.9	10.5	13	14	14	11	8.6	4.6	2.4	7.6
Precipitation mm	190	152	170	114	106	132	148	190	283	271	259	235	2250

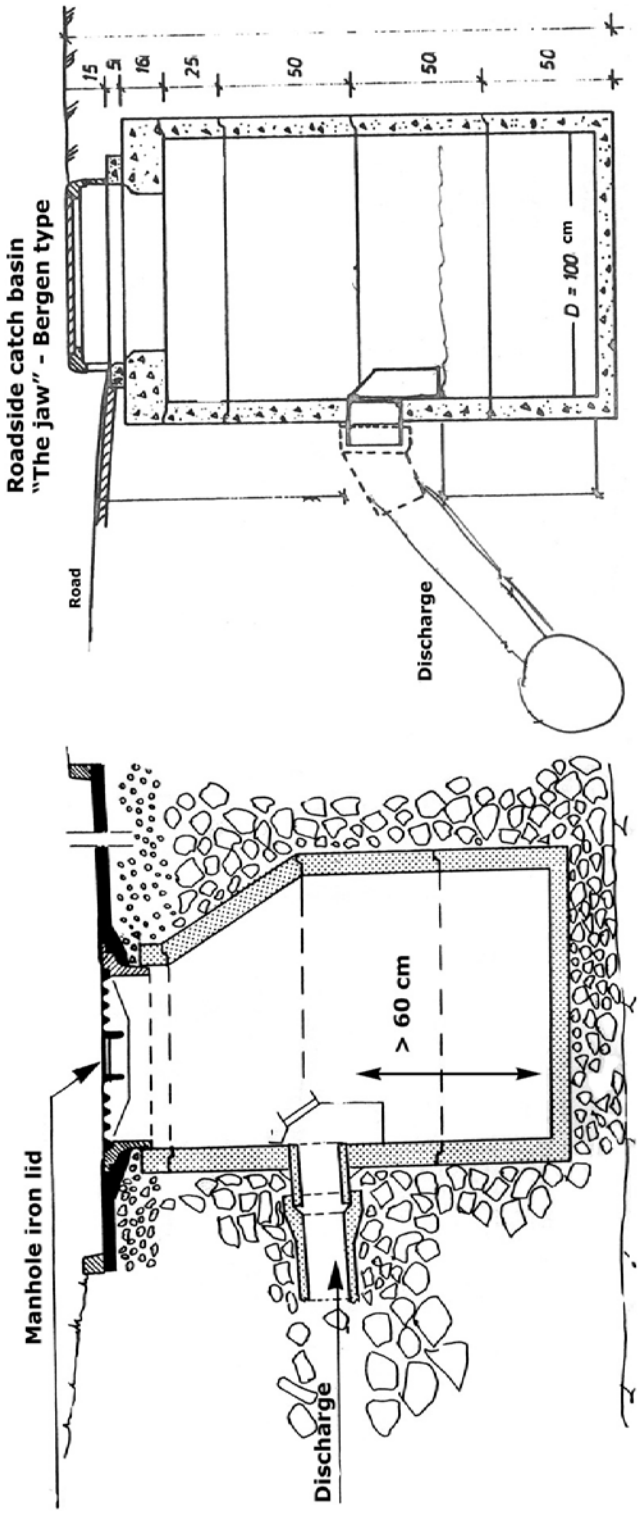


Fig. 2 – Two types of stormwater sediment traps; the typical parking lot type to the left, and the roadside sediment trap to the right.

3054 mm), creating large amounts of stormwater, which is directed to inlets in the impervious surfaces surrounding the Bergen harbor. Structures described in this paper as stormwater sediment traps (Figs. 2 and 3) will capture materials that are transported via urban runoff. The sedimentation rate will depend on particle size, specific gravity (SG) of particles, temperature, and the length of time until the next storm event (Butler et al., 1995). The stormwater sediment traps are features designed to slow down the stormwater before it enters into either the ground directly or into the combined or separated sewer system. However, in the areas adjacent to the harbor in our study area, the discharge from stormwater sediment traps may be released directly into the sea. The stormwater trap design may vary from country to country, and it is referred to in many papers as a gully pot which will collect urban sediments in a similar way (Pratt and Adams, 1984; Butler and Karunaratne, 1995).

The survey area around Bergen harbor consists of 80–100% impervious surfaces, such as streets, parking lots, and buildings. Given the amount of rainwater in the Bergen area, the overflow from impervious surfaces is discharged directly into the harbor about 30% of the time. The stormwater sediment traps will capture some of the suspended materials transported by stormwater, but this effect is reduced with decreasing particle diameter and stormwater intensity, as shown by Sansalone et al. (1998). The average catchment area for each inlet is about 600 m^2, and the maximum runoff distance on the surface is 50–80 m. Each stormwater sediment trap has a diameter of 1 m and a depth of about 3 m with an effective sediment trap of >60 cm located at the bottom of each stormwater sediment trap, giving each unit a capacity to capture 450–500 L of sediment. The total quantity of runoff sediments captured by the stormwater sediment traps in Bergen city is estimated to be about 13,000 metric tonnes annually. The municipality and the Norwegian Public Roads Administration collect sediments from stormwater sediment traps located in their respective areas of responsibility on an irregular basis, on average once a year (Bergen municipality, 1998-2004). This means that sediments found in each trap were deposited within the last year, which will reflect ongoing contamination from local sources in that particular catchment.

Sediments from 68 different stormwater sediment traps located in impervious urban surfaces around Bergen harbor were sampled in October and November 2004. Small catchments were selected in order to reflect local pollution sources, and stormwater sediment traps as close to the harbor as possible were selected to reflect the possible environmental load on the harbor sediments. The manhole cover for each unit was opened with an iron jack. Sediments deposited by the stormwater traps were collected using a long-handled aluminum scoop, which was cleaned with materials from the given trap to avoid cross contamination. Sample sizes of 1 L were collected in alcohol-cleaned white plastic containers with a tight cover and transported to the laboratory facility at the Geological Survey of Norway where all the containers were positioned on a bench top for the particles in suspension to settle. All samples were subsequently split into two equal fractions in wet condition. One fraction of each sample was sent to a commercial laboratory for the determination of PCBs and PAHs. The other fraction was air dried at 30 °C for 7 days and subsequently sieved through a 2 mm nylon screen. The <2 mm fraction was retained for determinations of metals, TOC, and grain size distribution.

2.2. *Chemical analyses*

To determine the PCB content, a representative sample of 10 g homogenized sediment was extracted with 20 mL acetone and 1 mL internal standard of PCB#53 (0.10 μg/mL). The extracts were then 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 using a rotavapor (50 °C) and then cleaned using 1 mL 2-propanol and 1 mL tetrabuthylammonium

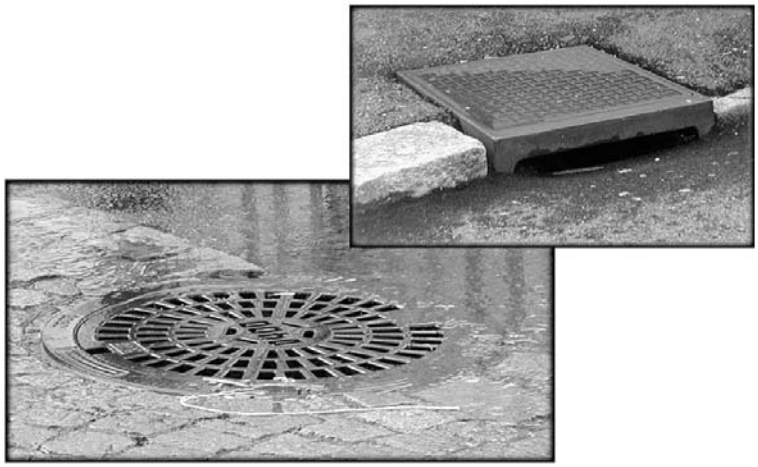


Fig. 3 – Pictures of characteristic inlets from impervious surfaces, the roadside stormwater trap (the Jaw), and the typical parking lot type (bottom).

Table 2 – Quality control of 9 selected elements (As, Cd, Cr, Cu, Hg, Ni, Pb, Sn, Zn), PAH_{16}, and PCB_7 in certified reference materials; MESS-3 from NRC[a] and RTC[b]

Compound	N samples	Certified value		Measured, mean	Relative error	SD	CV (%)
As	190	21.2	mg/kg	20.2	−4.85	1.86	9.22
Cd	176	0.240	mg/kg	0.230	−4.28	0.024	10.4
Cr	339	105	mg/kg	31.4	−70.1	2.00	6.37
Cu	343	33.9	mg/kg	27.8	−18.0	1.10	3.96
Hg	263	0.091	mg/kg	0.096	5.50	0.008	8.33
Ni	339	46.9	mg/kg	36.7	−21.8	1.40	3.81
Pb	343	21.,1	mg/kg	17.6	−16.6	1.40	7.95
Sn	15	2.50	mg/kg	1.57	−37.3	0.91	58.0
Zn	343	159	mg/kg	133.6	−16.0	5.60	4.19
PAH_{16}	23	96.3	mg/L	103.8	7.79	9.68	9.30
PCB_7	3	253	mg/L	238	−5.93	36.0	15.1

(CV = coefficient of variation).
[a] Concentration of certified reference materials from National Research Council of Canada for As and heavy metals in comparison to results achieved at the Laboratory of Geological Survey of Norway.
[b] Concentration of certified values from Resource Technology Corporation for PAH_{16} and PCB_7 in comparison to results received at AnalyCen.

hydrogen sulphate (TBA) with sodium sulphite in excess. 5 mL of deionized water was then added, the solution was thoroughly mixed, and the organic phase was finally cleaned using concentrated sulphuric acid. The quantification of 7 PCB congeners (PCB_7: IUPAC-numbers 28, 52, 101, 118, 138, 153, 180) was carried out using a Gas Chromatograph (Instrument HP6890) with electron capture detection (GC-ECD). The column used was a DB5 122-5062 with 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). A

Table 3 – Main results; statistical summary of selected chemical components in 68 samples of urban runoff sediments

Component	Unit	Detection limit	Min	Max	Arithmetic mean	Median
As	mg/kg	1	<1	56	6.5	3.7
Cd	mg/kg	0.02	0.02	11.1	1.13	0.42
Cr	mg/kg	0.2	11	135	30	25
Cu	mg/kg	0.5	16	6600	273	97
Hg	mg/kg	0.01	<0.01	2.81	0.20	0.06
Ni	mg/kg	1	7.4	309	32	24
Pb	mg/kg	1	9	675	126	61
Sn	mg/kg	3	<3	444	19	6
Zn	mg/kg	1	51.3	4670	698	403
TOC	%	0.1	0.4	39	5.2	4.3
PAH_{16}[a]	mg/kg	0.2	<0.2	80	7.6	3.4
PCB_7	mg/kg	0.0004	<0.0004	0.704	0.080	0.029
Al	mg/kg	20	4860	16,300	10,007	9595
B	mg/kg	5	<5	347	8.2	<5
Ba	mg/kg	1	70	841	191	156
Ca	mg/kg	200	2440	123,000	15,194	9215
Ce	mg/kg	2	10.6	80.7	30.8	29.7
Co	mg/kg	0.1	3.78	162	12.3	9.0
Fe	mg/kg	2	8940	75,400	18,676	17,200
K	mg/kg	100	750	4100	2393	2305
La	mg/kg	1	7.6	42	16	15
Li	mg/kg	1	2.1	53	8.0	7.2
Mg	mg/kg	100	1790	17,900	6415	6135
Mn	mg/kg	0.2	128	676	298	277
Mo	mg/kg	0.5	<0.5	72.9	4.4	2.4
Na	mg/kg	200	330	10,800	1073	733
P	mg/kg	10	398	2510	1046	1020
S	mg/kg	500	<500	183,000	7353	3695
Sc	mg/kg	0.1	0.3	3.2	1.9	1.8
Sr	mg/kg	1	28	335	66	57
Ti	mg/kg	1	269	1850	1027	962
V	mg/kg	1	12.6	69.0	33.2	30.8
Y	mg/kg	0.1	2.7	14.7	8.0	7.6
Zr	mg/kg	1	1.2	26	5.2	4.2

[a] PAH_{16} analysis: N = 57 samples.

