



report

IVL Swedish Environmental Research Institute

WFD Priority substances in sediments from Stockholm and the Svealand coastal region

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Summary

The occurrence of the 32 water framework directive (WFD) priority substances has been determined in surface sediments from Stockholm and the adjoining Baltic Sea coast. In addition, six other metals, PCBs, DDTs, tetrabromobisphenol A and chlordanes were determined. Three different environments were investigated: urban area, suburban lakes, and coastal region, in total 34 stations. Two composite samples of sewage sludge were also analysed, in order to elucidate whether wastewaters have a role in the release of these substances.

The following substances were detected in most sediment samples: 4-nonylphenol, 4-tert-octylphenol, PAH, di(2-ethylhexyl)phthalate, C₁₀₋₁₃ chloroalkanes, pentachlorophenol, hexachlorocyclohexanes, chlorfenvinphos, polybrominated diphenyl ethers, tributyltin, As, Cd, Co, Cr, Cu, Hg, Ni, Mn, Pb, Zn, PCBs, DDTs and chlordanes. Chlorobenzenes were only detected sporadically. Simazine and α -endosulfan were detected with GC-ECD in several coastal sediments, but these results have not been verified with GC-MS.

These substances were not detected in any sediment sample:
Alachlor, atrazine, diuron, isoproturon, chlorpyrifos, trifluralin, TBBPA, hexachlorobutadiene, benzene, 1,2-dichloroethane, dichloromethane and chloroform.

Disregarding the most volatile substances and those known to degrade rapidly in sediments, there is excellent agreement between which substances that were found in sediments and what could be expected on the basis of known uses and emissions, regulation etc. Chemicals that are present in consumer products, i.e. DEHP, alkylphenols, PBDE, C₁₀₋₁₃ chloroalkanes, Cu and Zn, are enriched in central Stockholm and the lakes relative to the coast. Also Cd, Hg, Pb, PAH, PCBs, TBT, Σ DDT follow this pattern. Of the pesticides, mainly those known to undergo atmospheric long-range transport were found.

This study demonstrates that a number of chemicals are highly elevated in central Stockholm and the surrounding lakes. However, an influence of Stockholm on the coastal region is clearly apparent only at the nearest station (östra Askrikefjärden).

Although there was a good match between which substances that were found in sediments and in sludge, the spatial trends in the sediments suggest that these substances are also released by other sources than STPs. DEHP and the alkylphenols are unique in the sense that concentrations are generally higher in the lakes than in central Stockholm. It is proposed that a major factor contributing to this pattern is the relatively modern communities in many lake catchments, as compared to central Stockholm.

Finally, the organic substances were ranked with respect to the risk they pose in these aquatic environments. This assessment is approximate, due to methodological uncertainties. Nonetheless, risk is considerably better than concentrations as a measure of the environmental problem with these chemicals. Most 4-6 ringed PAHs and chlorfenvinphos are ranked highest, followed by nonylphenol, octylphenol, tributyltin, lighter PAH, and the non-WFD substances PCBs and DDTs. This is a generalization and results differ in some respects between the three geographical groups, and indeed between the stations.

Sammanfattning

De 32 prioriterade ämnena från EU:s vattendirektiv har analyserats i ytsediment från Stockholm och den angränsande Östersjökusten. Dessutom har 6 andra metaller, PCB, DDT, TBBPA och klordan analyserats. Tre typmiljöer har undersökts: centrala Stockholm, sjöar i Stockholms närhet, och Svealandskusten – totalt 34 stationer. Två samlingsprov på rötslam ingick också, som en markör på om de aktuella ämnena sprids via avloppsvatten.

Följande ämnen detekterades i flertalet sediment: PAH, di(2-etylhexyl)ftalat, 4-nonylfenol, 4-tert-oktylfenol, pentaklorfenol, hexaklorcyklohexan, klorfenvinfos, bromerade difenyletrar, tributyltenn, C₁₀₋₁₃ kloralkaner, As, Cd, Cr, Cu, Hg, Ni, Mn, Pb, Zn, PCB, DDT-gruppen och klordaner. Klorbensener detekterades sporadiskt. Simazin och α -endosulfan detekterades med GC-ECD i flera kustsediment, men resultaten har inte verifierats med GC-MS och bör betraktas som preliminära.

Inte i något sediment kunde följande ämnen detekteras: alaklor, atrasin, diuron, isoproturon, klorpyrifos, trifluralin, TBBPA, hexaklorbutadien, bensen, 1,2-diklorethan, diklormetan och kloroform.

Om man bortser från de mest flyktiga ämnena samt de som snabbt omvandlas i sediment, så är det en utmärkt överensstämmelse mellan vilka ämnen vi återfunnit i sediment och vad man kunde förvänta sig utifrån känd användning och kända utsläpp. DEHP, alkylfenoler, PBDE, kloralkaner, koppar och zink är alla vanligt förekommande i konsumentprodukter och uppträder generellt i betydligt högre halter i Stockholm och dess omgivning, än i kustregionen. Detta gäller även Cd, Hg, Pb, PAH, PCB, TBT och Σ DDT. Av de pesticider som undersökts så återfinns vi huvudsakligen de som sprids via långväga atmosfäriskt transport. Sammanfattningsvis visar denna studie att ett antal "farliga ämnen" sprids till miljön i Stockholms stad och dess omgivningar, men att dessa utsläpp inte generellt förefaller påverka kustmiljön utanför Askrikefjärden.

DEHP och alkylfenolerna är speciella såtillvida att de vanligen uppträder i högre halter i sjöarna än i centrala Stockholm. Vi föreslår att detta, åtminstone delvis, beror på att sjöarnas avrinningsområden karaktäriseras av yngre samhällen än centrala Stockholm.

Slutligen har de organiska ämnena rankats med avseende på vilken de risk de utgör för dessa vattenmiljöer. Denna bedömning är ungefärlig, p.g.a. olika antaganden som riskbedömningsmetodiken bygger på. Ändå anser vi att risk är betydligt bättre än koncentrationer som mått ämnens inbördes miljöfarlighet. Flertalet 4-6-ringade PAH och klorfenylfos utgör störst risk, följt av nonylfenol, oktylfenol, tributyltenn, mer volatila PAH, PCB och DDT (PCB och DDT är ej prioriterade i Vattendirektivet). Detta är en sammantagen bild av hela datamaterialet. Vissa skillnader föreligger mellan de olika typmiljöerna och framförallt mellan olika stationer.

1 Background - Water Framework Directive and the Priority Substances

As part of the European Community water framework directive (2000/60/EC), 32 priority substances (PS's) have been identified (Annex X, decision 2455/2001/EC). These substances were selected on the basis of their risk to the aquatic environment, or to human health via the aquatic environment. Eleven of these substances were classified as hazardous priority substances (HPS's): all emissions and releases of those compounds must cease within the next 20 years. A further 15 of the substances are presently being evaluated for possible classification as HPS's. The list of priority substances is presented in Table 1, together with some brief information on the substances.

The prioritization of substances followed a risk assessment methodology, where exposure was considered in an EU perspective. Some of the PS's have not been permitted in Sweden for a long period of time (see Table 1), wherefore this EU perspective results in certain of the PS's possibly being of lower relevance to Sweden. However, the fact that a certain substance is banned is no guarantee for its absence in the environment: long-range atmospheric transport, unregistered use, release from imported goods, unintentional formation and former emissions of highly persistent substances may all lead to contamination of the environment. Monitoring the environment is thus crucial. Many of these substances have only rarely been measured in the Swedish environment. There is thus an urgent need for a consistent dataset of all the PS's that may help to focus the Swedish work on the implementation of this EC directive.

The major goals of this study are:

- Identify which of the priority substances that pollute an urban region and its adjoining coastal environment.
- Assess whether pollution is generated locally or is a result of large-scale atmospheric transport.
- Assess if the coastal region is polluted due to the proximity of Stockholm.
- Assess whether certain pollutants pose a risk to the aquatic biota.

In the context of the water framework directive, these compounds could be measured in the water phase, in the sediments or in biota, e.g. fish. Each of these matrices has some advantages and some disadvantages. We have selected surface sediments because they

represent a longer time span (a few years) as compared to water samples which only reflect a momentary situation. A majority of the substances also have properties that favour their transport from water to sediments. A primary goal of this study is to identify those of the PS's that are emitted in significant amounts in the region. For this purpose, sediments are generally more suitable than fish due to the process of biological uptake and metabolism that discriminate strongly between different substances. Sediment is also an important pool of many pollutants that may be assimilated by sediment dwelling organisms and subsequently transported up through the food chain.

However, a few of these substances are not likely to partition to sediments, and some substances may undergo chemical changes in sediments. These aspects are treated explicitly in the discussion section.

In addition, we have studied the PS's in two samples of sewage sludge from major STPs in the region. The occurrence of a certain compound in sewage sludge indicates that the compound is emitted via waste waters. Such information is therefore helpful in constraining the sources of the compound. Furthermore, effluents from STP's may influence the pollutant levels in the local aquatic environment.

Table 1. The substances investigated, their major uses or emission sources, and their legal status in Sweden. The Hazardous priority substances are marked in bold. Many substances have a wide and diverse use in society as well as many possible emission sources. This table is only intended as a rough guide to these aspects. The regulation column shows whether the compound is used or banned in Sweden. The Swedish government decided in 1990 that the use of certain chemicals should successively be phased out in Sweden. These compounds are denoted "phase-out".

CAS-no	Name	Uses or emission sources	National regulation	Present in consumer products
	PAH	Incomplete combustion		
120-12-7	Anthracene	Incomplete combustion		
91-20-3	Naphthalene	Incomplete combustion		
12002-48-1	Trichlorobenzene	Industrial chemical	No intended use since 1998	
608-93-5	Pentachlorobenzene	No known uses		
118-74-1	Hexachlorobenzene	Biocide; unintended formation	No intended use since 1980	
15972-60-8	Alachlor	Pesticide	No approved use since 1978	

CAS-no	Name	Uses or emission sources	Regulation	Present in consumer products
1912-24-9	Atrazine	Pesticide	Banned in 1989	
470-90-6	Chlorfenvinphos	Pesticide	No approved use since 2001	
330-54-1	Diuron	Pesticide	Banned in 1993	
115-29-7, 959-98-8	Endosulfan	Pesticide	Banned in 1996	
608-73-1	Hexachlorocyclohexane (HCH)	Pesticide	Banned	
58-89-9	(gamma-HCH, lindane)	Pesticide	Banned in 1989	
34123-59-6	Isoproturon	Pesticide	Restricted use	
2921-88-2	Chlorpyrifos	Pesticide	Restricted use	
87-86-5	Pentachlorophenol	Pesticide	Banned in 1978	
122-34-9	Simazine	Pesticide	Banned in 1995	
1582-09-8	Trifluralin	Pesticide	Banned in 1990	
—	PBDE (## 47, 99, 100)	flame retardant	phase-out, banned from Aug 2004	Yes
25154-52-3 104-40-5	Nonylphenol 4-para-nonylphenol	Industrial chemical; forms through degradation of NP-ethoxylates	phase-out	Yes
1806-26-4 140-66-9	Octylphenol 4-tert-octylphenol	Industrial chemical; forms through degradation of OP-ethoxylates	No restriction	Yes
87-68-3	Hexachlorobutadiene	Industrial chemical; by-product from chlorinated solvent production	No restriction	
85535-84-8	C₁₀₋₁₃-chloroalkanes	Lubricant; cutting fluid	Phase-out	Yes
117-81-7	Di(2-ethylhexyl)phthalate	Plasticiser	1999: restricted in children's toys	Yes
71-43-2	Benzene	incomplete combustion; component in petroleum products	Restricted use	Yes
67-66-3	Trichloromethane (chloroform)	Solvent	Restricted use	
107-06-2	1,2-Dichloroethane	Solvent	Restricted use	

CAS-no	Name	Uses or emission sources	Regulation	Present in consumer products
75-09-2	Dichloromethane	solvent in medical industry	Banned 1993 in consumer products, 1996 for prof. use	
-----	Tributyltin	Antifoulant; preservative; stabiliser in plastics; forms through degr. of TBTO	1993: all ships under 25 m; no new use after 2003	Yes
-----	Pb	Numerous	phase-out	Yes
-----	Hg	Numerous	phase-out	
-----	Ni	Numerous		Yes
-----	Cd	Numerous	Restricted use	
Non-WFD substances that are included in this study				
-----	PCB (28, 52, 101, 118, 153, 138, 180)	Numerous	1972: ban of open use 1978: extended ban 1995: total ban	
-----	DDT (DDE, DDD)	Pesticide	1970: Restricted use 1975: total ban	
79-94-7	TBBPA	flame retardant		yes
-----	As	wood preservative	Restricted use	
-----	Co			
-----	Cr	steel, pigment etc		Yes
-----	Cu	Numerous, e.g. roofs, car brakes, water pipes		Yes
-----	Mn			
-----	Zn	Numerous		Yes

2 The study area

The study area is located in eastern Sweden and encompasses both freshwater in the Stockholm region and the brackish coastal waters of the Baltic Sea (Figure 1). Surface sediments from 34 stations were investigated: 14 in the coastal region of Svealand (Figure 3) and 20 in the Stockholm municipality (Figure 2). Furthermore, two composite samples of digested sludge were analysed from the two major STPs in the area.

Details of the stations are presented in Table 2 and Table 3. Stockholm is divided into two groups: (suburban) lakes and central Stockholm. The coastal stations are all true accumulation bottoms. This is also true for most stations in Stockholm, although it may be difficult to find completely undisturbed sediments in certain areas of central Stockholm. ^{210}Pb profiles have been determined for several Stockholm stations (Östlund et al., 1998; Sternbeck, 1998). These profiles showed that the upper ca 5 cm were mixed in most lakes. In central Stockholm, surface sediments showed much less influence of mixing.

Due to varying sediment accumulation rates (SAR), the upper 2 cm that we analysed represent slightly different time periods. In coastal region, SARs of 0.4-1.2 cm/yr correspond to ca 1-5 yrs. With the SARs in central Stockholm, the 0-2 cm layers correspond to 1-2 years. Due to the mixing of surface sediments in the lakes, the time period must be estimated from upper ca 5 cm, which translates to ages of 2.5 –5.5 years.

Hydrologic residence times in the lakes span over a range from one month to four years (Table 3). Certain lakes receive large amounts of storm water, due to proportionally large catchments and to the presence of impervious surfaces in the catchments. Forests cover a major part of the catchment of other lakes, e.g. Judarn and Kyrksjön. The influence of impervious surfaces is also important for all central Stockholm stations.

All central Stockholm stations are hydrologically connected. Freshwater runs eastward from the large lake Mälaren and, after passing a lock, mixes with brackish water at the station Strömmen (Figure 2). Hydrologic residence times are generally not known for central Stockholm, but are probably lower than in most lakes (Table 3). Sewage treatment plants are known to emit some of these substances. Effluents from the three STPs in Stockholm can directly influence only two of the Stockholm stations (Strömmen & Fjäderholmarna, Figure 2). None of the lakes receives input from any STP.