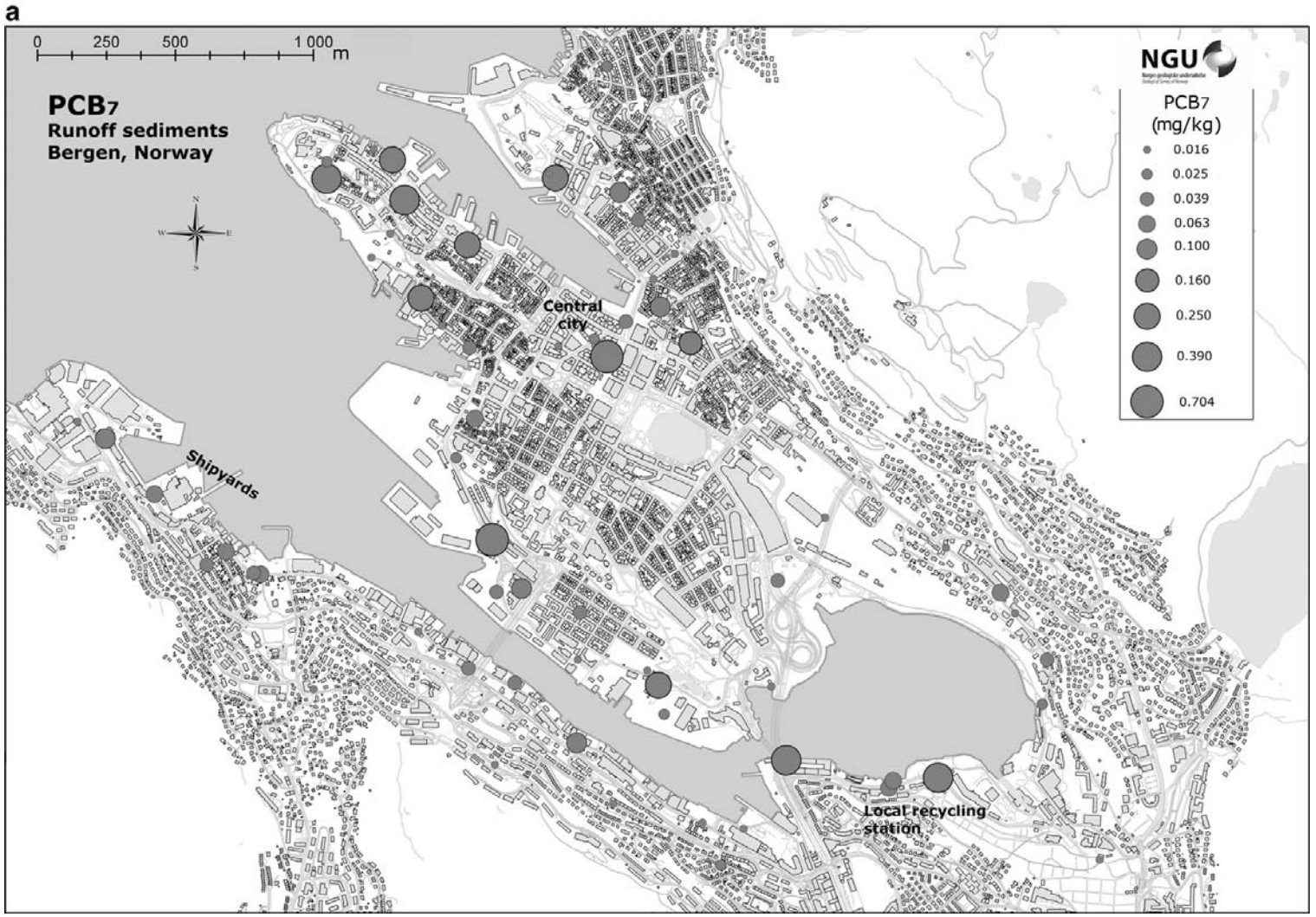
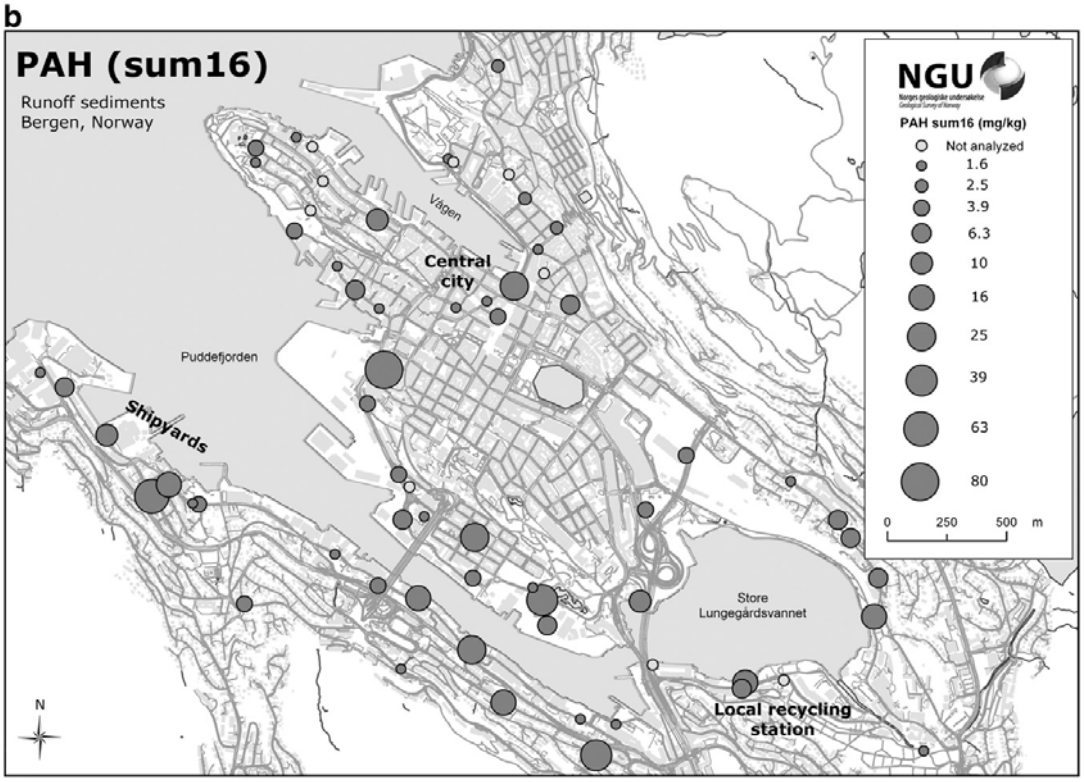


Fig. 4 – Geochemical maps showing the spatial distribution of selected environmental components in 68 samples of urban runoff sediments in Bergen, Norway. a) PCB_7, b) PAH_{16}, c) Cd, d) Hg, e) Pb, and f) Zn.



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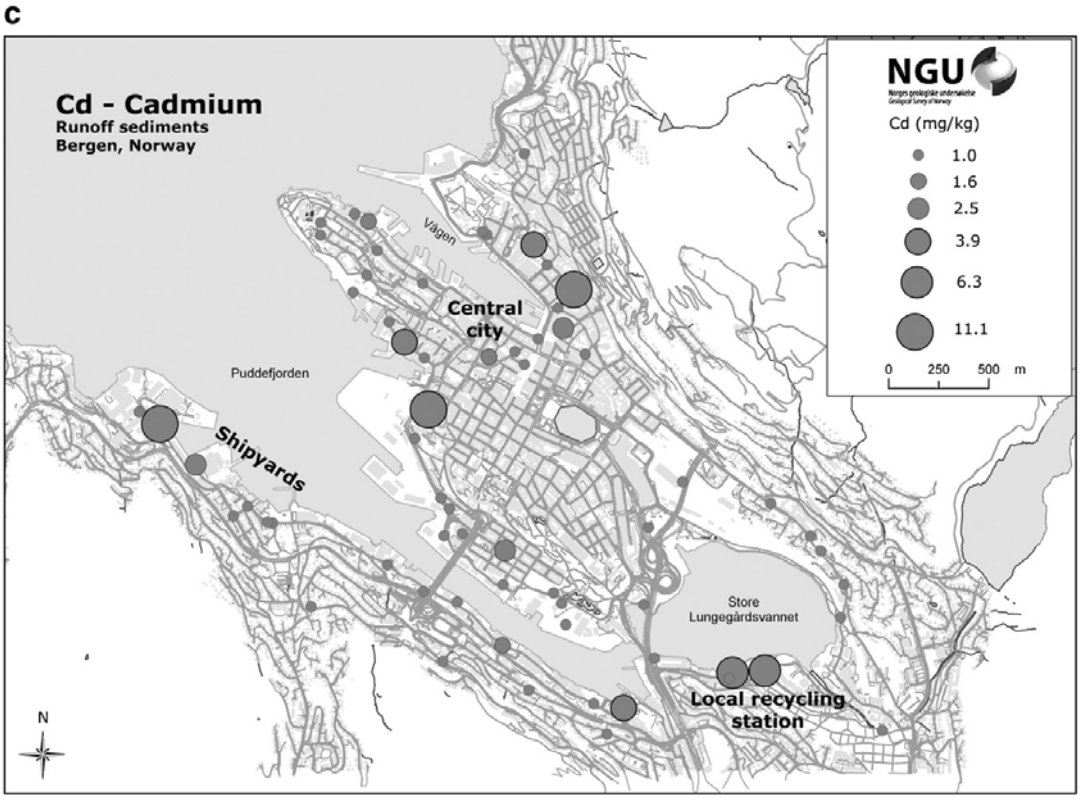
representative sample of about 40 g homogenized sediment was used for PAH analysis. Pyrophosphate (20 mL, 0.05 M) was then added to bring sediment clods to dissolve. Internal standards in this method were *d*-acenaphtene, *d*-phenanthrene and *d*-perylene solved in cyclohexane/ethylacetate from Larodan fine Chemicals. The 16 PAH compounds (PAH_{16}, see Table 2) were quantified using a GC-MS (HP 6890 — Agilent 5973) with splittless injection and a HP-5 5% phenyl methyl siloxane column. The method is based upon the Nordtest Technical Report No. 329 (Karstensen et al., 1997). The PCB and PAH analyses were performed at AnalyCen AS, Moss, Norway. The laboratory is accredited for hydrocarbons analyses according to the European standards of NS-EN ISO 17025, NS-EN ISO 9001, and NS-EN ISO 14001. The precision of the analytical method is 15–20%. For quality control a certified reference material bought from Resource Technology Corporation (RTC) was analyzed regularly. Table 2 shows the certified values in comparison to the values received at AnalyCen for replicate analyses carried out over a period of 5 years.