The Stockholm stations were investigated previously, as part of a larger study on the distribution of PAHs, PCBs, metals and hydrocarbons in sediments (Östlund et al., 1998; Sternbeck och Östlund, 2001). The general pattern that emerged was that stations in central Stockholm were more polluted than most of the surrounding lakes, although concentrations in central Stockholm show a decline over the last decades.

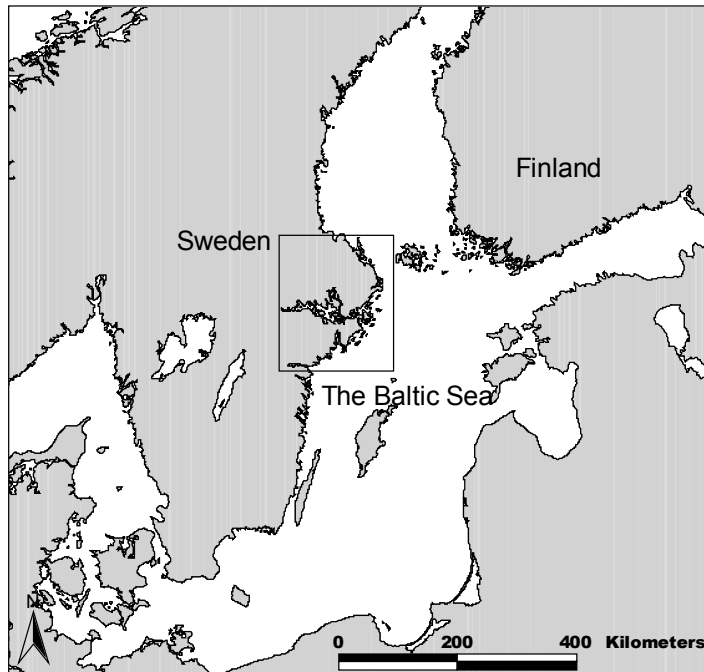


Figure 1. Regional overview. The investigated area is marked with a square.

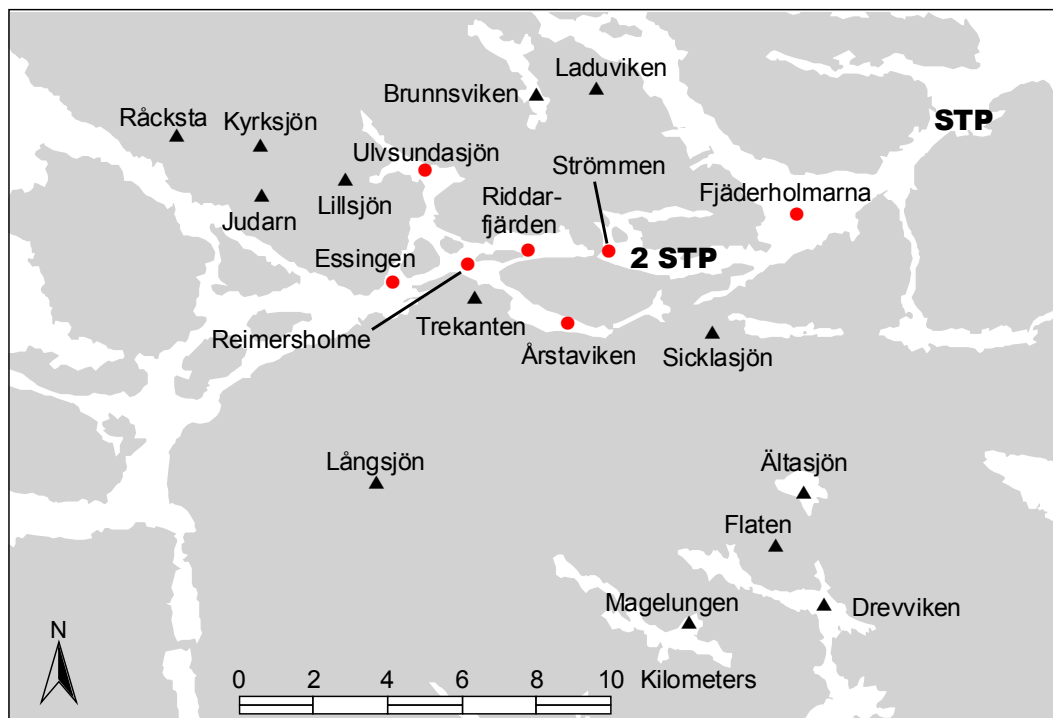


Figure 2. Stations in the Stockholm municipality. Stations in the lakes group are marked with black triangles, and stations in the central Stockholm with red circles. The outlets of three STPs are also shown.

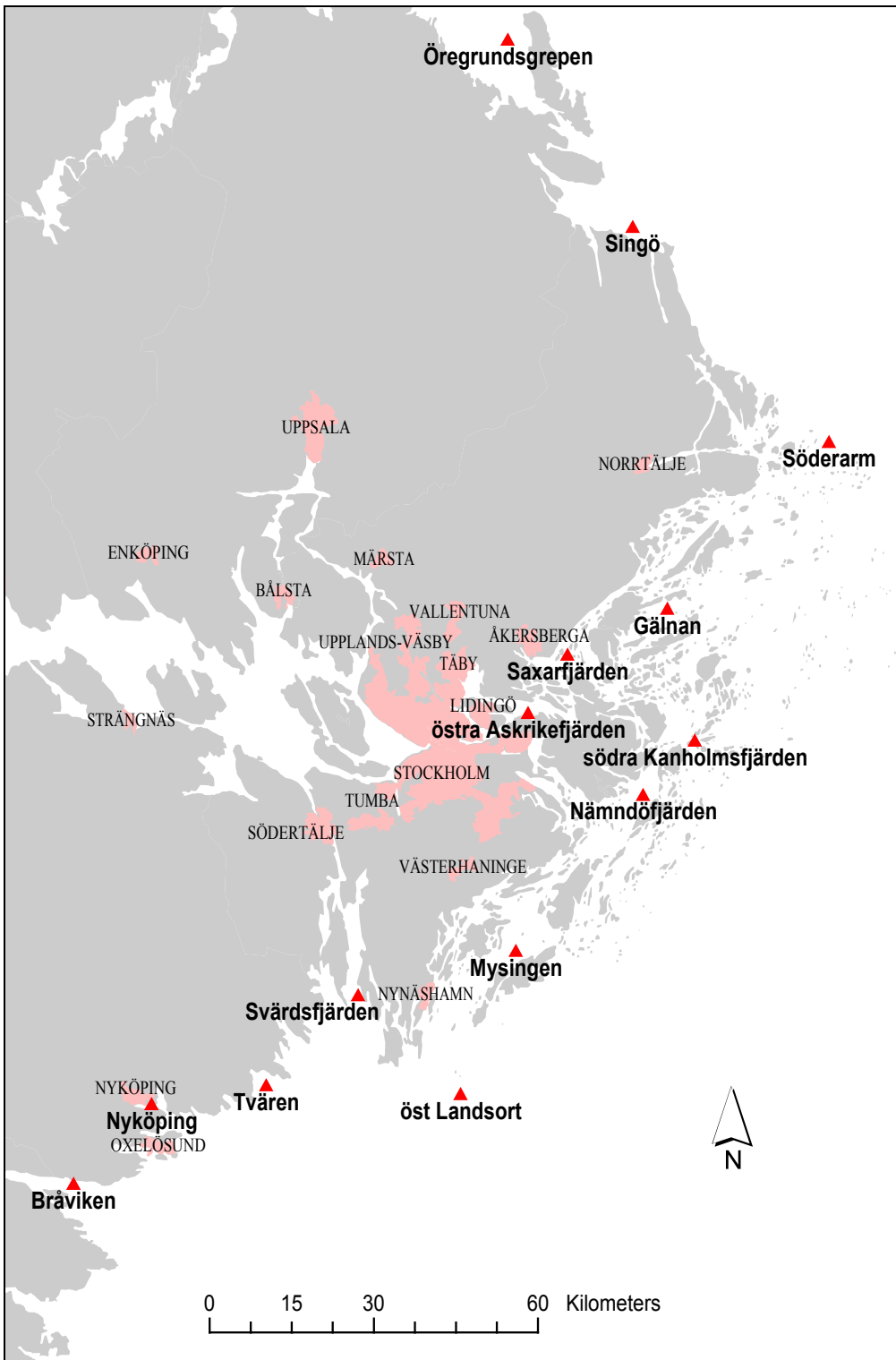


Figure 3. Stations along the Svealand coast. Larger communities are shown in red. Stockholm (figure 2) is located in the center.

Table 2. Sampled stations along the Svealand coast.

Station	sed. depth (cm)	Latitude	Longitude	sampling depth (m)
Bråviken	0-1	58.61662	16.81733	48.1
Gälnan	0-2	59.52558	18.76064	30.3
Mysingen	0-2	58.97791	18.23225	45.1
Nyköping	0-1	58.74433	17.06748	1.7
Nämndöfjärden	0-2	59.2239	18.65679	77.6
Saxarfjärden	0-2	59.45754	18.43316	48.2
Singö	0-2	60.15024	18.70259	46.1
Svärdsfjärden	0-2	58.91404	17.72776	34.3
Söderarm	0-2	59.78409	19.30911	71.2
södra Kanholmsfjärden	0-2	59.30886	18.82971	65.5
Tvären	0-1	58.77127	17.42997	75.9
Öregrundsgrepen	0-2	60.46402	18.31577	53.4
öst Landsort	0-2	58.74742	18.04221	64.3
östra Askrikefjärden	0-2	59.36507	18.29903	52.1

Table 3. Sampled stations in the Stockholm municipality (lakes and central Stockholm). Station numbers > 100 refers to lakes; < 100 to central Stockholm.

Station	Station nr	Latitude	Longitude	Sampling depth (m)	Hydrological residence time (months)	catchment divided by lake area
Brunnsviken	144	59.360217	18.046883	7	not known	6.5
Drevviken	105	59.23485	18.173767	12	11	8.5
Flaten	126	59.2496	18.152533	11	48	6.4
Judarn	135	59.3377	17.915483	2.8	11	11
Kyrksjön	139	59.349783	17.91565	2.5	7	7
Laduviken	145	59.360833	18.074967	3	3	20
Lillsjön	132	59.340883	17.955267	3	not known	10
Långsjön	111	59.26805	17.965317	3	10	8.5
Magelungen	102	59.23165	18.110133	9	5	7.8
Räcksta träsk	136	59.353017	17.8763	2	0.7	100
Sicklasjön	110	59.30115	18.125983	4.5	1.5	14
Trekanten	129	59.311817	18.014633	5	36	4.3
Ältasjön	107	59.261783	18.166467	5	21	6

Table 3. continued.

Essingen	80	59.315733	17.97635	29	not known	
Fjäderholmarna	66	59.3288	18.1681	28	not known	
Reimersholme	18	59.319433	18.0124	23	not known	
Riddarfjärden	34	59.322667	18.040883	19	8 hours- 2 weeks	0.76
Strömmen	52	59.321617	18.079017	27	not known	
Ulvsundasjön	6	59.342383	17.99385	16	not known	7.3
Årstaviken	45	59.304667	18.058033	8	not known	6.6

3 Methods

3.1 Sampling

Sediments in the Stockholm municipality were sampled with a gravity corer (Kajak) in May and June 2002. Surface sediments (0-2 cm) of at least eight cores from each station were mixed in the field. The coastal sediments were taken with a Gemini corer and sliced onboard. Surface sediments from four cores were mixed. The samples were collected and stored in precleaned and burned glass bottles. Prior to sampling, the liners were cleaned with Deconex. Samples were stored cold and in the dark until extraction.

Composite sludge samples were obtained from the digesters at the Bromma and Henriksdal sewage treatment plants. The individual samples were taken with a metal spoon three times a week during a period of two weeks. The sludge samples were stored in glass bottles and were frozen prior to analysis.

3.2 Chemical analysis

Table 4 gives a summary of the analytical methods used. Analyses were performed in the IVL laboratories, with the exception of final determination of chloroalkanes that was done by NILU, Kjeller, Norway. Chloroalkanes were determined with GC-high resolution-MS in negative ionization mode.

Table 4. Summary of analytical methods used. Unless presented in detail in this report, a reference is given where details on the analytical method are found.

Substance (group)	Analytical method	Reference
PAH	Soxhlet extraction, silica gel chromatography, HPLC w. fluorescence detection	
Endosulfan (α - & β -)	Extraction, column chromatography fractionation, GC-ECD	
PCBs Hexachlorocyclohexanes PBDE (47, 99, 100)	Soxhlet extraction, sulfuric acid treatment, aluminium oxide chromatography, GC-ECD	Östlund et al., 1998
Chlorobenzenes Hexachlorobutadiene	Soxhlet extraction, sulfuric acid treatment, aluminium oxide chromatography, GC-MS	
Alachlor, Atrazine Chlorfenvinphos Chlorpyrifos Diuron, Isoproturon Simazine, Trifluralin	GC-NPD	Lagerberg (2002)
Pentachlorophenol	Extraction, acetylation, GC-ECD	Palm et al., (2002a)
Nonylphenol Octylphenol	Extraction, acetylation, GC-MS	
C ₁₀₋₁₃ -chloroalkanes	GC-HRMS	Tomy et al., (1997)
Di(2-ethylhexyl)phthalate	Extraction, GC-ECD	Parkman & Remberger (1994)
Benzene	Extraction w. pentane, GC-FID	
Dichloromethane 1,2-Dichloroethane Trichloromethane	Extraction w. pentane, GC-ECD	
Tributyltin	Freeze drying, extraction w acetate/methanol, ethylation w NaBEt ₄ , GC-FPD	
Cd, Hg, Ni, Pb, Co, Cr, Cu, Mn, Zn in Stockholm	Freeze drying, digestion with HNO ₃ 120°C flame AAS	Östlund et al. (1998)
As in Stockholm	Ashing, hydride generation, AAS	Östlund et al. (1998)
Metals in coast	Total digestion, ICP-AES; ICP-MS	

PAH was analysed with HPLC using a fluorescence detector. Therefore, acenaphthylene was not obtained. Previous studies in these sediments have shown that this compound contributes less than 1% to $\Sigma 16$ -PAH, why we calculate that sum from the 15 PAH analysed. Detection limits for individual PAH are given together with results in the appendix.

Endosulfan eluted in a slightly polar fraction (10% MTBE in pentane) in the silica gel chromatography of the PAH-extract. It was gently treated with $H_2SO_4:H_2O$ and then fractioned on a aminopropylene column. Three fractions were eluted F1 (hexane), F2 (MTBE+hexane 1+9), and F3 (MTBE+hexane 1+3). Fractions F1 and F3 was combined and analyzed for α - and β -endosulfan with GC-ECD.

For analysis of chlorobenzenes and hexachlorobutadiene, seven deuterated PAHs were added as internal standards. The samples were extracted and cleaned up as for PCB analysis and analysed by GC-MS.

Identification and quantification of 4-nonylphenols was done on extracts for pentachlorophenol analysis by GC-MS SIR on nine peaks using one of the ions m/e 107, 121, 135, 149 and "4-nonylphenol, mixture of isomers" (Acros Organics) as a standard. 4-tert-octylphenol, being a single substance (4-(1,1,3,3,-tetramethylbutyl)-phenol), was quantified on the ion m/e 135 using a standard (Aldrich).

The analytical precision of chloroalkanes is reported to $\pm 20\%$. The precision of PBDEs, alkyphenols and DEHP is estimated to $\pm 15\%$, and pesticides to $\pm 20\%$. For the individual PCB congeners, precision was previously reported (Östlund et al., 1998) to vary between 3 and 10 %. For metals in Stockholm and the lakes, precision is reported to be $\pm 10\%$, except for Hg with a precision of $\pm 15\%$ (> 2 mg/kg dw) or $\pm 20\%$ (< 2 mg/kg dw). IVL Laboratories are accredited for analysis of metals, PAH and PCBs in sediments. The routines used for these substances are also applied for all other substances.

3.3 Statistics and graphical presentation

The three datasets (lakes, central Stockholm, coastal) commonly show pronounced overlap even when average values differ. Whether two datasets really are significantly different in a statistical sense, a test such as the t-test is commonly applied. However, an underlying assumption for such tests is that data distribution is normal or close to normal. Environmental data are frequently not normally distributed, but sometimes follow log-normal distribution. It is not to be expected that the current datasets are normally distributed because the grouping of the stations is somewhat arbitrary. In particular, the lakes differ widely in terms of anthropogenic load, catchment characteristics etc. An earlier study comprising ca 100 stations in central Stockholm and

the lakes (Östlund et al., 1998; Sternbeck and Östlund, 2001) demonstrated that concentrations of Cd, Hg, Pb, Cu, As and Zn were log-normally distributed, whereas $\Sigma 16\text{PAH}$ and $\Sigma 7\text{PCB}$ were neither normally nor log-normally distributed. Therefore, average values and standard deviations are of little informative value.

Several of the substances in this study show a very heterogeneous concentration distribution and can be described as heavy tailed and with unequal variances in the three geographical groups. A few high outliers may strongly influence the average values, the comparison of which clearly is doubtful. It is not straight-forward to compare such datasets statistically (e.g., Skovlund and Fenstad, 2001). Furthermore, the standard deviations are in certain cases larger than the average value.

Instead, we have chosen to show data on box-whisker plots (an example is shown in Figure 4). An advantage of these plots is that they give the median values and, through the other percentiles, an illustrative view of how heterogeneous the datasets are. The outer percentiles must be considered as approximate due to limited number of samples, especially for central Stockholm that only consists of seven samples. Percentiles can be calculated with several different methods, why results obtained with different softwares may differ. We have chosen the Cleveland method, which gives less emphasis on single extreme values.

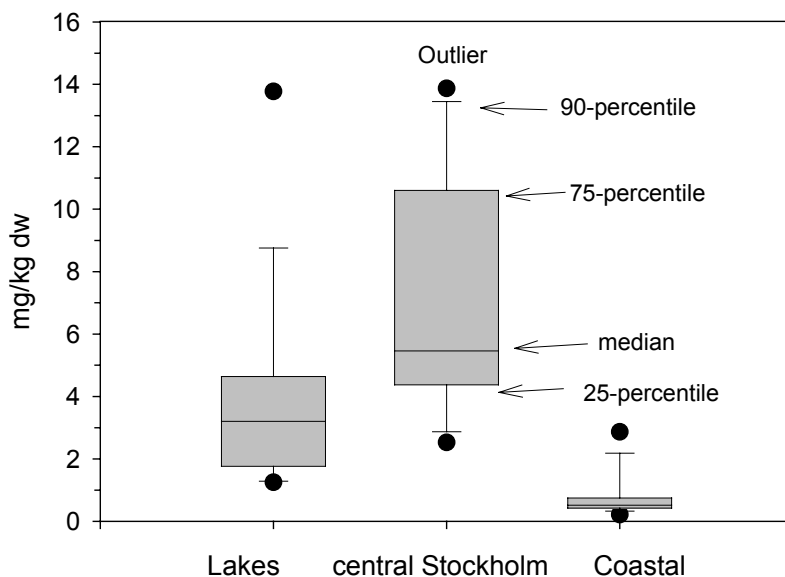


Figure 4. Illustration with explanations of the graphical data presentation method. This type of plot is used for most substances in the Results chapter.

Data are also presented on maps to provide a quick and efficient overview of the spatial trends. For this purpose, well-established “environmental quality guidevalues” would

have been useful for classifying datapoints in low, high etc. In Sweden, such values have been published by the EPA, but only for a limited number of the compounds studied (Anon., 1999). Those values were not derived from risk for effects. We have chosen not to use those values when creating the intervals for the maps because (1) they are available for only a few of the substances in this study, and (2) the maps should provide as much information as possible on the spatial trends. We have therefore chosen to divide each dataset in four (or five) intervals. The starting point is the percentile distribution of the entire dataset for each substance, but inherent delimitations in the datasets are also considered. The percentiles given below are therefore very approximate:

Blue: concentrations up to the 25-percentile;

Green: concentrations between the 25-percentile and the median;

Yellow: concentrations between the median and the 75-percentile;

Red: all values above the 75-percentile.

Values below detection limits are also informative and shall be included when calculating the percentiles. We have chosen to represent these values with a zero when calculating percentiles. For a few substances, values below detection limit constitute more than 25 % of the dataset. In those cases, the upper limit for the blue labels as well as the lower label for the green has been adjusted. For example, TBT was not detectable in 12 out of the 34 samples. Consequently, the blue label represents the 35-percentile. Certain compounds display a strongly skew distribution, with a few very high values. In those cases, a fifth interval is included that represent these values, roughly the 90-percentile. This interval is represented with a larger red label.

Finally, it is important to realise that the total concentration range differs between the substances. The shift from one label to another is therefore not of equal significance for all substances.

4 Results

All data are presented in tables in the appendix. The following subchapters provide a summary with graphical illustrations of the results. Different aspects of the data are discussed in chapter 5.

4.1 Metals

In addition to the four priority pollutant metals (Cd, Hg, Ni and Pb), the study also comprises As, Co, Cr, Cu, Mn, and Zn. The coastal samples were analysed using a stronger digestion method (chapter 3.2), wherefore those metal levels are not entirely comparable to those from Stockholm. If the coastal samples show lower concentrations than in Stockholm, this trend is true. In the opposite case, however, the difference may possibly be due to the different digestion methods. The data from Stockholm are compared with results from the 1997 study in chapter 5.3.

The concentrations of Hg, Cd, Cu, Pb, Ni, Zn, Cr and As are compared between the three geographical groups in Figure 5, Figure 6, Figure 7 and Figure 8. One coastal sample remains to be analysed, why the graphs are preliminary. Central Stockholm is clearly enriched in mercury compared to the lakes. This pattern is not obvious for As, Cd, Cu, Ni, Pb or Zn, but to some extent for Cr. It is obvious that concentrations of Hg, Cd, Cu, Pb, Ni and Zn are lower in the coastal region, though a progressive decline from Stockholm to the more remote coastal stations can be seen (Figure 9, Figure 10, Figure 11). Care must be taken when interpreting metal concentrations in the Baltic Sea, because redox processes and primary production may also influence the concentrations of certain metals, e.g. Cd (e.g., Manheim, 1961; Sternbeck et al., 2000).

Manganese and Co show no sign of anthropogenic enrichment (not shown on graphs). It is well-known that variations of Mn concentrations in sediments mainly depend on redox conditions. The median values of Co are almost identical to preindustrial values in the Baltic Sea (Anon., 1999).

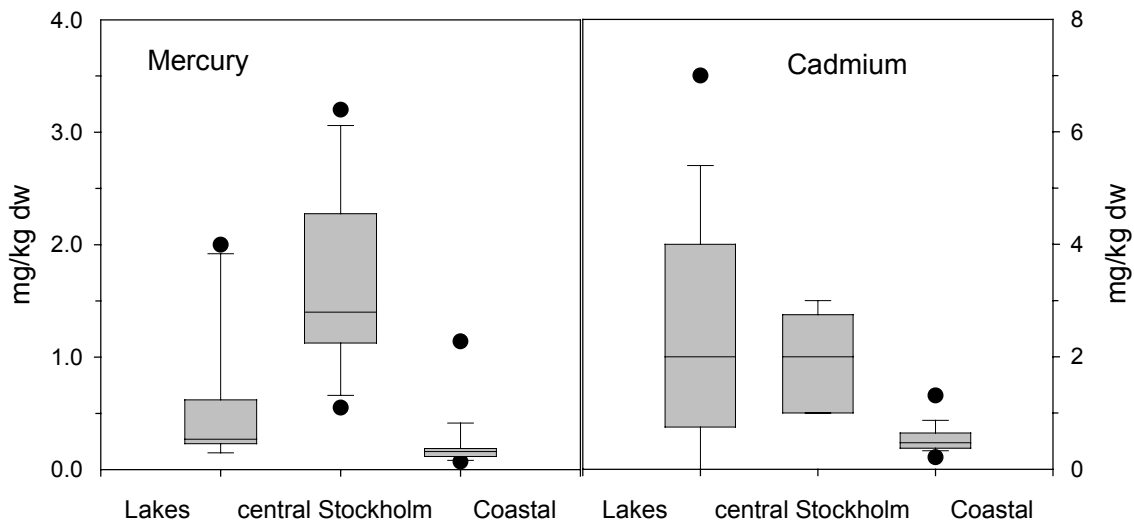


Figure 5. Concentrations of Hg and Cd in the three groups. For explanation of the plot, see chapter 3.3.

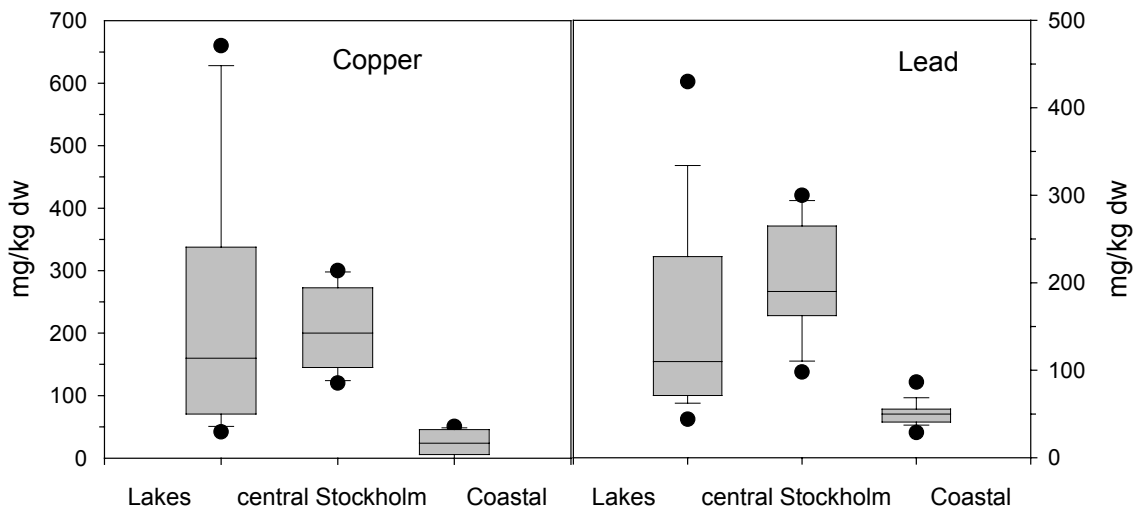


Figure 6. Concentrations of Cu and Pb in the three groups. For explanation of the plot, see chapter 3.3.

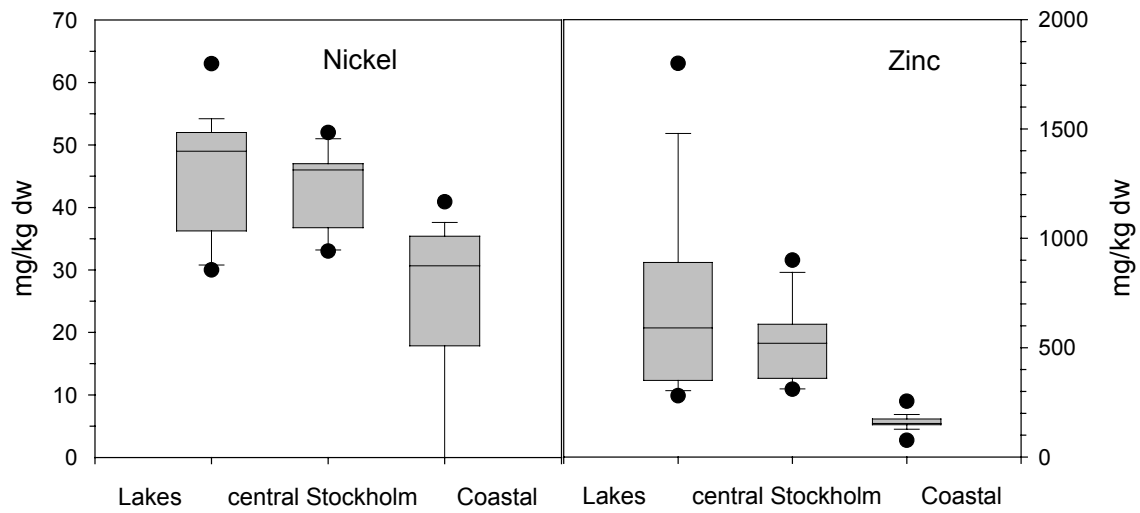


Figure 7. Concentrations of Ni and Zn in the three groups. For explanation of the plot, see chapter 3.3.

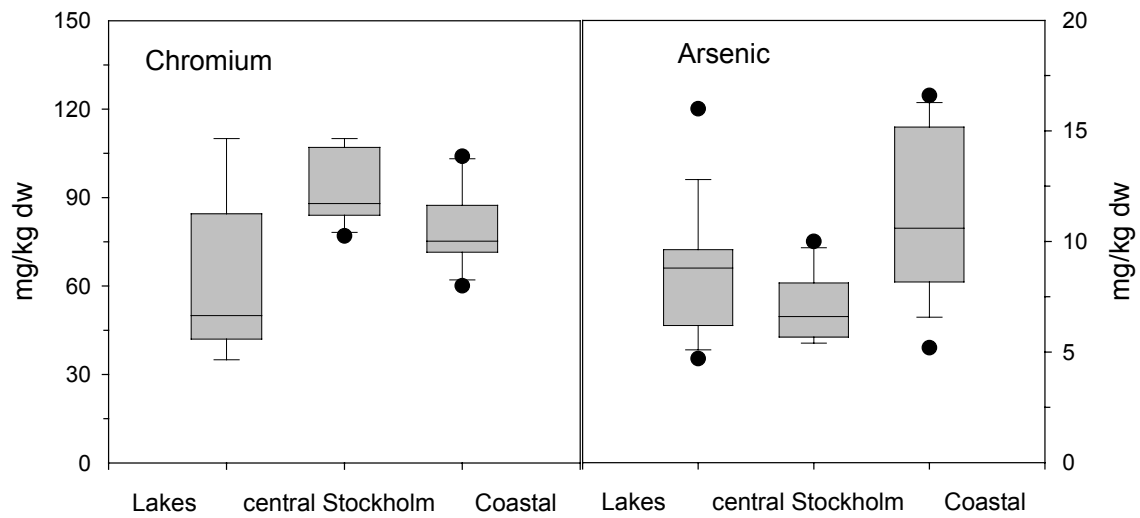


Figure 8. Concentrations of Cr and As in the three groups. For explanation of the plot, see chapter 3.3.