All the analyses for metals, TOC, and grain size were performed at the Geological Survey of Norway Laboratory. The digestion of a 1.000 g dry sample in the <2 mm fraction for metal analysis was done with 20 mL 7 N nitric acid in an autoclave for 30 min at 120 °C, according to NS 4770 (Norwegian Standard, 1994). The solution was analyzed using a Perkin Elmer Optima 4300 Dual View ICP-AES (Inductively Coupled Plasma Atomic Emission Spectrometer) for 31 elements (Al, As, B, Ba, Ca, Cd, Ce, Co, Cr, Cu, Fe, Hg, K, La, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sc, Sn, Sr, Ti, V, Y, Zn, and Zr) and a Perkin Elmer SIMAA 6000 graphite furnace atomic absorption spectrometry (GFAAS) for As, Cd, and Sn. Hg was determined using a CETAC M-6000A Hg Analyzer (cold vapor atomic absorption spectrometry, CVAAS). Quality control for 9 selected elements (As, Cd, Cr, Cu, Hg, Ni, Pb, Sn, Zn) determined by analysis of certified marine sediments (MESS-3) from NRC is shown in Table 2.

Dry samples of the <2 mm fraction were treated with hydrochloric acid prior to analysis of total organic carbon (TOC) to remove inorganic (carbonate) carbon. TOC was determined with a LECO SC-444 instrument. The uncertainty of the TOC measurement is ±15% up to 3% TOC, and ±10% when concentration of TOC exceeds 3%, determined by analysis of a house standard. Blanks were included in every batch. Grain size, or particle size, distribution was performed on a random selection of 21 samples of the original 68 by laser diffraction using a Coulter LS 200 instrument. The analysis was done on dry material within particle diameter range of 0.4–2000 μm. The results are presented on the basis of volume, by a cumulative volume percentage. The uncertainty of this measurement is ±3%.



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2.3. *Principal component analysis (PCA) and mapping*

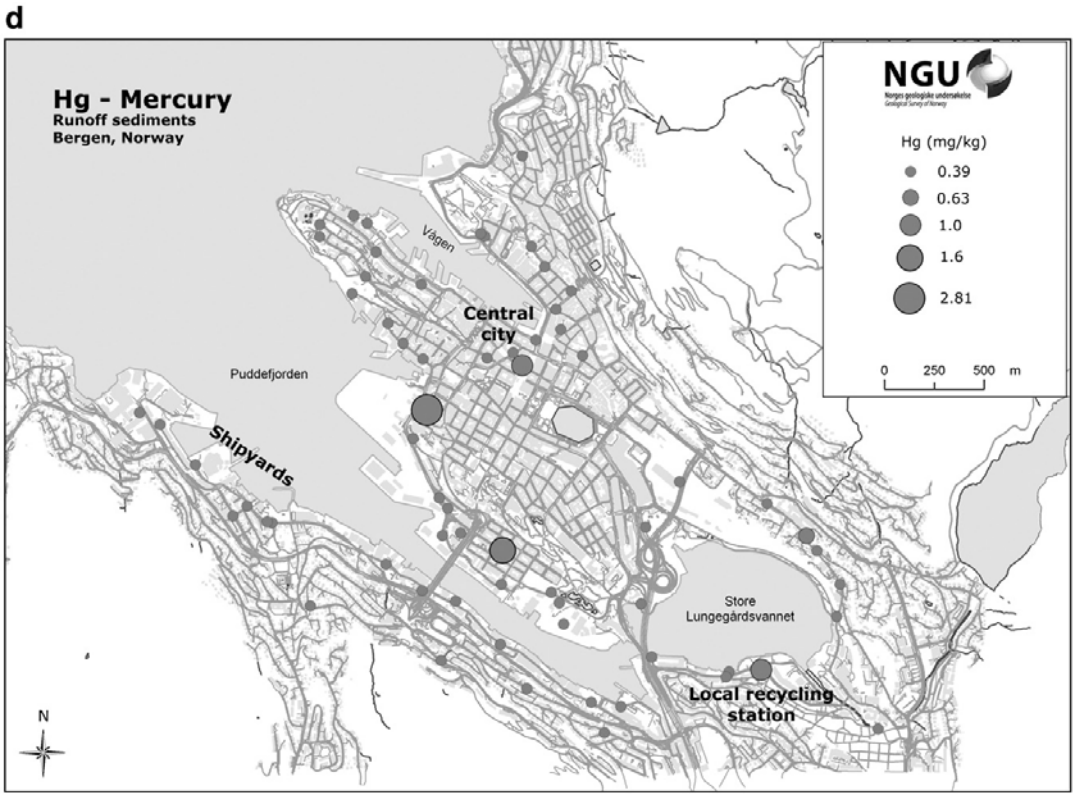
Principal component analysis (PCA) was performed on the complete dataset in addition to simple statistical description of the data including cumulative frequency distributions. PCA was conducted using Simca-P version 10.0.4.0 by Umetrics. UTM-coordinates and the results of chemical analysis have been loaded into ArcMap™ to visualize the spatial distribution in the Bergen city area. The first class of concentration ranges on the maps, represented by the smallest dot, includes 30 percentile of the results for each component. A 5-point division of the logarithmic scale defines the subsequent concentration classes.

3. Results

Analytical results of the urban runoff sediment samples are summarized in Table 3 listing the detection limits, the concentration ranges (min–max), arithmetic mean and median concentrations for heavy metals, PCB_7, PAH_{16}, and TOC.

PCB_7 was detected in 63 out of 68 samples. This was somewhat of a surprise compared to previous studies of urban soil in both Bergen and other Norwegian cities where the concentrations of PCB_7 were relatively high in a few samples while most of the samples were below the detection limit (Ottesen and Volden, 1999; Andersson et al., 2004). 14 out of 68 samples had concentrations of PCB_7 above 0.100 mg/kg dw, which is considered heavily polluted for Norwegian marine sediments (Molvær et al., 1997). The concentration of PAHs found in the urban runoff sediments is within the expected range, based on studies of urban soil pollution in Norwegian cities (Ottesen and Volden, 1999; Jartun et al., 2003) but relatively low compared to a study of urban soil from New Orleans, USA, a much larger city than Bergen (Wang et al., in press). The median concentration of PAH_{16} in urban runoff sediments from Bergen is 3.4 mg/kg dw compared to as high as 11.2 mg/kg dw for a median surface soil PAH_{16} value of an inner city community in New Orleans. It is important to notice that these two sample media are different in composition, but will be contaminated by the same sources in an urban environment.

Spatial variation of selected pollutants within the urban sampling area is shown in Fig. 4a–f. The geographical distributions of the pollutants are shown with ArcMap™ geochemical maps of the urban environment surrounding Bergen harbor. The stormwater sediment trap locations are shown as red dots. Fig. 4a gives the distribution of PCB_7, and the general impression is that the concentrations are fairly low. However, the map clearly shows that there are several



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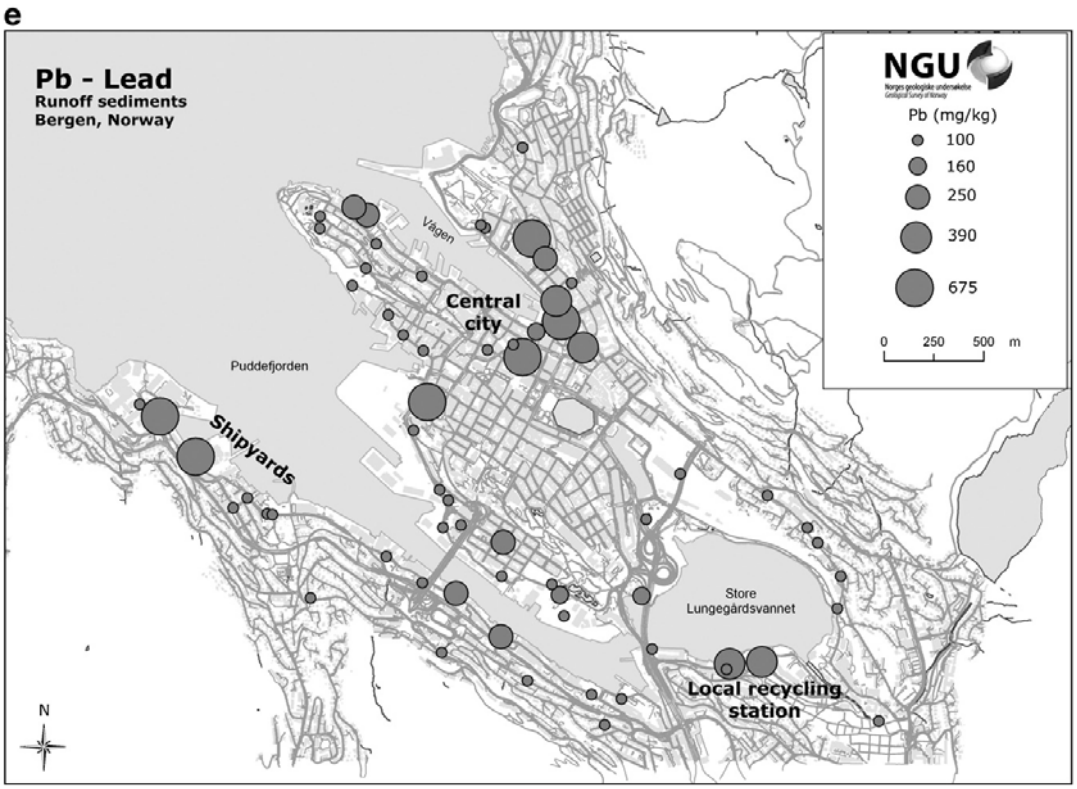
active sources of PCBs close to the harbor. This corresponds to earlier studies of PCBs in the Bergen area, showing that some building facades of the same type and age are contaminated with PCBs and others are not (Andersson et al., 2004). However, there are no previous studies of specific PCB-sources within the small catchments in the interface between the urban and marine environment. The distribution of PAHs in the urban environment is characterized by the same pattern as the PCBs (Fig. 4b). Fig. 4c–f represents the geographical distribution of four selected heavy metals (Cd, Hg, Pb, and Zn, respectively) close to the Bergen harbor area. High concentrations of Cd and Pb are found in selected areas representing areas such as old, central parts of the city, shipyards, and small areas with recycling stations for household waste. Leaded gasoline, lead based paint, batteries, and wheel weights are well known sources of Pb in an urban environment (Nriagu, 1990; Root, 2000; Laidlaw et al., 2005; Mielke, 2005), while high concentrations of Cd follows the contamination patterns as Zn. Zn is found in high concentrations widely distributed in the whole sample area, whereas the concentration of Hg is slightly elevated in only one single sample (concentration 2.81 mg/kg) of urban runoff sediments close to an excavation site in the inner city. The urban runoff sediments trapped by stormwater traps are mostly inorganic, with a median TOC content of 4.3%, ranging from 0.4–39%. The extreme value of 39% TOC is explained by deposition of waste from a renovated building, such as pressed wallboard and insulation draining to that particular stormwater sediment trap. The content of TOC in the urban runoff sediments appears to correlate well with PCB_7, PAH_{16}, and Hg, but not so well with other pollutants such as Pb, Zn, and Cd. A thin film of oil was also seen in some samples, most likely originating from car spills and asphalt. An extensive study of the environmental importance of asphalt cores is currently being published by the Geological Survey of Norway.