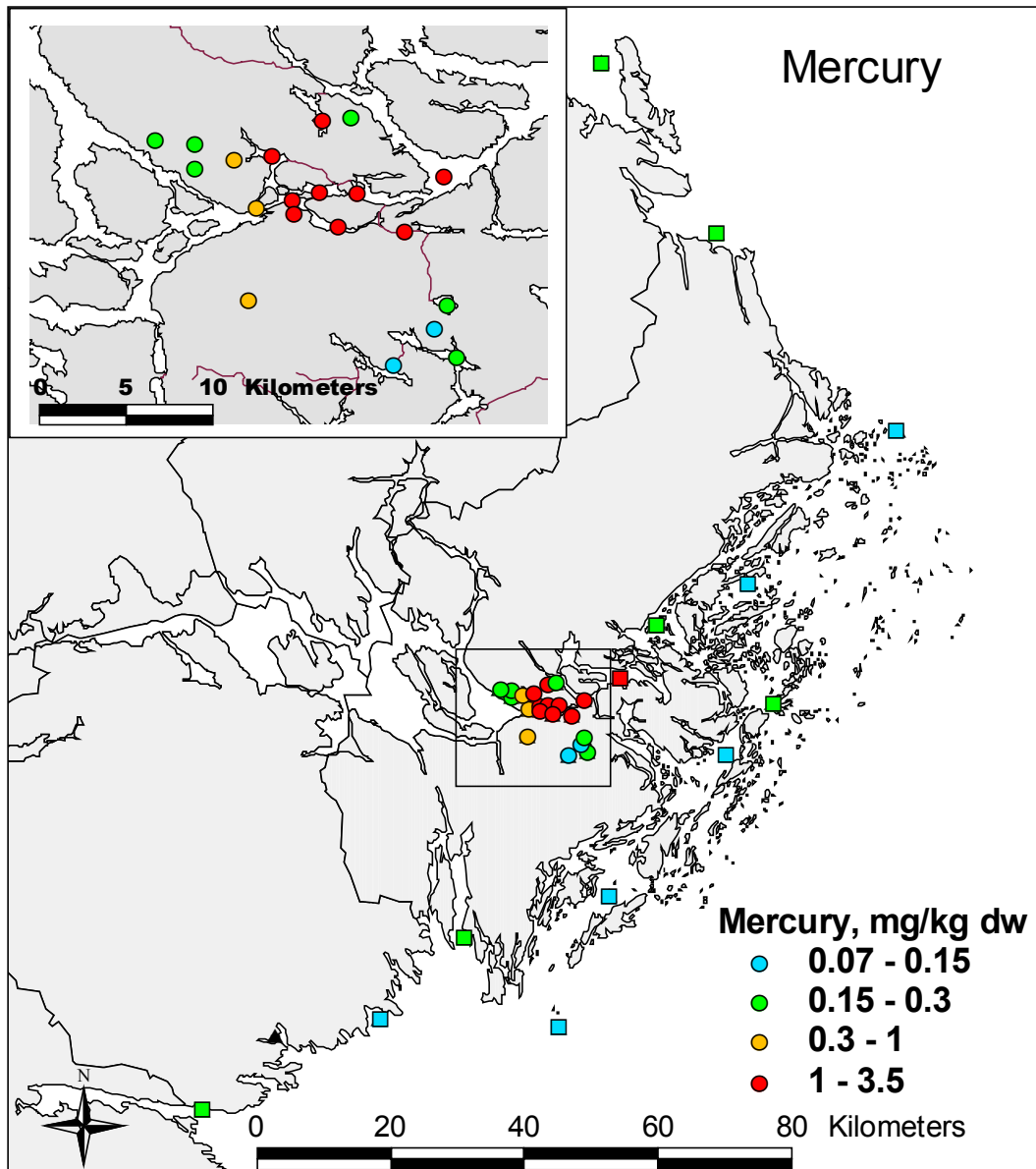


Figure 9. Mercury concentrations. A close-up of Stockholm is provided in the upper left. The coastal stations are analysed following a stronger digestion (see methods) and are therefore marked with squares. One coastal station is not analysed and is marked with a black triangle.

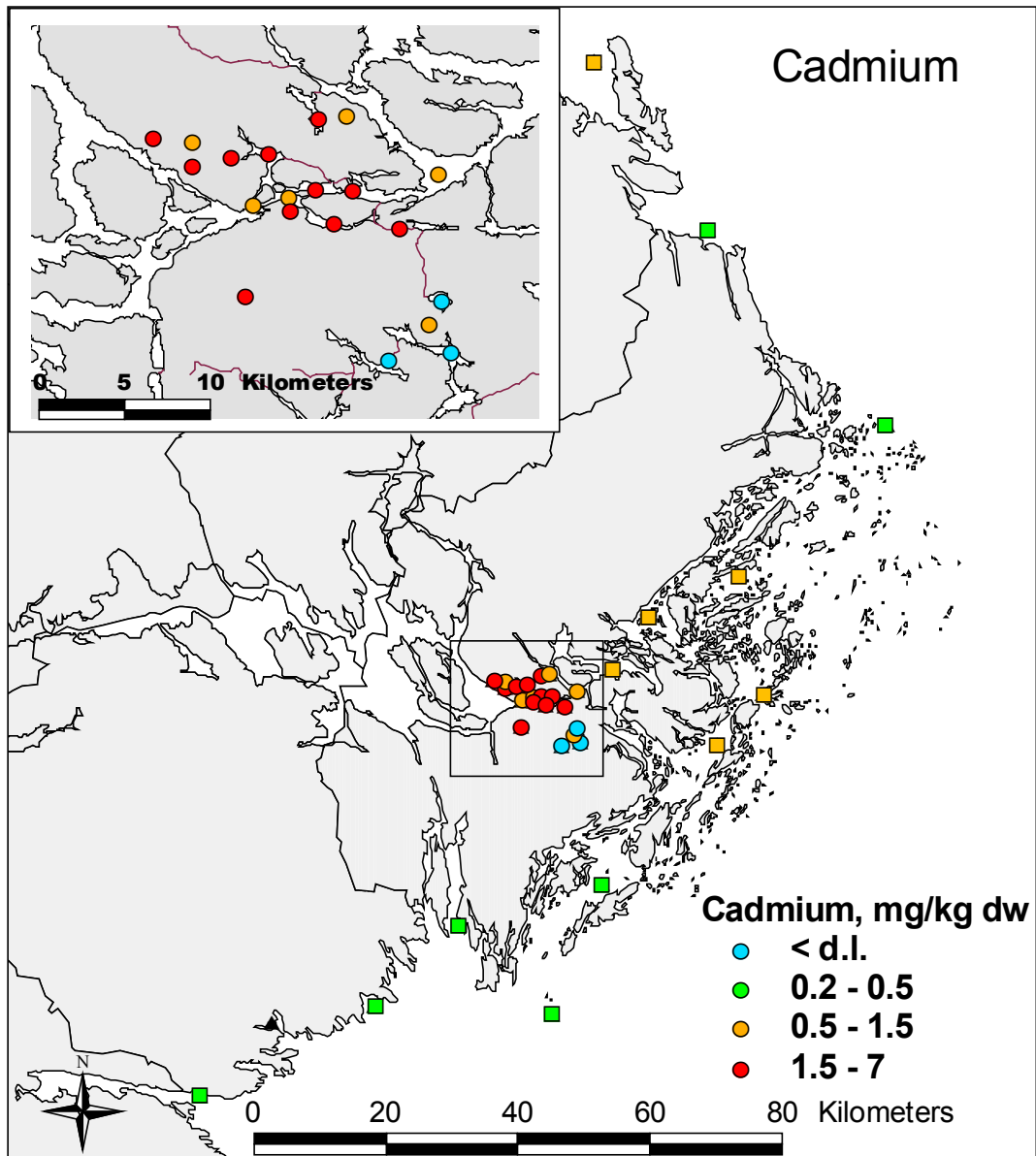


Figure 10. Cadmium concentrations. A close-up of Stockholm is provided in the upper left. The coastal stations are analysed following a stronger digestion (see methods) and are therefore marked with squares. One coastal station is not analysed and is marked with a black triangle.

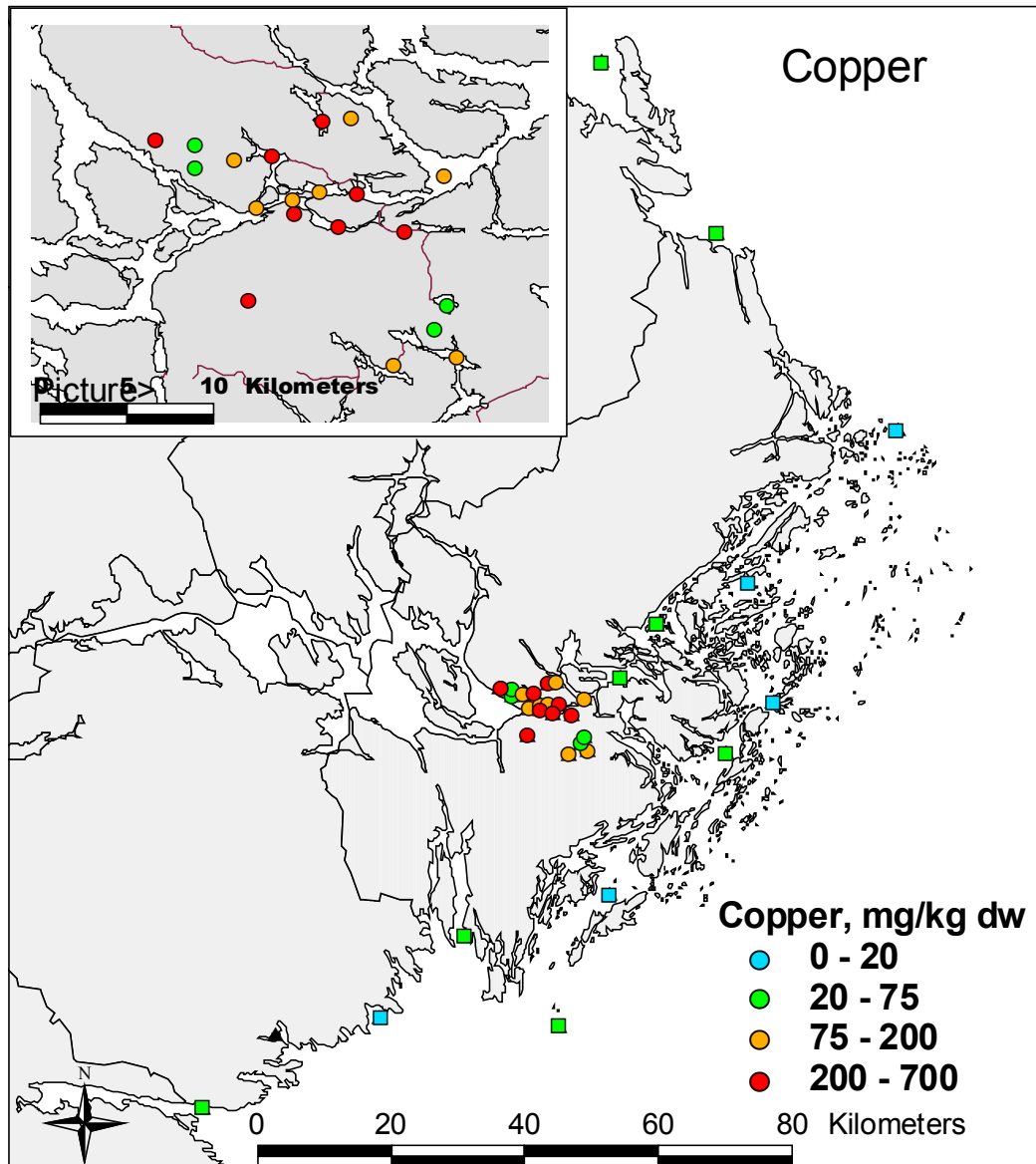


Figure 11. Copper concentrations. A close-up of Stockholm is provided in the upper left. The coastal stations are analysed following a stronger digestion (see methods) and are therefore marked with squares. One station was not analysed and is marked with a black triangle.

4.2 Polycyclic aromatic hydrocarbons (PAH)

Concentrations of $\Sigma 16$ -PAH in the three geographical groups are compared in Figure 12, and are displayed geographically in Figure 13. Concentrations of the individual PAHs are presented in the appendix. Generally, $\Sigma 16$ -PAH decreases in the following order: central Stockholm > lakes > coast. Except for naphthalene, all PAHs intercorrelate very strongly. With the analytical technique used, data is obtained for several lakes where concentrations could not be detected in the previous study (Östlund et al., 1998). The present data are compared with results from the 1997 study in chapter 5.3.

The coastal region is relatively homogenous with an average $\Sigma 16$ -PAH concentration of $0.52 \pm 0.17 \mu\text{g/g dw}$, if we omit the two anomalously high values at Östra Askrikefjärden and södra Kanholmsfjärden (Figure 13). Those stations are discussed in chapter 5.2.3. This homogeneity indicates that local influence on PAH is limited. As a reference, surface sediments from open Baltic Sea, sampled during the 1990's, were in the range $0.8\text{-}1.5 \mu\text{g/g dw}$ (Witt and Matthäus, 2001). Along the Swedish west coast, the median $\Sigma 11$ -PAH was $0.9 \mu\text{g/g dw}$ ($n=105$; Cato, 1997) as compared our median of $0.47 \mu\text{g/g dw}$. These comparisons support our conclusion that the local influence along the Svealand coast is limited.

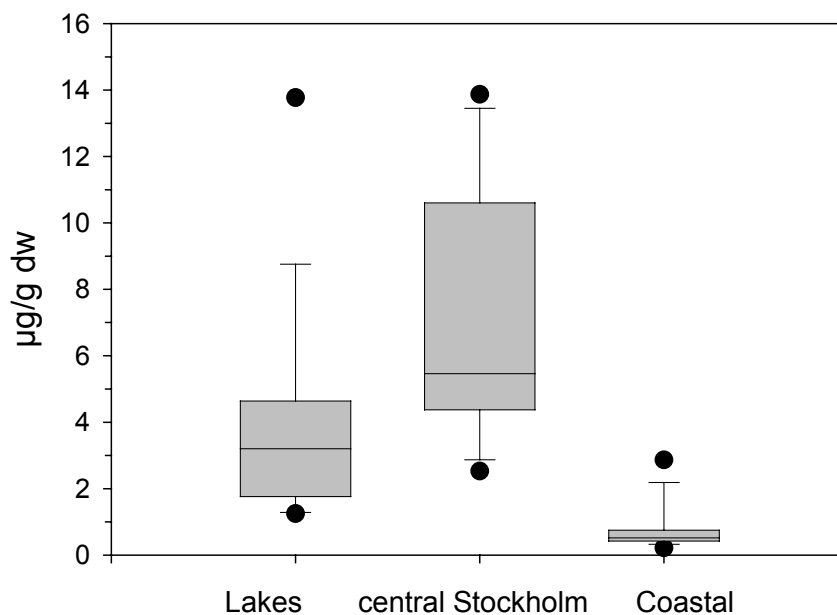


Figure 12. Percentile distribution of $\Sigma 16$ -PAH in the three groups. For explanation of the plot, see chapter 3.3.

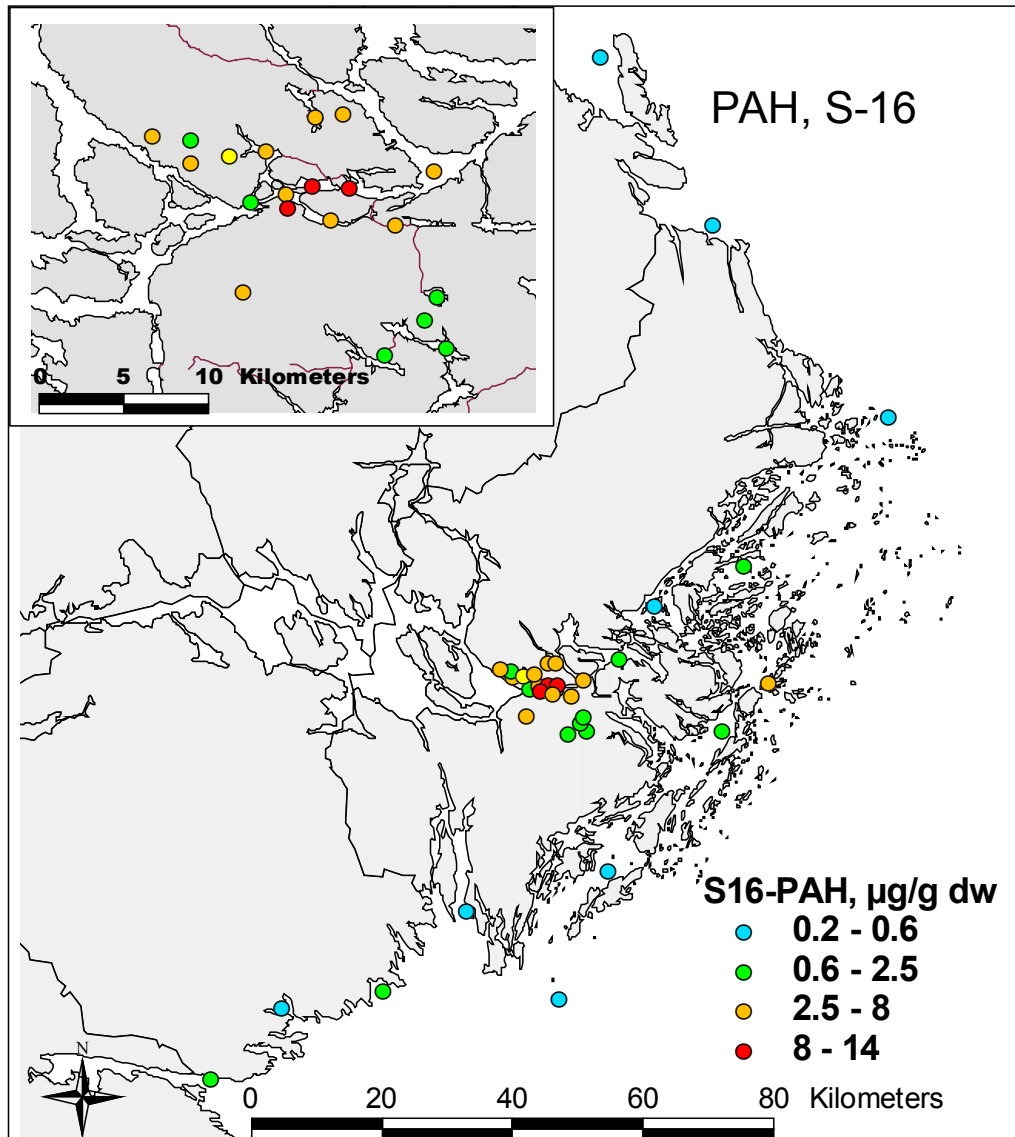


Figure 13. Concentrations of $\Sigma 16$ -PAH. A close-up of Stockholm is provided in the upper left.

4.3 Chlorobenzenes

Eleven different chlorobenzenes were analysed. In most of the samples, neither chlorobenzene was above detection limit (Table 5). Detection limits are given in appendix, and are lower for the coastal sediments. In the Stockholm municipality, one or more chlorobenzenes were found in the following stations: Lillsjön, Brunnsviken,

Trekanten, Reimersholme, Riddarfjärden, Årstaviken and Ulvsundasjön. These are all stations with high concentrations of many other pollutants.

Concentrations of hexachlorobenzene (HCB) in the range 2-19 ng/g dw were recently found in settling matter (sediment traps) from central Stockholm (Broman et al., 2001). In contrast, the present data from central Stockholm are mostly below detection limits (1-7 ng/g dw). The fact that HCB is higher in settling matter than in surface sediments may possibly be due to anaerobic dechlorination in sediments. Under experimental conditions and at 25 °C, HCB has a half-life of ca 27 days in anaerobic sediments (Masunaga et al., 1996). The study also shows that 1,4-DCB is at least an order of magnitude more stable than other chlorobenzenes. Actually, 1,4-DCB is the most commonly found chlorobenzene in our study.

In Stenungsund, SW Sweden, where several chemical industries are located, sediments contained HCB in the range 1.7 – 60 ng/g dw, whereas most other chlorobenzenes were in the range of our detection limits or slightly lower (Brorström-Lundén and Granmo, 1995).

Table 5. Chlorobenzenes. Number of samples with detectable concentrations and maximum concentrations. Detection limits are given in the appendix.

Substance	Lakes		Central Stockholm		Coast	
	samples > d.l.	max, ng/g dw	samples > d.l.	max, ng/g dw	samples > d.l.	max, ng/g dw
1,3-dichlorobenzene	0		0		1	
1,4-dichlorobenzene	1	150	3	26	9	2
1,2-dichlorobenzene	1	16	0		0	
1,3,5-trichlorobenzene	0		0		0	
1,2,4-trichlorobenzene	2	6	3	6	0	
1.2.3-trichlorobenzene	0		0		0	
1,2,3,5-tetrachlorobenzene	0		0		0	
1,2,4,5-tetrachlorobenzene	1	8	0		0	
1,2,3,4-tetrachlorobenzene	0		0		0	
Pentachlorobenzene	1	6	2	4	0	
Hexachlorobenzene	0		1	3	0	

4.4 Benzene and chlorinated solvents

Benzene, 1,2-dichloroethane, dichloromethane and chloroform were below detection limits in all sediments (see appendix for detection limits). This is not surprising because they are volatile substances that do not partition strongly to sediments (chapter 5.4).

4.5 Pesticides

The following pesticides are prioritised in the WFD: alachlor, atrazine, diuron, endosulfan, hexachlorocyclohexanes (HCHs), isoproturon, chlorfenvinphos, chlorpyrifos, pentachlorophenol (PCP), simazine and trifluralin. Only a few of these pesticides are permitted in Sweden (Table 1). In addition, data on the non-WFD pesticides DDTs and chlordanes are presented in chapter 4.13. Of the WFD pesticides, HCHs, chlorfenvinphos and PCP were detected. In addition, simazine and α -endosulfan were detected in a few samples (see appendix), but the identity of these peaks has not been verified with GC-MS and results must be regarded as preliminary.

Neither isoproturon nor chlorpyrifos was detected although they are used in Sweden. This agrees with another recent Swedish study, although isoproturon was found at low levels in a few samples from agricultural areas (Sundin et al., 2002).

Chlorfenvinphos was detected in 17 out of the 34 samples, and in all three geographical groups (Figure 14). The identity of chlorfenvinphos has been verified with GC-MS. Certain samples hold fairly high concentrations. The highest value was obtained from Brunnsviken, which also displays the highest level of α -endosulfan. Actually, an institute for plant protection was located here, and contaminated landfills have been identified leading to the classification as a high-risk area (LST, 2003).

In a compilation based on data submissions from the member states in EC (Klein et al., 1999), chlorfenvinphos was not reported to have been studied in sediments but was occasionally found in the water phase, with mean values of 0.02 $\mu\text{g/l}$. It was not found in water or sediment in the Swedish pesticide monitoring (Sundin et al., 2002; Ulén et al., 2002). Information from the Swedish National Chemicals Inspectorate state that chlorfenvinphos is resistant to degradation in the environment.

Pentachlorophenol shows only moderate variations between the geographical groups (Figure 15). There is a tendency to somewhat lower concentrations in the coastal region (Figure 17) but the total concentration range is rather narrow. Concentrations are very similar to previous measurements in Sweden (Broman et al., 2001; Palm et al., 2002a), although higher concentrations may occur close to former industrial sites (e.g., Palm et al., 2002a).

Three different hexachlorocyclohexanes were determined (α -HCH, β -HCH & γ -HCH). Although β -HCH is a minor isomer in technical HCH, it was a major part (ca 70%) of Σ -HCH in sediments. This is in agreement with β -HCH generally being more persistent than other HCHs. Concentrations of HCHs are fairly similar in the three geographical groups, although substantial variations occur within each group (Figure 16, Figure 18). γ -HCH in the southern Baltic Sea were previously reported with an average of 0.16 and a max of 1.5 ng/g dw (Dannenberger and Lertz, 1996). This is in good agreement with our results. In conclusion, most data do not indicate any strong local influence on the concentration of HCHs.

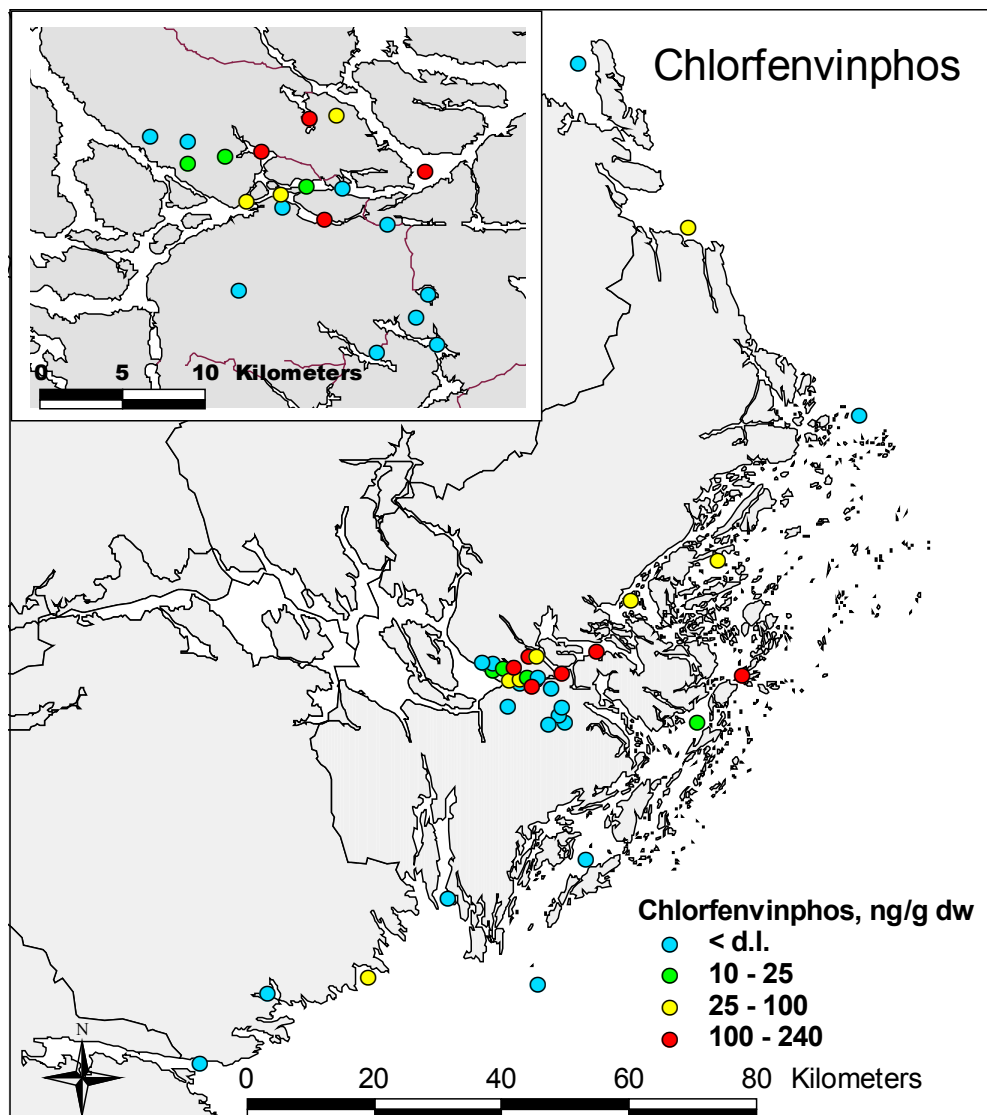


Figure 14. Concentrations of chlorfenvinphos. A close-up of Stockholm is provided in the upper left.

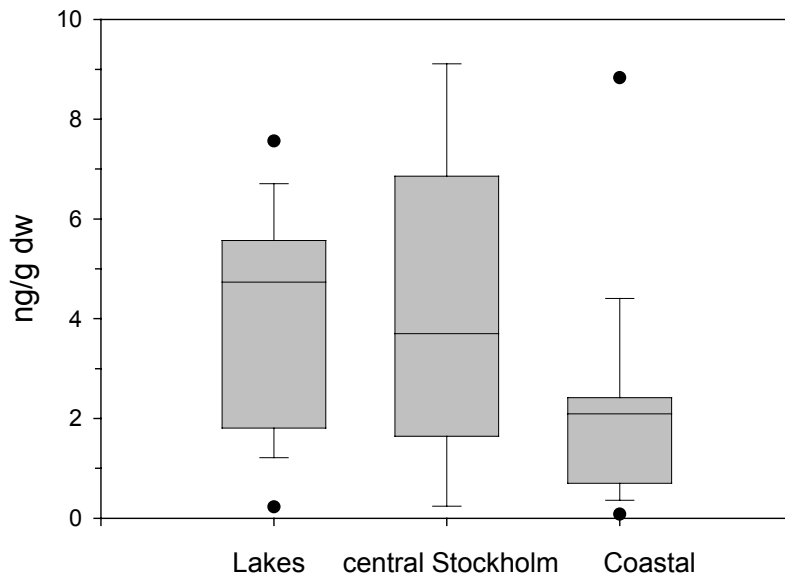


Figure 15. Percentile distribution of pentachlorophenol in the three groups. For explanation of the plot, see chapter 3.3.