Principal component analysis (PCA) was used to obtain an overview of the chemical data. In this study, the first, second and third principal components (PCs) described only 56% of the total variation in pollutant patterns of the 68 urban runoff samples. Each chemical constituent had to be present above the detection limit in at least 75% of the samples in order to be included in the PCA. The elements Ag, B, and Be were thus excluded. Variables were scaled to unit variance prior to the analysis. Fig. 5 shows the loading plot with PCB_7, PAH_{16}, metals, and TOC for the three PCA components, revealing distinguished groups with covariating constituents. Pb, Zn, Sn, and Zr appear to correlate well, which may be an indication of paint residues from buildings in the older parts of the city, corresponding to similar findings in urban soil in inner parts of Norwegian cities (unpublished data). Vehicle pollution might



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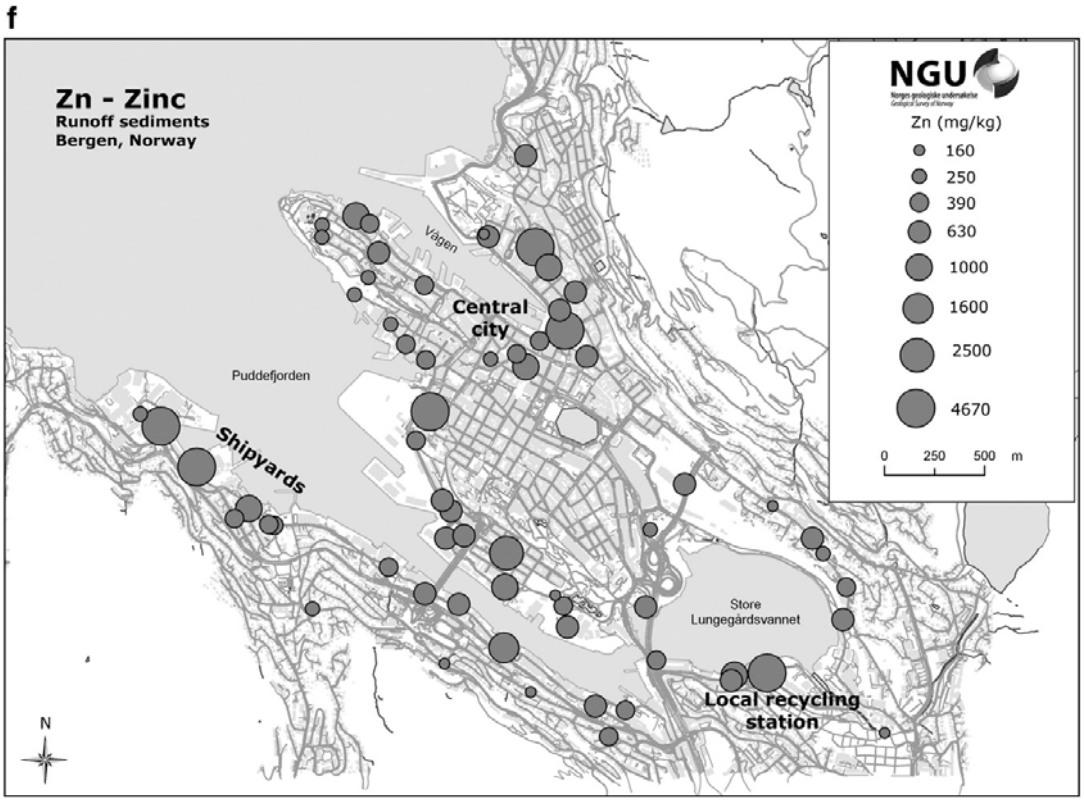
be a source for Pb and Zn via leaded gas and tire wear (Mielke et al., 1999). A correlation is indicated to occur in a group consisting of TOC, Hg, PCB_7, and PAH_{16}. A strong sorption between PCBs and PAHs to soil organic matter has been described by Krauss et al. (2000), and the relationship between organic matter and Hg has been described in other papers by Sanei and Goodarzi (2006).

Table 4 gives a summary of the grain size distribution analysis, showing particles with a diameter between 0.4 and 2000 μm. The table presents the results for the 10 percentile, median (50 percentile) and 90 percentile in 21 samples of urban runoff sediments from stormwater traps. Grain size distribution for the coarsest, the median and the finest range of the original 21 samples are given in Fig. 6, presented with a logarithmic scale on the abscissa axis representing the grain size diameter (μm). Table 4 indicates a wide variety of grain size distributions between the samples, ranging from mostly clay and silt, to samples with a main fraction of coarse sand. The median grain size ranges from 23–646 μm, with diameters 250–300 μm being the most frequent.

Stormwater quality in Norway generally focuses on dissolved compounds in the water phase. This survey however describes the concentrations of important environmental components in the sediments of stormwater traps, originating from urban soils and the anthropogenic activities in the impervious urban area. With a few exceptions, all heavy metal results are within the concentration range found in studies of urban soils from Norway (Ottesen et al., 1995; Ottesen and Volden, 1999; Ottesen et al., 2000a; Ottesen and Langedal, 2001; Langedal and Ottesen, 2001; Jartun et al., 2003; Andersson et al., 2004). In Table 5, the median values and the concentration range for selected pollutants in urban soils and stormwater sediment traps are compared. The median concentrations are higher in the trap sediments, except for Hg and Pb. The median values are slightly elevated compared to what is thought to represent the regional baseline for selected inorganic components in this area (Ottesen et al., 2000b). Cumulative frequency distributions are shown for the 9 environmentally most important elements As, Cd, Cr, Cu, Hg, Ni, Pb, Sn, and Zn (Ottesen and Langedal, 2001) in Fig. 7, with a logarithmic scale on the abscissa axis (concentration). The curves illustrate a relatively narrow range of concentrations for each element, although high concentrations occur in a small percentage of samples for each element indicating the presence of local, active sources.

4. Discussion

Urban runoff sediments trapped in small stormwater traps may originate from a wide variety of sources, such as the



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weathering of buildings, renovation, demolition, city fires, and traffic. The study of the chemical properties of these sediments will mostly reflect the ongoing, active contamination within the limited catchment area for each stormwater sediment trap, which makes this a suitable method for locating the actual pollution sources. Results from our study in Bergen show that the contamination of selected pollutants such as PCBs, PAHs, Pb, and Zn may be high in several parts of an urban area, and that particle bound pollutants are easily dispersed via stormwater from impervious surfaces to traps. Urban runoff sediments seem to be contaminated by the same means as urban soil, previously described by Mielke et al. (1999), Ottesen and Langedal (2001), and Mielke et al. (2004). The site-specific sources of PCBs in the urban environment of Bergen are especially important to reveal, because of severe contamination of the inner harbor sediments reported in a proposed remediation plan for Bergen harbor (Kryvi et al., 2005). The specific sources responsible for the marine contamination in Bergen have so far not been revealed, although the Geological Survey of Norway has conducted some studies of PCBs in building materials (paint, plaster, and concrete) where about 33% of the buildings exhibited PCB-contaminated facades (Andersson et al., 2004). However, only a few of the buildings studied by Andersson et al. (2004) were located in the immediate area surrounding the Bergen harbor. The stormwater sediment traps that were sampled in our study were all situated close to the harbor.

Due to old pipeline systems we cannot be completely certain whether a given stormwater sediment trap drains directly to the marine sediments or via the sewage system to a stormwater treatment plant. Given the annual amount of rain in Bergen, generating large volumes of stormwater, the sewage system is in any case overflowed about 30% of the time in some areas. Sustainable stormwater management, such as increased infiltration, bioretention and use of detention basins, is more or less absent in the impervious urban landscape, creating large volumes of stormwater. Rainfall patterns in Bergen as showed in Table 1 indicate that the precipitation falls more or less evenly throughout the year with a slight increase in late summer and in the fall. The average temperature in the Bergen area is above freezing point the whole year, which means that the precipitation seldom falls as snow. With large urban impervious areas surrounding the harbor, the direct runoff to the harbor may be considerable. Materials accumulated on top of impervious surfaces are easily eroded during heavy rainfall and may be scoured off, transported in suspension, and delivered to an inlet structure such as a small stormwater sediment trap and subsequently to a point of discharge. Sampling and chemical analyses of urban runoff sediments in our study have been proven to be

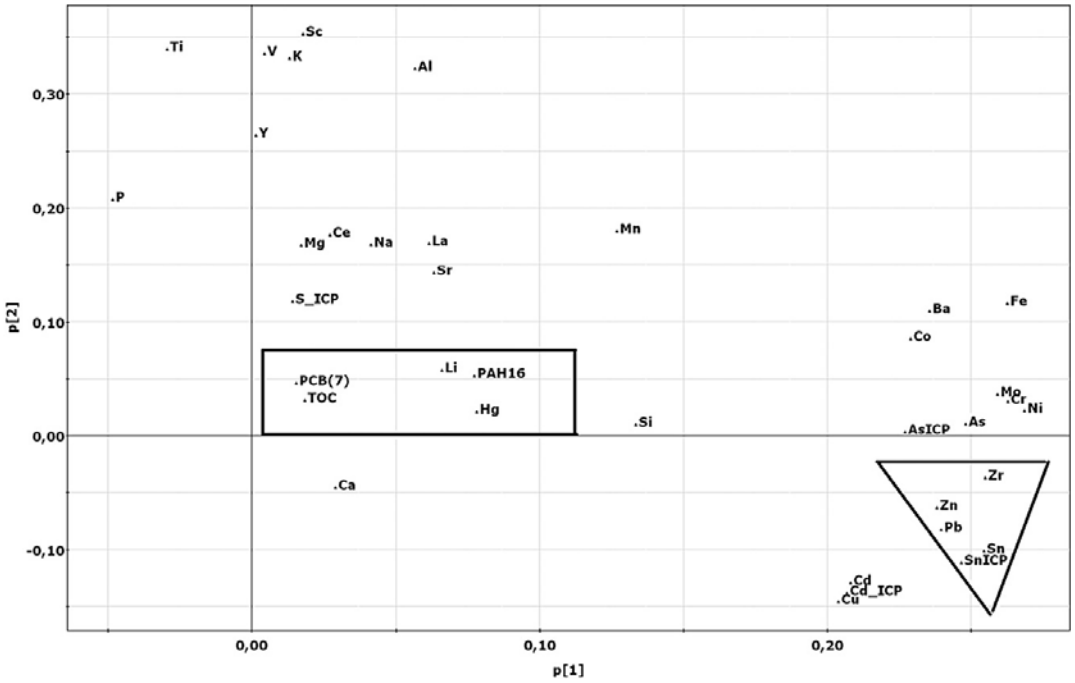

Fig. 5 – Loading plot showing the PCA analysis of the investigated components (except B, Be and Ag) and their internal correlation in 68 samples of urban runoff sediments trapped in small stormwater sediment traps in Bergen, Norway.