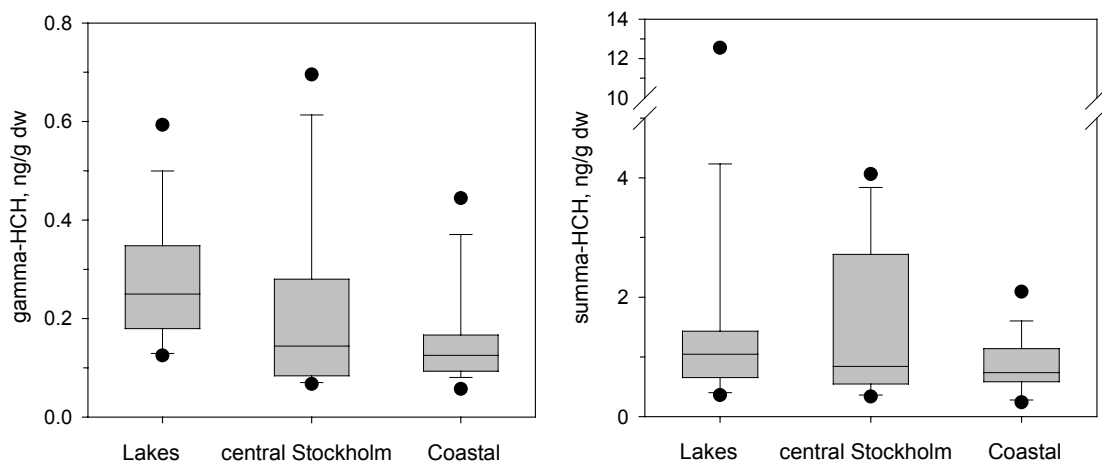


Figure 16. Percentile distribution of γ -HCH and Σ -HCH in the three groups. Please note the break in the y-axis for Σ -HCH. For explanation of the plot, see chapter 3.3.

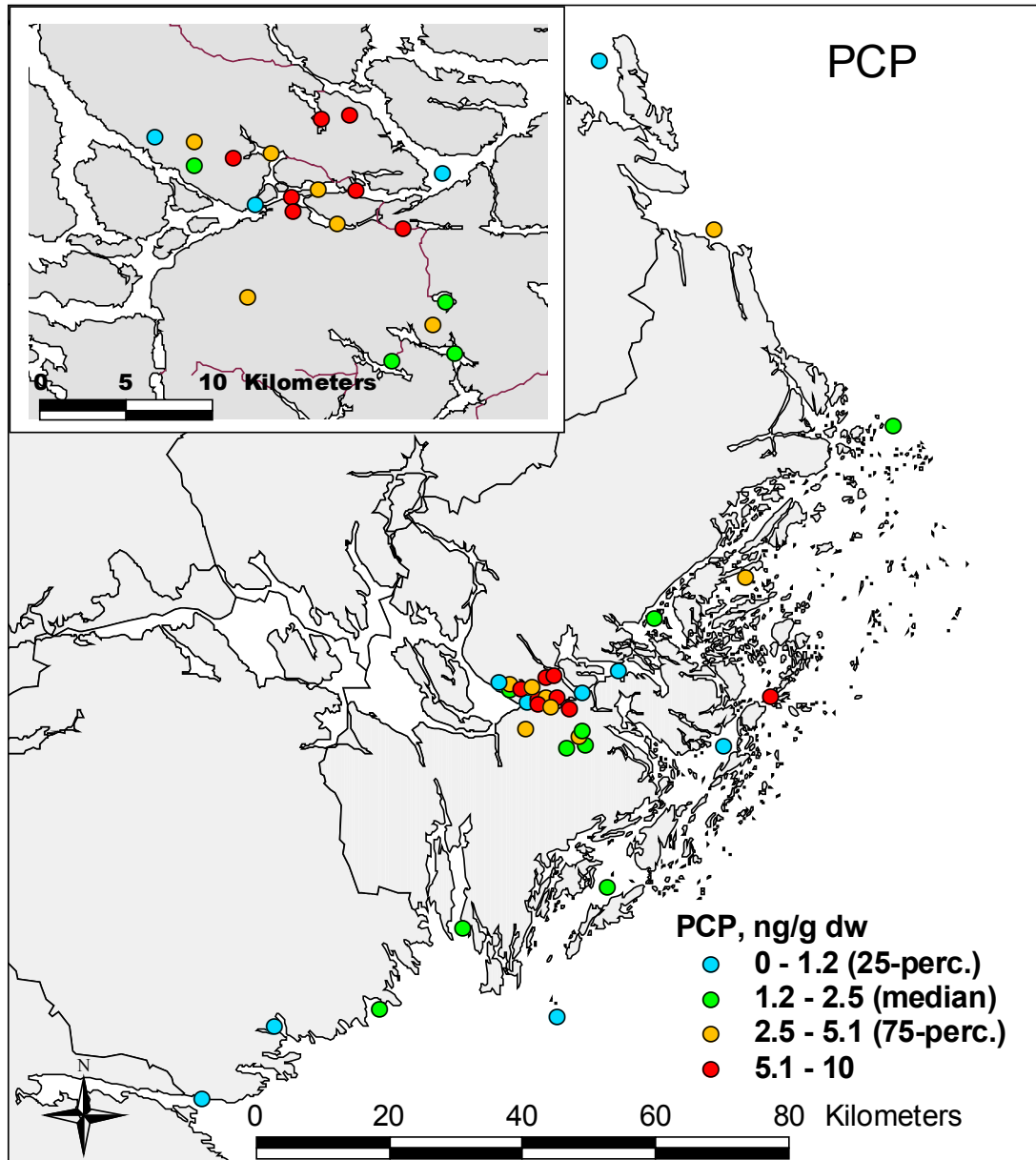


Figure 17. Geographical pattern of pentachlorophenol. A close-up of Stockholm is provided in the upper left.

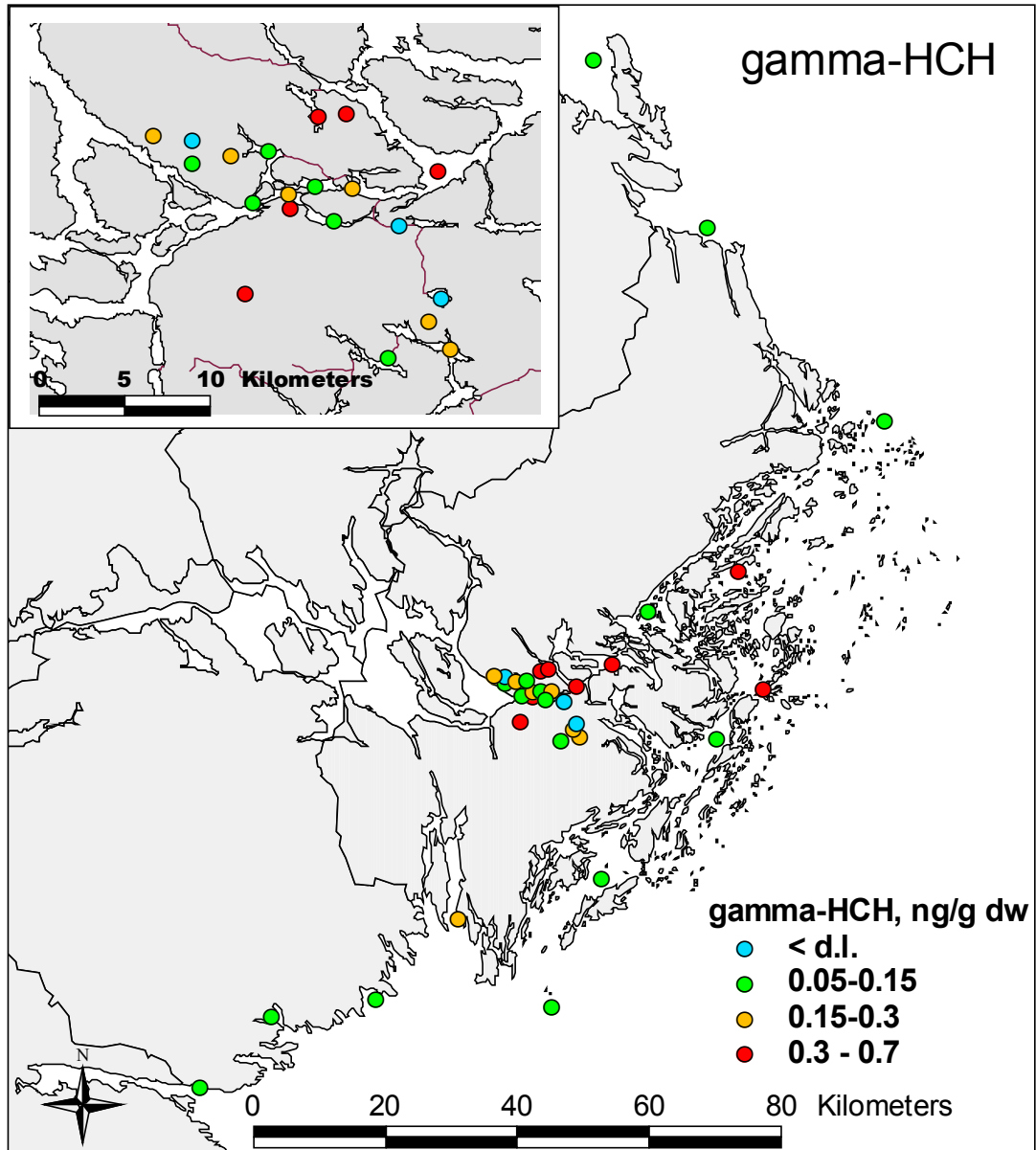


Figure 18. Geographical pattern of gamma-HCH. A close-up of Stockholm is provided in the upper left.

4.6 Di (2-ethylhexyl) phthalate (DEHP)

The concentrations of di-(2-ethylhexyl)phthalate (DEHP) in the three geographical groups are compared in Figure 19 and spatial trends are shown in Figure 20. The general pollution level decreases in the order lakes > central Stockholm > coastal. This contrasts with e.g., PAHs, PCBs, PBDEs and mercury, for which central Stockholm is most polluted. However, because sediment accumulation rates commonly are somewhat lower in the lakes than in the central Stockholm (Östlund et al., 1998; Sternbeck, 1998), the fluxes differ less than the concentrations.

The heterogeneous nature of the lakes is strikingly evident, concentrations spanning over three orders of magnitude. The lowest concentrations are similar to the range previously reported from remote Swedish lakes (10-400 ng/g dw; Parkman and Remberger, 1994). Also central Stockholm is fairly heterogeneous whereas the coastal region is relatively uniform with respect to DEHP, except for one sample located close to Stockholm (östra Askrikefjärden). An evaluation of studies on the degradability of DEHP in sediments conclude that sedimentary DEHP is stable under environmental conditions (KemI, 2001a). Thus, the wide concentration range we observe is likely to result from differences in the load. This suggests that local sources in each catchment have a major control on the distribution of DEHP, particularly in the lakes. This is further discussed in chapter 5.2.2.

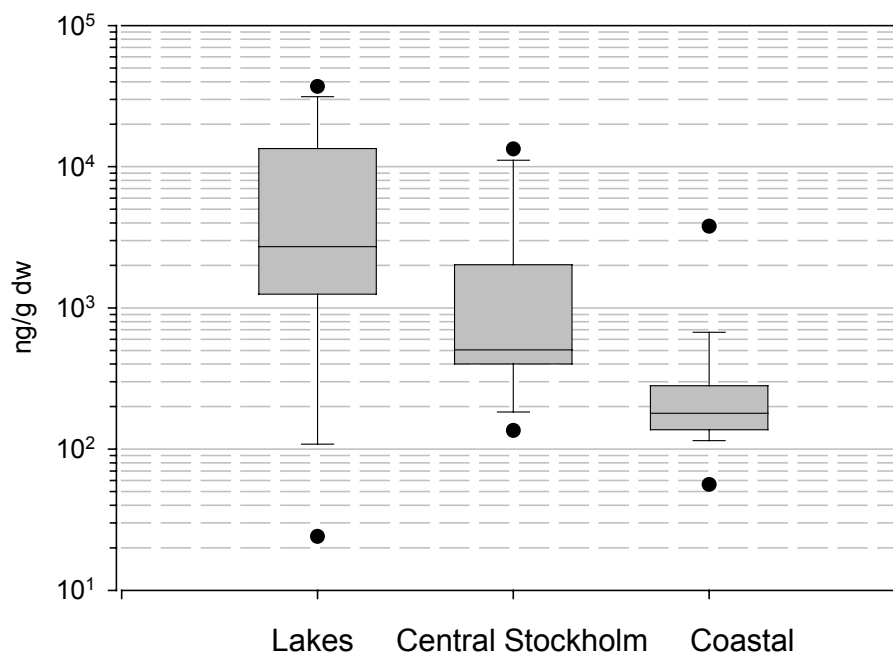


Figure 19. Concentrations of DEHP in the three groups. Note the logarithmic y-axis.