essential approaches in studying the presence and dispersion mechanisms of active land-based pollution sources close to the harbor. The sampling methods presented in this study are easy to use, and will provide local environmental authorities in other cities with a simple tool to reveal possible local pollution sources and investigate the dispersion of pollutants by stormwater over impervious surfaces. The results of the Bergen study indicate that important pollutants, such as Pb, Zn, PAHs and PCBs are being actively transported from site-specific sources within the urban environment towards the marine environment by stormwater in several areas of the city. Some of the pollutants may be held back in stormwater sediment traps for a period of time, but the study has disclosed ongoing contamination because each stormwater sediment trap is emptied about once a year. However, in a city such as Bergen, with over 200 storm events each year, there is a high risk that a large amount of urban runoff sediment is being discharged directly into the harbor in overflow because of a significant percentage of the sediment is within the range of suspended transport. This means that sediment contributions from the urban areas are very likely to increase the concentration of the same pollutants in the marine sediments just outside the urban area. The consequences will have to be that remediation strategies of the marine sediments (dredging,

Table 4 – Summary of particle diameter (d, μm) in 21 selected samples of urban runoff sediments

Sample ID	d at 10% vol.	d at median vol.	d at 90% vol.	Sample ID	d at 10% vol.	d at median vol.	d at 90% vol.
1	14.83	438.6	1604	40	16.54	511.7	1453
6	9.140	550.0	1712	45	29.30	599.3	1583
8	26.66	586.2	1435	47	4.917	275.9	1157
12	12.89	164.0	1303	51	8.448	256.3	918.8
13	18.75	171.0	1129	53	2.074	23.18	139.4
17	3.540	48.70	614.6	56	8.045	341.5	1569
21	3.119	23.92	160.4	61	13.17	337.5	1321
22	9.161	285.5	1333	65	5.315	99.52	1376
23	32.50	231.7	1297	67	1.956	19.77	215.7
37	22.95	646.7	1608	68	5.051	69.70	857.1
38	6.132	226.3	1295				

Particle diameter in μm at 10%, 50% (median) and 90% of sample volume.

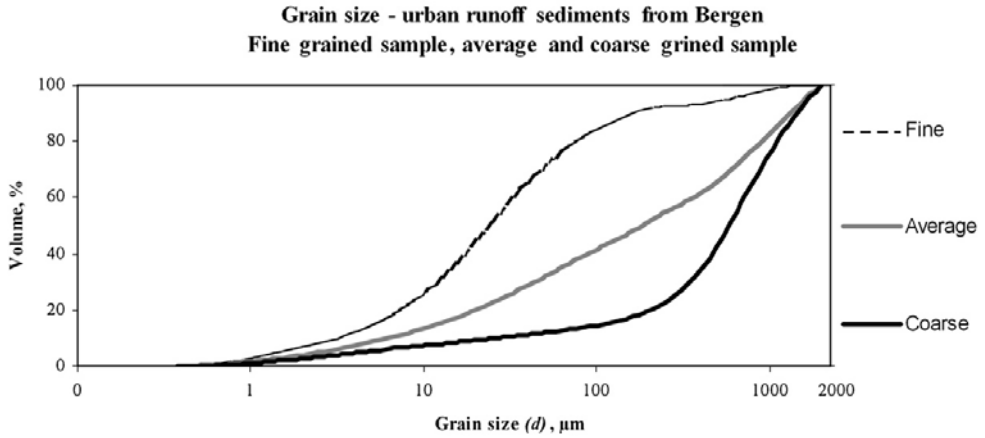

Fig. 6 – Grain size distribution for 3 selected samples of urban runoff sediments from Bergen, Norway. The 3 lines represent a fine-grained, median and coarse-grained sample. The abscissa axis represents a logarithmic expression of grain diameter (*d*) in μm, and the ordinate axis represents cumulative volume of sample for a particular diameter.

cover) in such areas only will have a short-time effect because of the more or less constant supply of pollutants. If the sources of contamination can be revealed by the present method, the harbor and city authorities will have a much better foundation of making the proper strategic decisions for harbor remediation plans.

The results from this study show that the concentration of a given pollutant may be high in one specific stormwater trap and significantly lower in a nearby one collecting sediments from a different catchment area (see maps in Fig. 4a–f). These variations may exist because of local differences such as the age of nearby buildings, time of renovation or local waste sites, which may generate various chemical signatures. First of all, two adjacent buildings may exhibit different contamination patterns in their façade. Second, the stormwater generated from rain falling on the roof, façade or outside areas from each building may drain to different stormwater sediment traps.

Table 5 – Statistical summary of selected chemical components in samples of urban runoff sediments trapped in stormwater sediment traps (results from Table 3) and urban soils from Bergen, Norway (Ottesen and Volden, 1999; Andersson et al., 2004)

Compound	Median mg/kg		Range mg/kg	
	Urban runoff sediments	Urban soils	Catch basin sediments	Urban soils
As	3.7	3.2	<1–56	0.5–18
Cd	0.4	0.3	0.02–11	0.04–1.5
Cr	25	21	11–135	6–215
Cu	97	29	16–6000	15–2850
Hg	0.06	0.2	<0.01–2.81	0.02–1.9
Ni	24	13	7–309	3–310
Pb	61	81	9–675	3–886
Zn	403	128	51–4670	41–998
PCB_7	0.031	0.014	<0.0004–0.704	<0.001–28

Sn and PAH_{16} were not determined in the urban soil studies.

Previous studies by Garnaud et al. (1999) showed a marked enrichment of metal pollutants from rain samples to samples of stormwater from the catchment outlet, demonstrating the importance of local pollution sources to the stormwater quality. Given the concentration range for each pollutant and their spatial variation (Table 3, Fig. 4a–f), it is evident that the impervious urban environment plays a significant role in the dispersion mechanisms of important environmental components such as PCBs, PAHs, Pb, Zn, and Cd. However, the exact sources of input in each area need further and more detailed study. Current investigations, based on the results from our study of stormwater sediment traps in Bergen, are being carried out to reveal the actual pollution sources in a few important catchments. Further work is also needed to get a detailed understanding of the actual quantities of different pollutants that are being discharged directly to the harbor. The fine sediment particles, at least up to the fine sand fraction of about 250 μm, may easily be transported in suspension. Studies carried out by Deletic et al. (2000) indicate that sediments up to this size are likely to be transported throughout both the catchment area and the drainage systems without being deposited. Table 4 and Fig. 6 indicate that a large amount of the sediments from urban runoff in Bergen is remarkably fine-grained, and significant amounts of the sediments have a particle diameter suited for transport in suspension to a final recipient. Generally, high particle bound concentrations of pollutants are associated with smaller particles (Xanthopoulos and Hahn, 1990) due to large surface-to-volume relationship and the good adsorption properties of especially clay minerals (Krumgalz et al., 1992). The diameter of runoff sediments in stormwater traps varies depending on the main sources, such as highway surfaces. Median particle diameters of 600–1000 μm have been reported by Ellis and Harrop (1984), Grottker (1990), and a summary table in Deletic et al. (2000). These particles may be eroded from the sediment bed in the bottom of the trap during heavy storms and transported farther into the sewer system to a point of discharge.

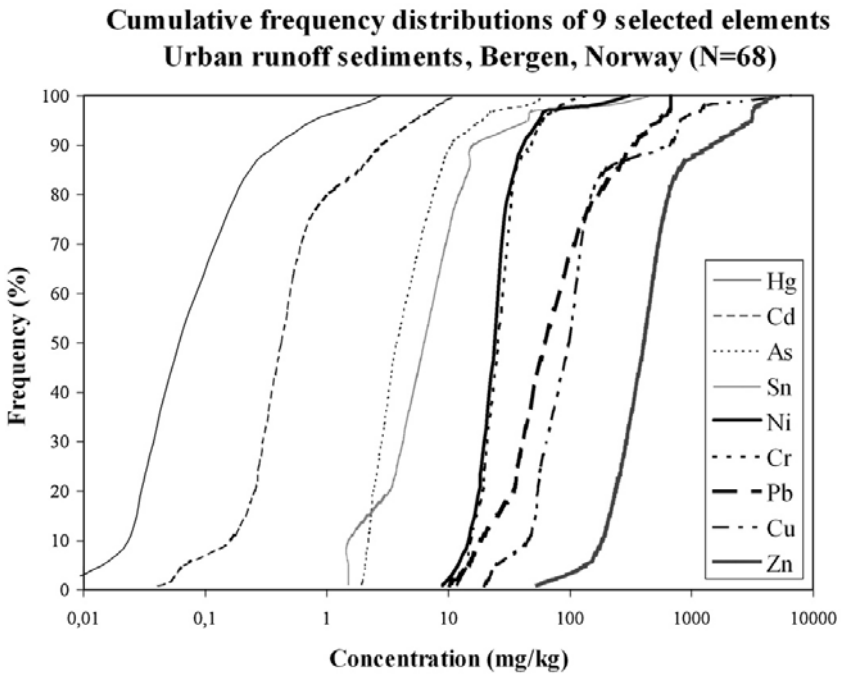

Fig. 7 – Cumulative frequency distributions of 9 selected elements (Hg, Cd, As, Sn, Ni, Cr, Pb, Cu, and Zn) in 68 samples of urban runoff sediments trapped in small stormwater sediment traps in Bergen, Norway.

The organic content (TOC) of the sediment samples is relatively high (see Table 3), which will promote the binding of different pollutants such as PCBs, PAHs, and Pb (Koh et al., 2006). A combined sewer, containing both stormwater and household sewage, will have a much higher content of organic matter than an urban stormwater system alone (Chebbo and Gromaire, 2004). Studies of urban stormwater quality have mostly been focusing on combined sewer systems, and in most cases the concentrations of pollutants have not been determined in the drainage sediments that only originate from restricted geographical catchments at the urban surface.

Pollutants in surface materials such as urban soil or residues from renovation may be re-mobilized by erosion processes such as urban runoff during a heavy storm event. The sediment mobilization from impervious surfaces into drainage systems has been computer modeled in several previous studies (Pratt et al., 1987; Deletic et al., 2000; Lindholm, 2004). Several of these computer models are exclusively site-specific, and the chemical data behind the dispersion models are often insufficient to be used extensively in real life. The most important contribution to understanding the contaminated urban environment and its importance to the possible contamination of nearby water bodies is more empirical data. Suggestions have been made to use recommended or default concentrations of pollutants to calculate the environmental load from urban drainage systems (Lindholm, 2004). It may, however, seem that too few empirical studies have been carried out to really understand the environmental impact of the urban runoff. Our study clearly shows that particle bound pollutants accumulated on top of impervious surfaces are easily picked up by stormwater and transported through the urban environment, and that the environmental load represented by different pollutants varies to a great extent from one area to another. The significance of this dispersion to the environmental condition of the marine sediments in Bergen will be further investigated. In addition, a detailed investigation on specific contamination sources within each catchment, shown by elevated concentrations of pollutants in stormwater sediment traps, will be performed based on the geochemical maps shown in Fig. 4a–f.

5. Conclusions

Detailed studies of recently deposited sediments from urban stormwater traps showed that there are sources within the impervious urban area supplying the stormwater system with particle bound pollutants such as PCBs, PAHs, and heavy metals. The stormwater sediment traps were evenly distributed in the interface between an urban area and the marine environment to get an impression of the possible dispersion of pollutants to the harbor sediments. Analysis of grain size distribution showed that a large percentage of the stormwater sediments are suitable for suspended transport. An old combined sewage system combined with large amounts of rain cause overflows and direct discharges to the marine

environment to happen at least 30% of the time. The approach explained in this paper presents the opportunity to reveal upstream pollution sources in addition to an overview of the dispersion mechanisms to a more vulnerable environment.

Acknowledgments

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Paper II

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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.

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. 23 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.

Keywords: PCB, paint, urban, building

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 –80s (Breivik et al., 2002a ; Breivik et al., 2002b). 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; Wania et al., 1998; Halsall et al., 1999; 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; Breivik et al., 2002b; AMAP, 2002; AMAP, 2003; Breivik et al., 2004) 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 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.

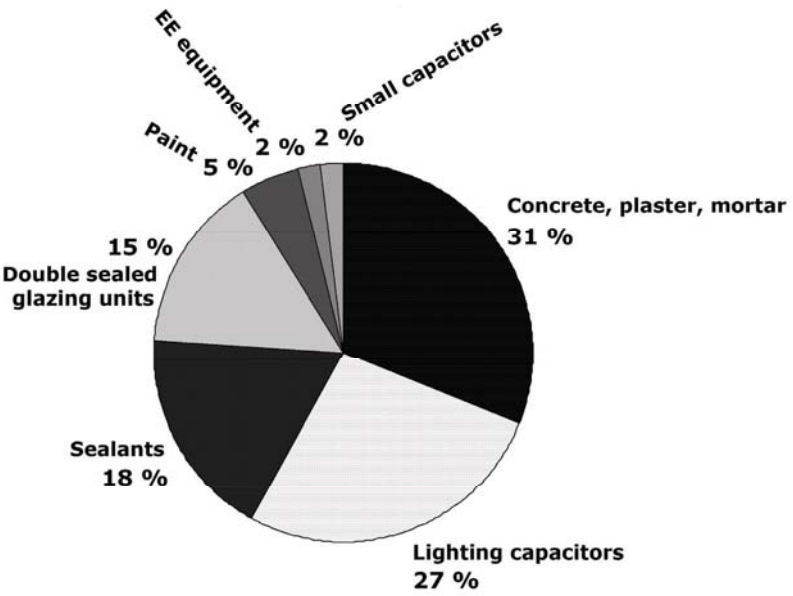

Fig. 1. Remaining polychlorinated biphenyls (PCBs) in products from Norway, shown by percentage of 155 tonnes. Numbers are from BNL (2005) and Norwegian Pollution Control Authority

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#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 tetrabuthylammonium hydrogen sulphate (TBA) with sodium sulphite in excess. Finally the organic phase was cleaned using concentrated sulphuric acid. The quantification of 7 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 x 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.

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).

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

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

[1] 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).

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 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 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.

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

…table 2 continued

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
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

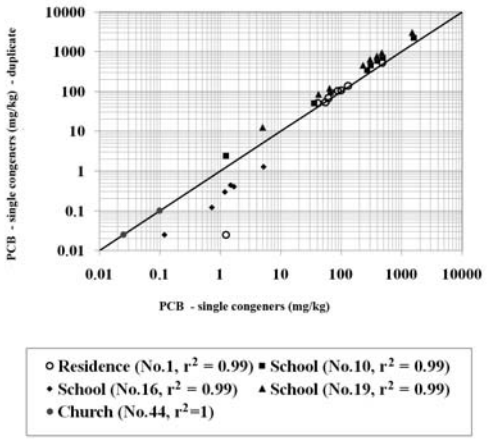

Fig. 2. Results from analysis of lab duplicates expressing the concentrations of the single congeners of PCB_7 (PCB no. 28, 52, 101, 118, 138, 153, 180) in samples of paint (N=5).

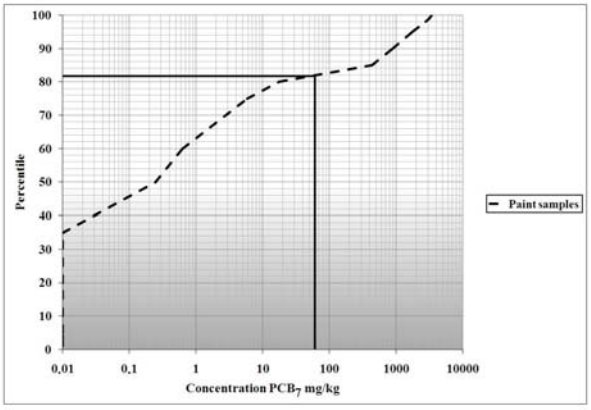

Fig. 3. Cumulative frequency distribution of PCB_7-concentration in 68 samples of paint from buildings and structures within an urban environment. Note the logarithmic scale on the abscissa. Bold line indicates the concentration of 50 mg/kg PCB_7, representing the lower limit of hazardous waste in Norway.

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.

Table 3
Concentrations of PCB_7 and individual congeners in 12 core samples from the Puddefjordsbroen bridge case study (samples no. P01-P12), including 4 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

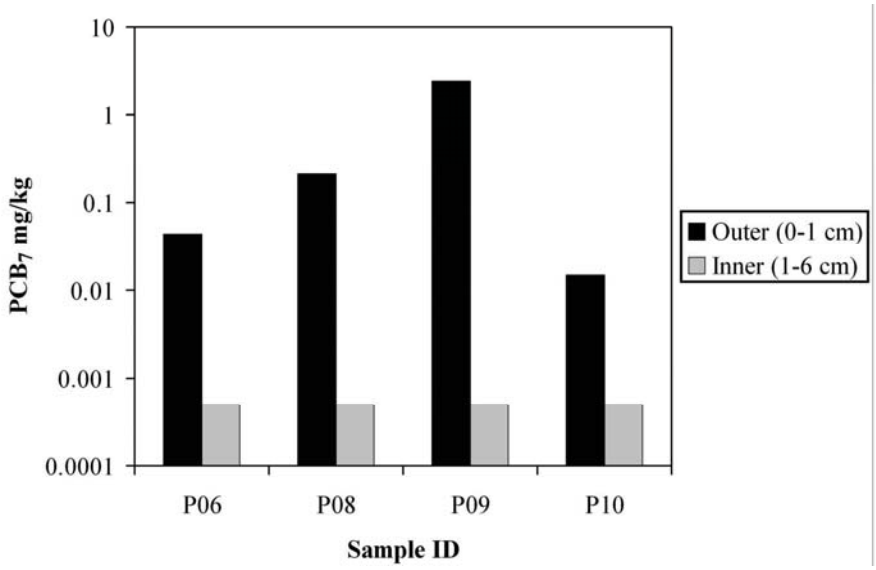

Fig. 4. Variation of surface (0-1 cm) and inner core (1-6 cm) measurements of PCBs in four samples of concrete from a bridge previously coated with a PCB containing paint.

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 (70^{th} 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 dataset. 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.

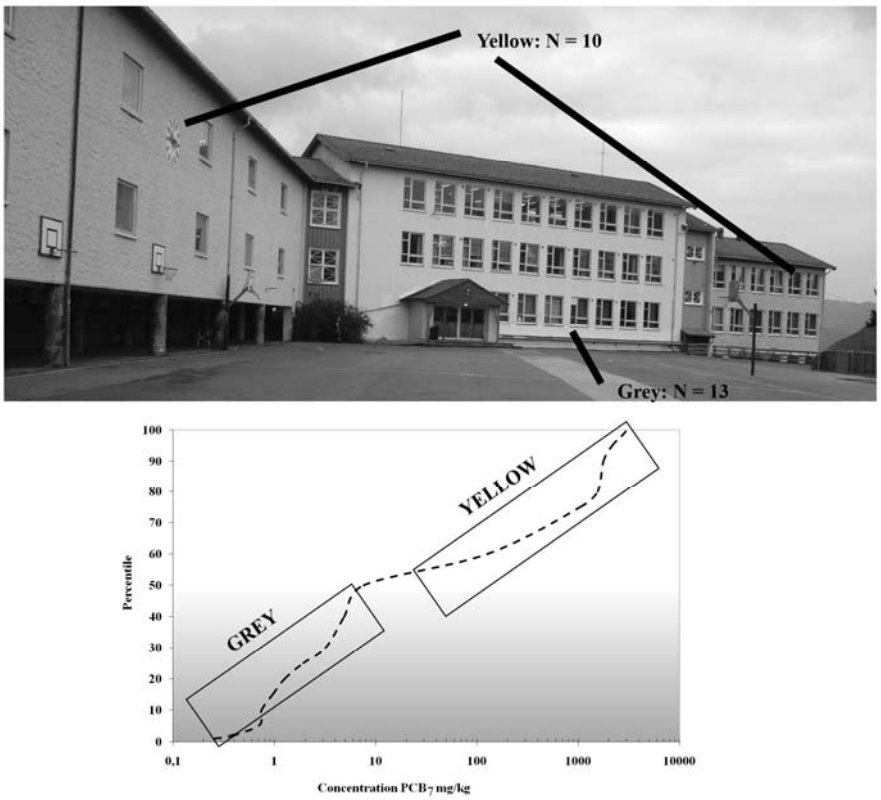

Fig. 5. PCBs in 23 samples of paint from a single school building façade. Two different types of paint show different ranges of PCB_7-concentration given in the cumulative frequency distribution. Note the logarithmic scale on the abscissa.

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; Bennet, 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 1970´s to the beginning of the 1980´s (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 the present 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. 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). Figure 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).