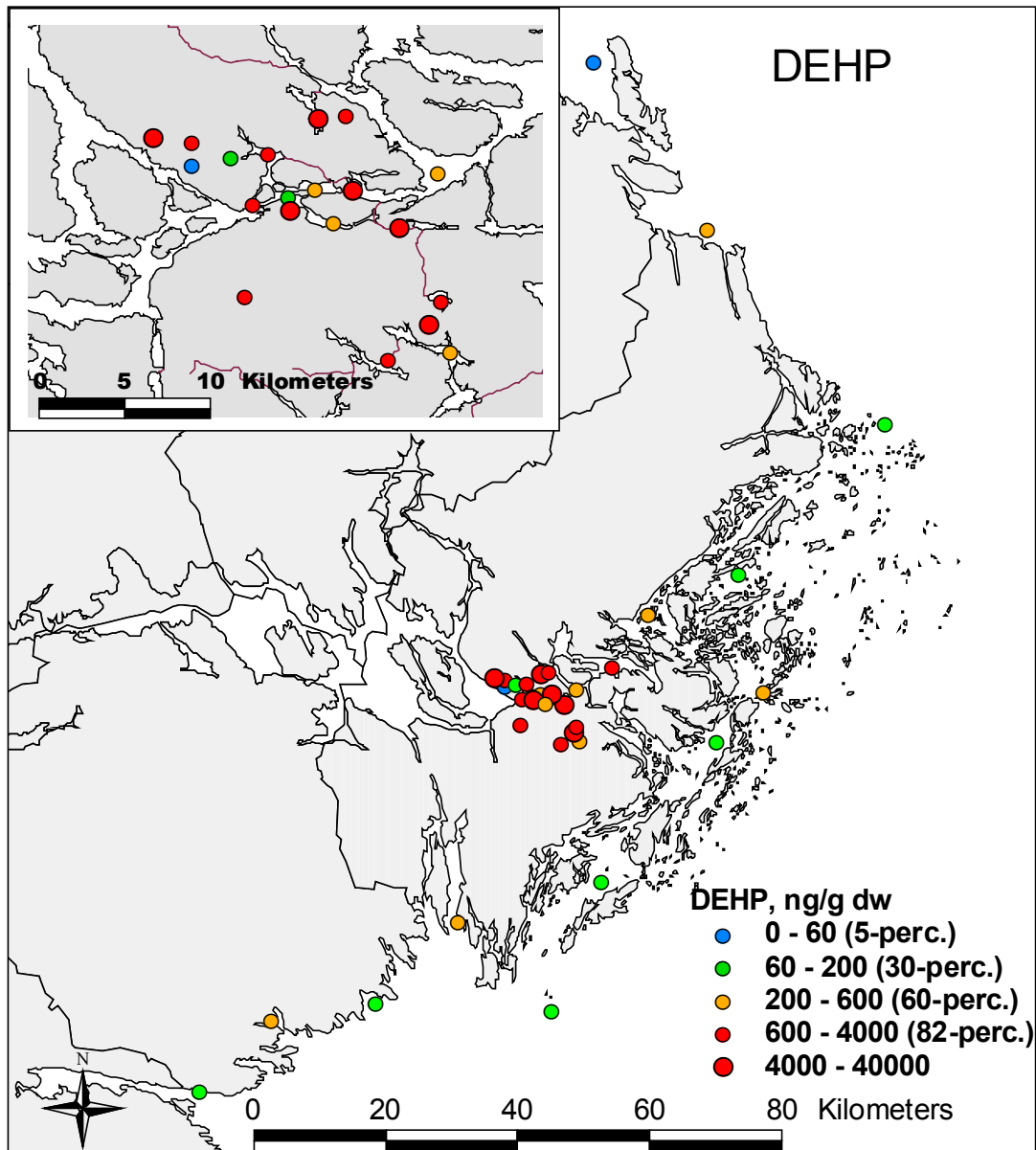


Figure 20. Concentrations of DEHP in the region. A close-up of Stockholm is provided in the upper left.

4.7 Alkylphenols

Concentrations of 4-nonylphenol (NP) and 4-tert-octylphenol (OP) decrease strongly in the order: lakes > central Stockholm > coastal (Figure 21). This is a pattern only shared by DEHP. Concentrations of NP in central Stockholm are very similar to recent data from sediment traps in this area (250-890 ng/g dw: Broman et al., 2001), suggesting that

concentrations in surface sediments are not strongly affected by degradation processes. Other studies have also concluded that NP is fairly persistent under sedimentary conditions (e.g., Hale et al., 2000; Ferguson et al., 2003). Thus, concentrations reflect the anthropogenic load of these substances.

Concentrations of both OP and NP are very low at Essingen (located a few km upstream the city center), increases rapidly in inner part of central Stockholm, and decline progressively from Stockholm to the more remote coastal stations (Figure 23). This clearly shows that OP and NP are released in significant quantities in the city center.

Similar to DEHP, NP and OP in the lakes span over a range more than two orders of magnitude, with lower levels in forest lakes. This illustrates the strong influence of local sources in the catchments, and that sewage treatment plants are not the only large emissions sources. There is a rough correlation between NP and OP in the Lakes and in central Stockholm (Figure 22), suggesting that similar emission pathways may be operating for these two compounds.

Sediment accumulation rates were previously determined for some of the lakes and for the central Stockholm, using ^{210}Pb and ^{137}Cs . Those data can be used to transform concentrations to fluxes of NP and OP. Fluxes differ less than concentrations between the lakes and central Stockholm, although also fluxes are widely distributed within each group. The cause of the high levels in the lakes is further discussed in chapter 5.2.2.

Would it be possible that the releases in Stockholm influence the concentrations of NP and OP in parts of the coastal region? Apart from the apparent releases in central Stockholm, the three STPs in Stockholm (see Figure 2) were estimated to release 41 kg NP to water during 2001 (Stockholm Vatten, 2002). These amounts may be transported to the coastal region by currents, although NP and OP will be removed by settling particles along the way. A study on the influence of a sewage treatment plant on DEHP and NP in water and sediments in Roskilde, Denmark, demonstrated that concentrations level off within a few kilometers, mainly due to settling (Vikelsøe et al., 2001). Similar conclusions were drawn from a study on both NP and OP in Tokyo (Isobe et al., 2001). Except for Östra Askrikefjärden, the nearest coastal stations are at least 20 km away from Stockholm. In analogy with these studies, the slightly elevated concentrations of NP and OP in the coastal region outside Stockholm cannot unambiguously be attributed to releases in Stockholm. Reference data from the open Baltic Sea have not been found.

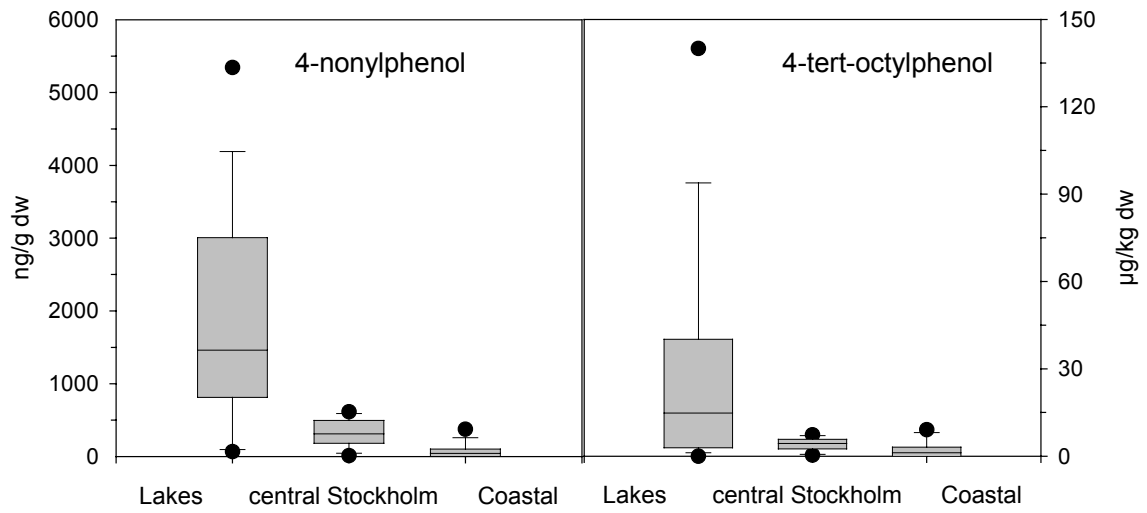


Figure 21. Concentrations of 4-nonylphenol and 4-tert-octylphenol in the three groups.

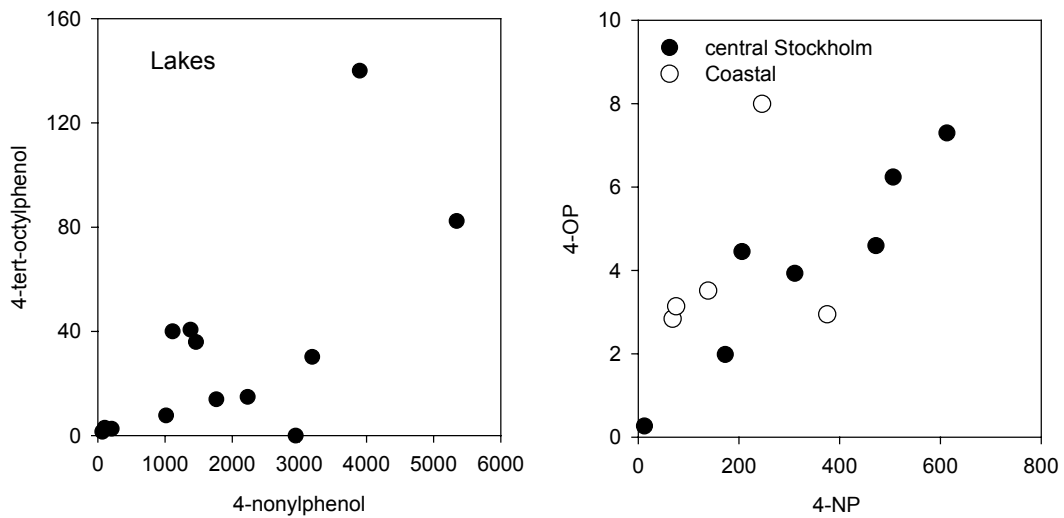


Figure 22. Correlation between NP and OP in the three groups. Concentrations in ng/g dw.

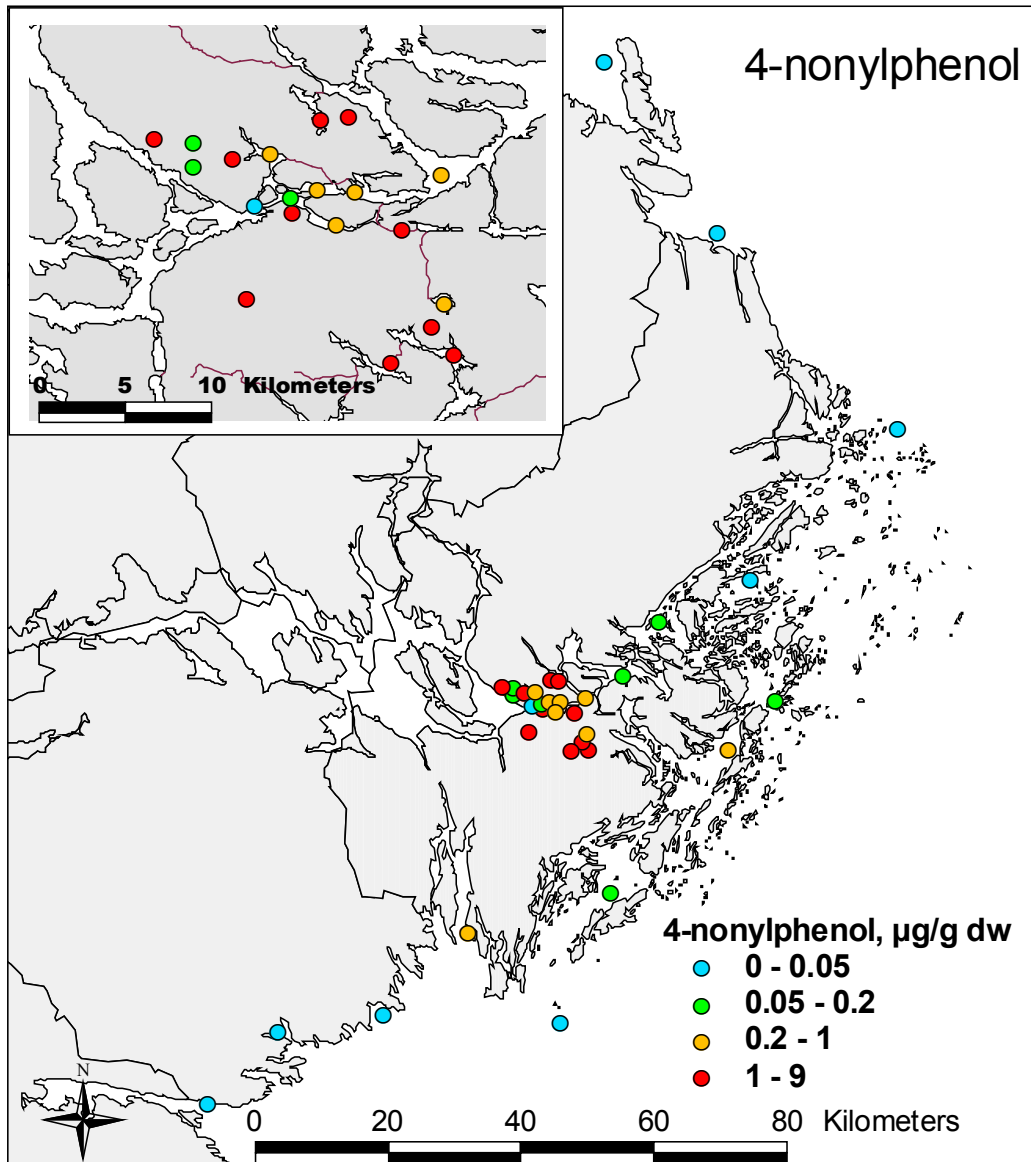


Figure 23. Geographical pattern of 4-nonylphenol. Concentrations are given in µg/g dw. A close-up of Stockholm is provided in the upper left.

4.8 Polybrominated diphenyl ethers (PBDE) and TBBPA

Concentrations of the three congeners generally decrease in the order: BDE-99 > BDE-47 > BDE-100. The PBDE congeners show a clear geographical pattern with highest concentrations in central Stockholm and lowest in the coastal region (Figure 24 and Figure 25). When excluding two high samples, the coastal region shows average concentrations of 0.062 ± 0.025 and 0.092 ± 0.04 µg/kg dw for BDE-47

and BDE-99, respectively. These values are close to other stations in the Baltic Sea (see de Wit, 2000). Thus, an influence from Stockholm on PBDE in the coastal region is not apparent.

Although central Stockholm is clearly enriched in PBDE relative to the lakes and the coastal region, concentrations are similar in sediments from the mouths of many European rivers (de Wit, 2002). Much higher concentrations have been found downstream of certain point sources in the UK (Allchin et al., 1999). There are no known major point source emitters of PBDE in Stockholm, and taken together it is suggested that diffuse emissions are responsible for the enrichment in central Stockholm.

We also analysed tetrabromobisphenol A (TBBPA), another common brominated flame retardant but not a WFD-PS. TBBPA was below detection limit, 5 ng/g dw, in all samples. In Sweden, TBBPA has previously been identified in sediments close to a point source (Sellström and Jansson, 1995) and at very low levels in a few other areas (Remberger et al., 2002). The absence of TBBPA in these sediments may possibly be explained by anaerobic degradation in sediments (Voordeckers et al., 2002).