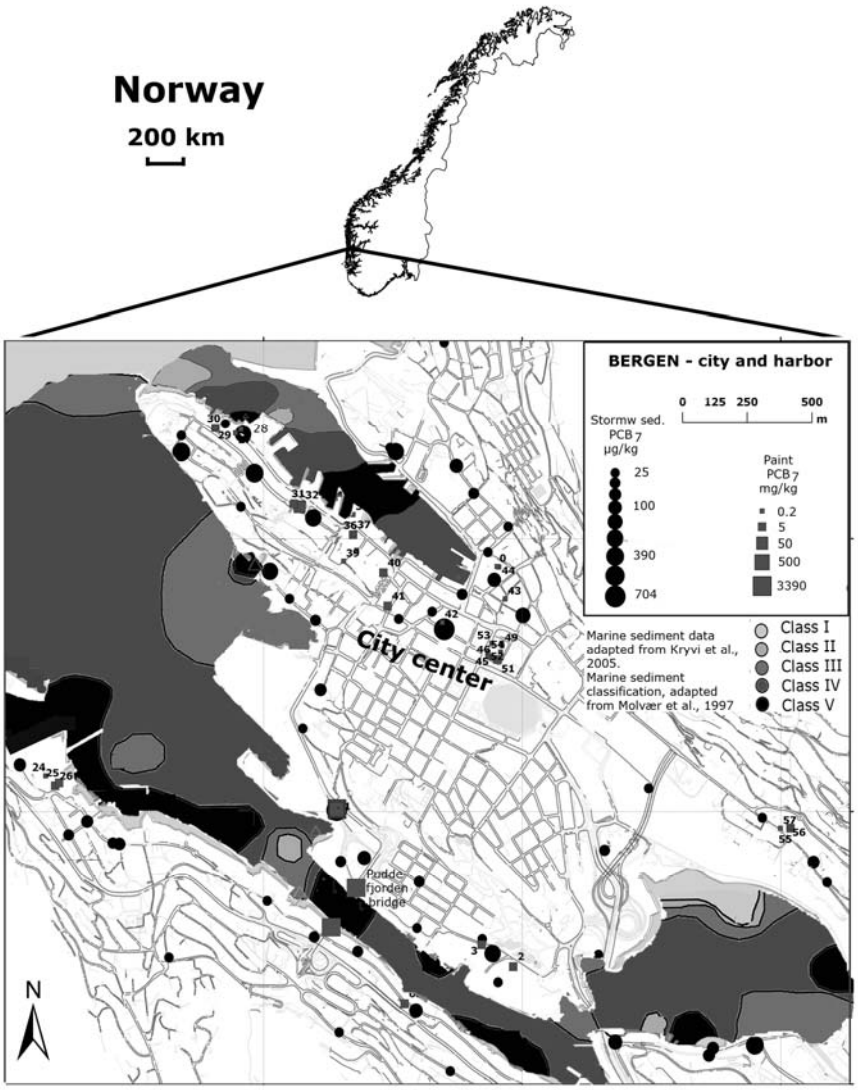

Fig. 6. Map of the study area showing the environmental condition of marine sediments based on the concentration of PCB_7 (Molvær et al., 1997; Kryvi et al., 2005), the locations of stormwater sediments studied by Jartun et al. (2008), and locations of PCB in paint (present study). The location of the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

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 liters 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 a 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
- 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, sealants) before a particular building is up for renovation or demolition. This appears

especially important for buildings erected or renovated between 1950 and 1970.

Acknowledgments

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Paper III

Jartun, M., Ottesen, R.T., Volden, T. and Lundkvist, Q. Local sources of polychlorinated biphenyls (PCBs) in Russian and Norwegian settlements on Spitsbergen Island, Norway. Submitted to Journal of Toxicology and Environmental Health, May 4th 2008.

Local sources of polychlorinated biphenyls (PCBs) in Russian and Norwegian settlements on Spitsbergen Island, Norway

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Abstract

Samples of surface soil, flaking paint, concrete, transformer oils, and small capacitors were collected from the three largest coal-mining settlements on Spitsbergen; Barentsburg (Russian), Pyramiden (Russian), and Longyearbyen (Norwegian), to study the role of potential local sources of PCB in the arctic areas (78° N). Median concentrations of PCB_7 in soil from Barentsburg and Pyramiden were 0.268 and 0.172 mg/kg respectively with a maximum concentration of 28.7 mg/kg. High concentrations found in paint (3520 mg/kg) and small capacitors (114 000 mg/kg) indicate that these two are the main sources of local PCB contamination. Only traces of PCB were found in the Longyearbyen samples compared to the results from the other two settlements. Large amounts of building refuse, electrical waste, and scrap metals constitute major pollution sources in Barentsburg and Pyramiden. Weathering and general decay will facilitate the mobilization of PCB from these sources to the local soil, which consequently is highly available for fluvial and eolian transport to the more vulnerable marine environment.

Keywords: PCB, Arctic, sources, dispersion

1 Introduction

Levels of polychlorinated biphenyls (PCBs) in various sample media in Arctic areas have so far mostly been accredited to long-range transport, including atmospheric transport (LRAT), ocean currents, trans-polar ice movements, or migrating animals (Macdonald et al., 2000; Skotvold and Savinov, 2003; AMAP, 2004; Kallenborn et al., 2007; Carroll et al., 2008). LRAT is described as a temperature-driven air-surface exchange controlled by a global fractionation process (Wania and Mackay, 1995; Wania and Mackay, 1996;

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Ockenden et al., 2003). Subsequent condensation and possible accumulation in Arctic areas is by far the most approved explanation for the high levels of PCBs found in samples of biota such as liver, blood, and brain of wildlife animals on Svalbard, including polar bears (Lie et al., 2003; Braathen et al., 2004; Dietz et al., 2004; Verreault et al., 2005; Sonne et al., 2007), seals (Wolkers et al., 2004), and birds (Henriksen et al., 1998; Knudsen et al., 2007; Murvoll et al., 2007; Verreault et al., 2007). Hop et al. (2001) used macro-benthos near urban settlements in Svalbard to study the contribution of local input of PCBs to the marine environment. Later studies of marine sediments have indicated elevated concentrations of PCB outside Barentsburg and Pyramiden (Evenset et al., 2006). The PCB-concentrations increased between 1997 and 2005 indicating an active source of contamination onshore.

PCBs have been added to a wide range of applications, including hydraulic oils, electrical transformers and capacitors, double-glazed windows, masonry coatings, sealants, and paint (Sundahl et al., 1999; Poland et al., 2001; Andersson et al., 2004; Herrick et al., 2004; Shin and Kim, 2006). The addition of PCBs to hydraulic oils as a fire retardant agent was especially essential for the coal mining industry during the decades preceding the national PCB ban in the 1980s. Based on the knowledge of PCB utilization in such a wide variety of applications, a study of possible local sources, and their significance to the local environment, was initiated by the Governor of Svalbard, Norwegian Pollution Control Authority, and Geological Survey of Norway within the three largest coal-mining settlements on Spitsbergen Island.

2 Methods

2.1 Sampling

Svalbard is a Norwegian archipelago in the Arctic Ocean consisting of several islands situated from 74 to 81 degrees North and 10 to 35 degrees East. Three of the islands are inhabited, including Spitsbergen where our study was carried out. A total of 133 samples of surface soil, paint, concrete, capacitors, and transformer oils were collected from Barentsburg, Pyramiden, and Longyearbyen, the three largest settlements on Svalbard. Surface soils (0-2 cm) were sampled into Rilsan ® bags using a small, paint-free spade. Samples of flaking paint were collected from random buildings using a filling knife. From some of the buildings, a drill core sample of the concrete was collected. Small capacitors and transformer oils were collected from random buildings in collaboration with the Russians and Norwegians living in Barentsburg and Longyearbyen, respectively. The Governor of Svalbard approved and supervised the sampling in the abandoned

settlement of Pyramiden. All samples were sent to our collaborative laboratory Alcontrol AB for PCB determination.

2.2 Chemical analysis

The extraction of PCBs followed the same procedure for all sample media, and started with 10 g of homogenized material. Acetone (20 mL), n-hexane (9.0 mL) and an inner standard (PCB-53, 100 µg/mL, 1 mL) were added followed by mixing and centrifugation. A solution of 15 mL phosphoric acid (0.1 M) and NaCl (0.2 M) was then added to the acetone/hexane-phase. 2 mL tetrabuthyl-ammonium hydrogen sulphate (TBA), 2 mL of propanol and a spatula of sodium sulphite was added to 2 mL of the hexane phase in a new test tube, followed by heating to 50°C. Deionized water and subsequently concentrated sulfuric acid were added to clean the hexane phase before the extract were condensed to 1 mL at 40°C. The quantification of 7 PCB congeners (PCB_7 IUPAC-numbers 28, 52, 101, 118, 138, 153, 180) was carried out using a Gas Chromatograph with electron capture detection (GC-ECD). The method is based upon the Nordtest Technical Report No. 329 (Karstensen et al., 1997). Certified reference materials (EC-2, sediment from National Water Research Institute, Canada) and analytical blanks were analyzed continuously in accordance to ISO/IEC 17025.

3 Results

Table 1 gives the results of the quality control comparing the measurements of PCB_7 in a reference material (EC-2) obtained from Alcontrol with the certified values from NWRI. The results are acceptable for all seven congeners.

Table 1
Results of quality control for PCB_7 (N=6) in a standard reference material obtained at Alcontrol compared to certified values from NWRI (EC-2).

Congener, IUPAC no.	Alcontrol (mg/kg dw)	SD[a]	CV[b] (%)	Reference (mg/kg dw)	CV (%)
PCB 28	0.0210	0.0038	18.0	0.028	60
PCB 52	0.0366	0.0064	17.6	0.034	70
PCB 101	0.0571	0.0103	18.0	0.053	45
PCB 118	0.0272	0.0073	26.7	0.035	49
PCB 153	0.0764	0.0123	16.1	0.081	59
PCB 138	0.0888	0.0158	17.8	0.089	43
PCB 180	0.0494	0.0079	16.0	0.048	49

[a]SD – Standard deviation
[b]CV – Coefficient of variation

Analytical results of our study of local, active sources of PCB on Svalbard are summarized in Table 2. PCBs were found in high concentrations in samples of surface soil, paint, concrete, and small capacitors. Low concentrations were detected in a limited number of samples of transformer oils and undefined plastic materials. The median values of PCB_7 found in surface soil within the two Russian settlements of Barentsburg and Pyramiden are 0.268 and 0.172 mg/kg, respectively, whereas the median value in Longyearbyen was below the detection limit of 0.004 mg/kg. Compared to a study of PCB in building materials and adjacent surface soil from Bergen, Norway (Andersson et al., 2004), the median soil concentrations of PCB_7 from the Russian settlements on Svalbard are considerably higher (0.268 and 0.172 compared to 0.150 mg/kg in Bergen). The concentrations of PCB_7 found in paint from buildings on Svalbard are also high, but at comparable levels with other studies from Norway (Andersson et al., 2004). Only traces of PCB were found in a limited number of samples of transformer oils and plastic materials, but the PCB concentrations found in small capacitors from the Russian settlements exceeds 11 %.

Table 2.
Analytical results of PCB_7 (mg/kg) in samples of soil, paint/concrete, oils, plastic materials, and small capacitors from Barentsburg, Pyramiden, and Longyearbyen. Results are shown for selected percentiles and the arithmetic mean.

mg/kg		N	Min	10 %	25 %	Median (50 %)	Ar.mean	75 %	90 %	Max
Soil	All	87	< 0.004	< 0.004	0.003	0.068	1.16	0.347	1.54	28.7
	Barentsburg	22	0.052	0.080	0.155	0.268	2.96	1.16	3.11	28.7
	Pyramiden	31	< 0.004	0.025	0.042	0.172	1.14	1.04	2.68	13.9
	Longyearbyen	30	< 0.004	< 0.004	< 0.004	< 0.004	0.010	0.007	0.019	0.131
Paint/ concrete	All	35	< 0.004	< 0.004	0.016	0.121	142	1.06	38.5	3520
	Barentsburg	16	< 0.004	0.042	0.200	0.621	229	10.8	55.6	3520
	Pyramiden	10	< 0.004	< 0.004	0.005	0.018	130	0.625	134	1290
	Longyearbyen	9	< 0.004	< 0.004	0.005	0.064	0.124	0.121	0.251	0.695
Oils	-	5	< 0.004	-	-	-	-	-	-	0.054
Plastics	-	2	< 0.004	-	-	-	-	-	-	0.021
Capacitors	-	8	< 0.3	-	-	-	-	-	-	114 000

Geographical distributions of PCB_7 in surface soil and paint within the three settlements are shown in Figures 1 and 2, respectively. For surface soil, high concentrations are distributed evenly within the sampled area of Barentsburg and Pyramiden, whereas the concentrations are consistently low in

Longyearbyen. The distribution pattern for paint is somewhat different, indicating that the surface coatings on buildings around the Heliport and in the southern part of Barentsburg are more contaminated with PCB than those in the central area around the main pier. In Pyramiden and Longyearbyen there are a few selected buildings containing PCBs, but the number of samples here are too low to provide a significant inventory of PCB-containing paint in these two settlements.

On the congener level, the PCB_7-profiles in soil samples from Barentsburg and Pyramiden exhibited a mid-chlorinated pattern mainly consisting of penta- and hexa CB. The same pattern is observed in the paint samples from these two locations. In Longyearbyen, the observed congener profiles in samples with detectable levels of PCB indicated a high-chlorinated pattern, represented by high relative concentrations of hexa- and hepta-CB. Small capacitors with high concentrations of PCB_7 exhibited a different congener composition, only represented by low-chlorinated homologues such as PCB 28.

4 Discussion

The environmental effects of persistent organic pollutants (POPs) in the Arctic are serious. High concentrations of PCBs, among other compounds, have been found in a wide range of animals and the concentrations increase in a process of biomagnification through the trophic levels (AMAP, 2004). Long-range transport is one of the screening criteria under the POPs protocol and the Stockholm Convention, whereas the role of local sources may have been underestimated in many of the discussions concerning the levels of pollutants in Arctic areas. As for PCB, this group of compounds has been added to an abundance of applications worldwide, and our study clearly shows that products containing high concentrations of PCB have been used to a large extent on Svalbard.

PCB concentrations in soils from areas heavily affected by anthropogenic activities are probably higher than in more pristine areas of Spitsbergen Island. The three largest settlements on the island constitute only a minor part of the whole Svalbard area. However, the concentrations of PCBs that we have discovered in surface soil and applications in these areas are a source of concern because of the potential of dispersion to larger areas. Surface soil collected in this study is available for erosion by creeks, rivers, rainfall, and snowmelt, facilitating a particle-bound transport to the marine environment. During early snowmelt, runoff may occur outside regular river channels. Scarce vegetation and permafrost may in turn reduce infiltration to the ground consequently making previously unavailable surface materials exposed to erosion (Bogen and Bønsnes, 2003). The

river system outside Pyramiden flushed through the abandoned settlement in 2006, transporting some of the surface materials directly to the sea. The dispersion of pollutants from the settlements facilitated by fluvial transport may be an environmental challenge for years to come. Dispersion mechanisms through eolian transport of sand and dust on dry, windy summer days may also originate from the settlements.

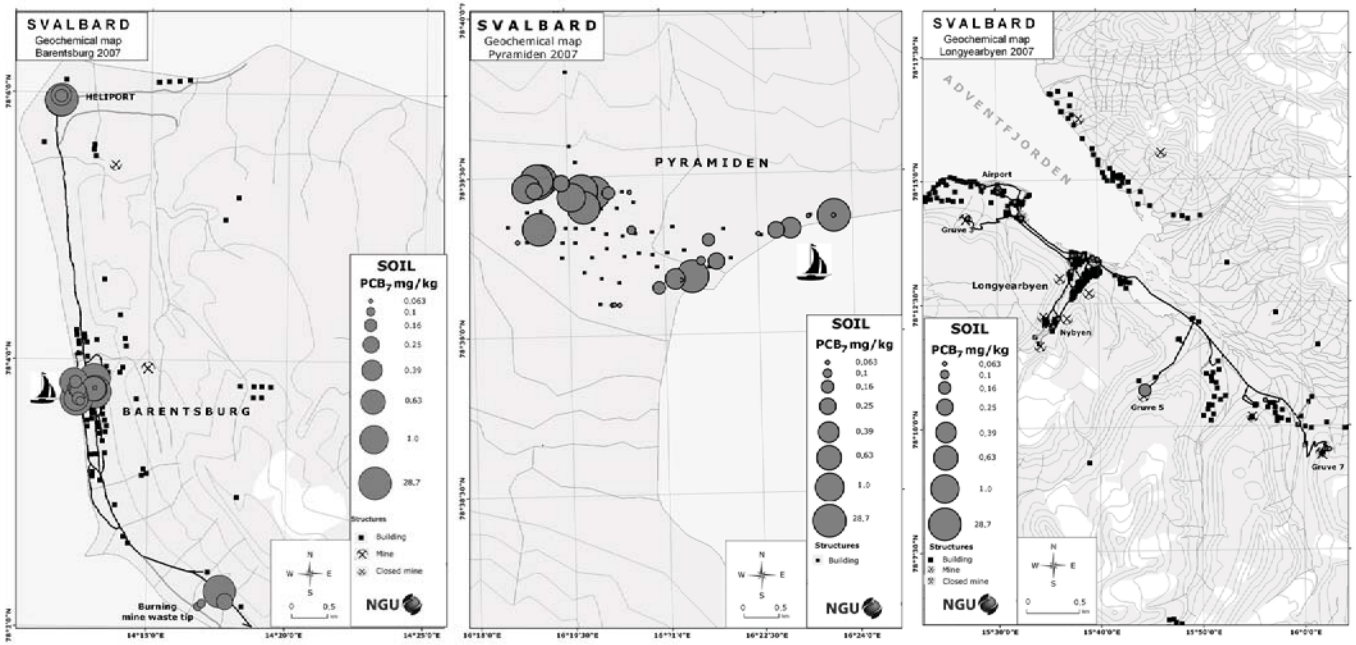

Figure 1. Geographical distribution showing the concentrations of PCB7(mg/kg) in samples of surface soil collected from (left to right) Barentsburg (N=22), Pyramiden (N=31), and Longyearbyen (N=30).

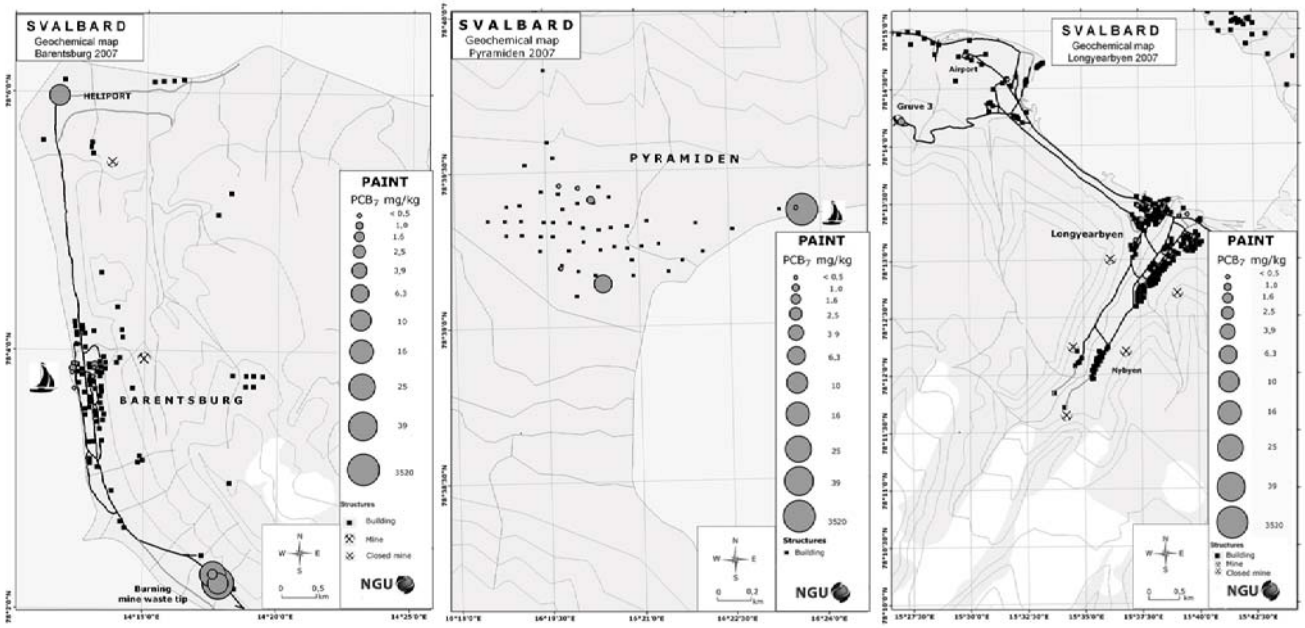

Figure 2. Geographical distribution showing the concentrations of PCB7(mg/kg) in samples of paint collected from (left to right) Barentsburg (N=16), Pyramiden (N=10), and Longyearbyen (N=9).

A large amount of waste such as building materials, electrical installations, barrels, and scrap metal is located in open terrain in and around the settlements of Barentsburg and Pyramiden. Weathering and decay are starting to mobilize the PCB added to e.g. paint and capacitors, first contaminating the adjacent soil and sediments, subsequently the marine environment. The typical small capacitor is a silver colored cylinder (~6 cm^2 x 10 cm), and each may contain as much as 50-60 g PCB. Hundreds of these were lying around as waste close to the shore in Pyramiden. Small capacitors manufactured before 1980 containing PCBs are now prohibited in Norway, including the Norwegian settlements on Svalbard, as of January 1st 2008 (Norwegian Legislation, 2004). No such regulation has been effectuated in the Russian settlements yet. Flaking paint collected from random buildings may in addition be an important source of PCBs because of the availability for further transport to the environment.

Whether or not the PCB found in surface soil and decaying products within these settlements is available for uptake by local or migrating animals such as seagulls will be an important scope of further studies. Given that PCBs are semi-volatile compounds it would also be interesting to study the soil-air flux from the heavily contaminated areas of Barentsburg and Pyramiden. The local contamination of PCBs in these areas is high, and the distance to the vulnerable arctic environment and food chains short. Some studies have indicated that PCBs may form de novo in thermal processes (Schoonenboom et al., 1995; Ishikawa et al., 2007). The coal power station in Longyearbyen and in Barentsburg, and also other combustion facilities should be subject to a thorough study of possible PCB emissions. The establishment of a ratio of long-range PCB contribution vs. local sources has not been the scope of our work so far. Both scenarios are likely to occur simultaneously. However, if the levels of PCBs in various sample media on Svalbard are to decrease, one have to start with the highly contaminated waste lying in open terrain throughout areas affected by anthropogenic activity.

5 Conclusion

Local contamination sources for PCB in the Russian and Norwegian coal-mining settlements on Spitsbergen Island have been detected. Surface soils within the Russian settlements are highly contaminated with PCB. Samples of PCB-containing exterior building paints were found in all the settlements. In the Russian settlements PCB was also detected in concrete/plaster and small capacitors. Immediate abatement of the contamination risk is needed.

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