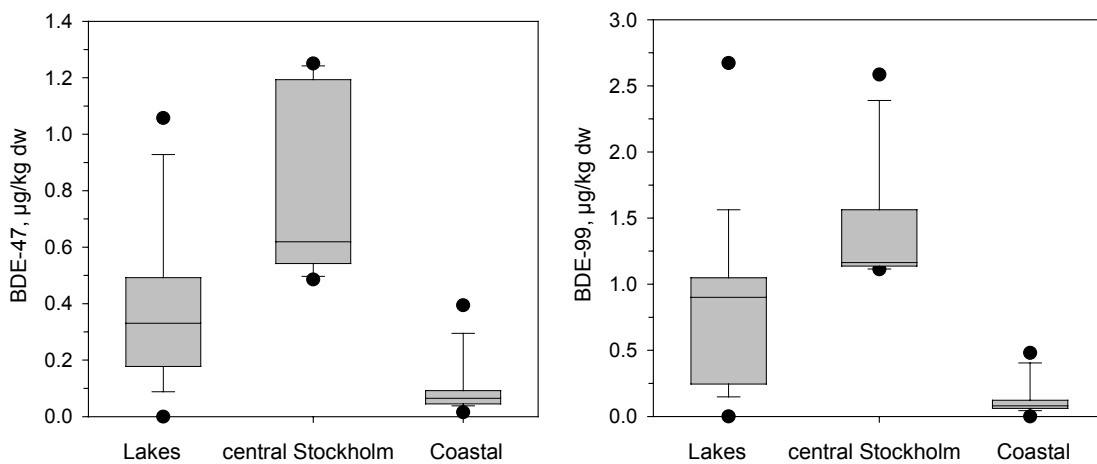


Figure 24. Percentile distribution of PBDE (BDE-47 and BDE-99) in the three groups. For explanation of the plot, see chapter 3.3.

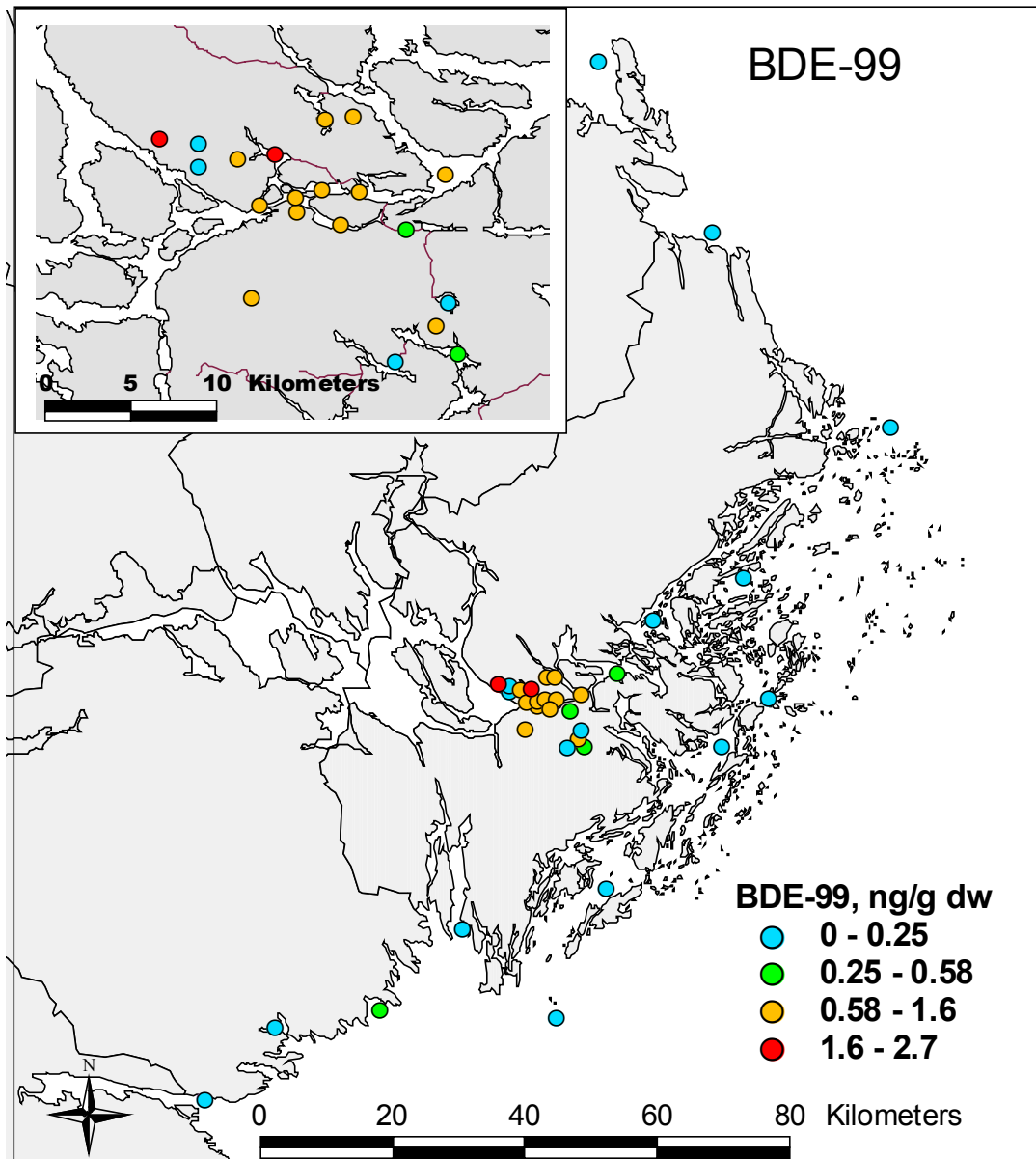


Figure 25. Geographical pattern of BDE-99. A close-up of Stockholm is provided in the upper left.

4.9 C₁₀₋₁₃ Chlorinated alkanes (SCCA)

C₁₀₋₁₃ chloroalkanes (SCCA) were detected (d.l.: 0.3 ng/g dw) in all but one sample. Concentrations generally decline in the order: central Stockholm > lakes > coast, but are highly variable in each group (Figure 26). There are a few exceptionally high values: Räcksta Träsk, Strömmen, and södra Kanholmsfjärden. The latter station is anomalous to the other coastal stations also with respect to e.g. Σ 7PCB (see chapter 5.2.3).

In the lakes, SCCA follow the same pattern as PBDEs and the alkylphenols, with forest lakes (e.g. Judarn and Kyrksjön) displaying relatively low concentrations and Trekanten, Räcksta and Brunnsviken at the high end. However, concentrations of SCCA span over a wider range.

There are only few studies available on the occurrence of SCCA in sediments. Concentrations of ca 2-5 ng/g dw were found in remote Canadian arctic lakes, and long-range atmospheric transport and deposition was suggested as the source of SCCA in these lakes (Tomy et al., 1999). Concentrations were higher in more polluted Canadian lakes, with a maximum value of 260 ng/g dw in surface sediments (Tomy et al., 1999). In a compilation of German studies, SCCA range from 6-83 ng/g dw (ECB, 2000). In this perspective, SCCAs in central Stockholm and several lakes are fairly high.

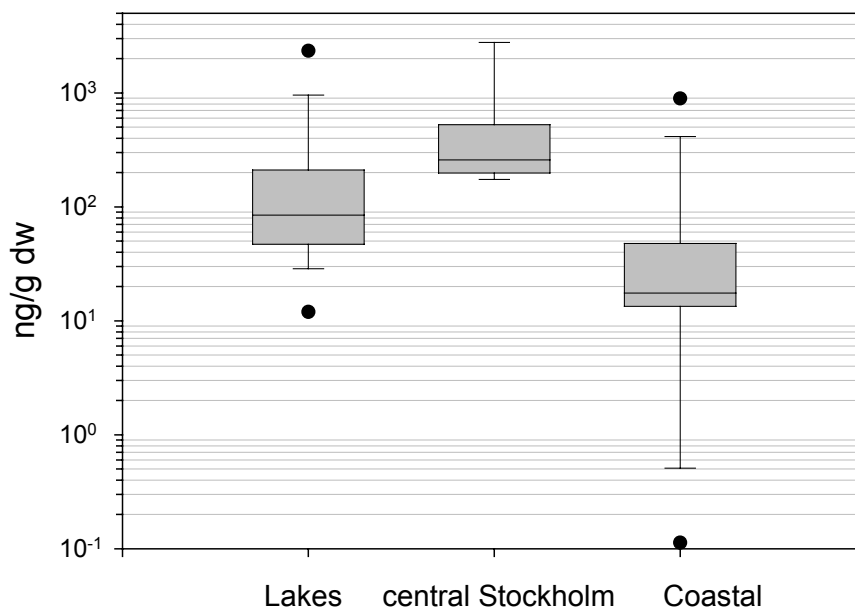


Figure 26. Percentile distribution of C₁₀₋₁₃ chloroalkanes in the three groups. For explanation of the plot, see chapter 3.3. Please note the logarithmic y-axis.

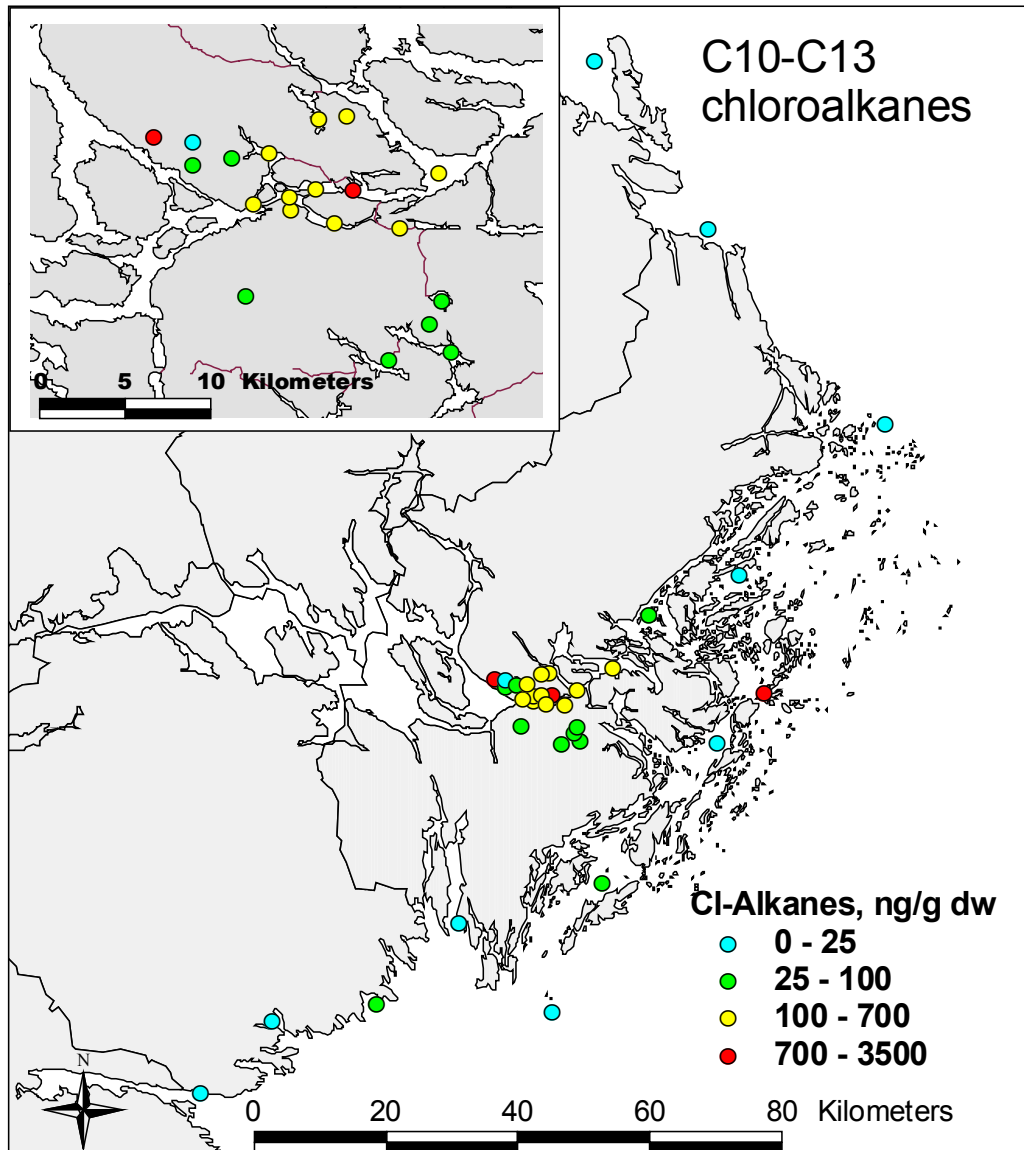


Figure 27. Geographical pattern of C₁₀₋₁₃ chloroalkanes. A close-up of Stockholm is provided in the upper left.

4.10 Hexachloro-1,3-butadiene (HCBd)

Hexachloro-1,3-butadiene was below detection limit (d.l.: 0.2-5 ng/g dw) in all samples. We are not aware of any other studies demonstrating the presence of this compound in sediments.

4.11 Tributyltin (TBT)

Concentrations of tributyltin, TBT, are presented in Figure 28. TBT was detected (d.l.: 2- 11 ng Sn/g dw) in all samples from central Stockholm and in 10 out of 14 coastal samples. In the lakes, however, only 5 out of 13 samples contained detectable amounts of TBT. Four of these five lakes are also polluted with several other substances.

Concentrations generally decrease in the order central Stockholm > coast > lakes. This general pattern is not surprising because a major use of TBT is as antifoulant in paints for large ships. The highest levels of TBT are found in central Stockholm, but in areas that are not typically trafficked by large ships (Figure 29). Although it can not be excluded that release from smaller boats is the explanation, TBT is also known to have other uses. For instance, it occurs as an impurity in other organotins that are used as stabilisers in PVC (KemI, 2000) and has occasionally been reported to occur in various household products. Concentrations in the coastal region are irregular (Figure 29), and similar to previous measurements in the region (Cato, 2003). Generally, concentrations are similar to or lower than on the Swedish west coast (compilation in Brack, 2002). In particular, high concentrations of TBT have been found in harbours and marinas along the Swedish coast and elsewhere (Cato, 2003).

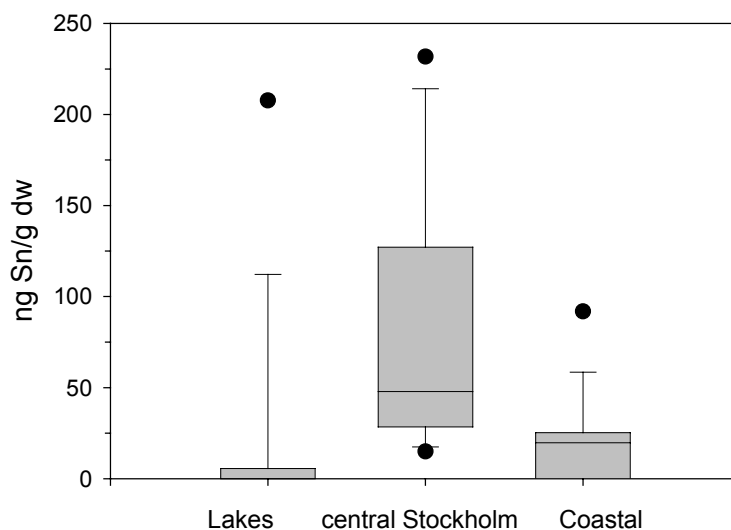


Figure 28. Percentile distribution of TBT in the three groups. In the Lakes, 8 out of 13 samples were below d.l., why no median value or lower percentiles were calculated. For explanation of the plot, see chapter 3.3.

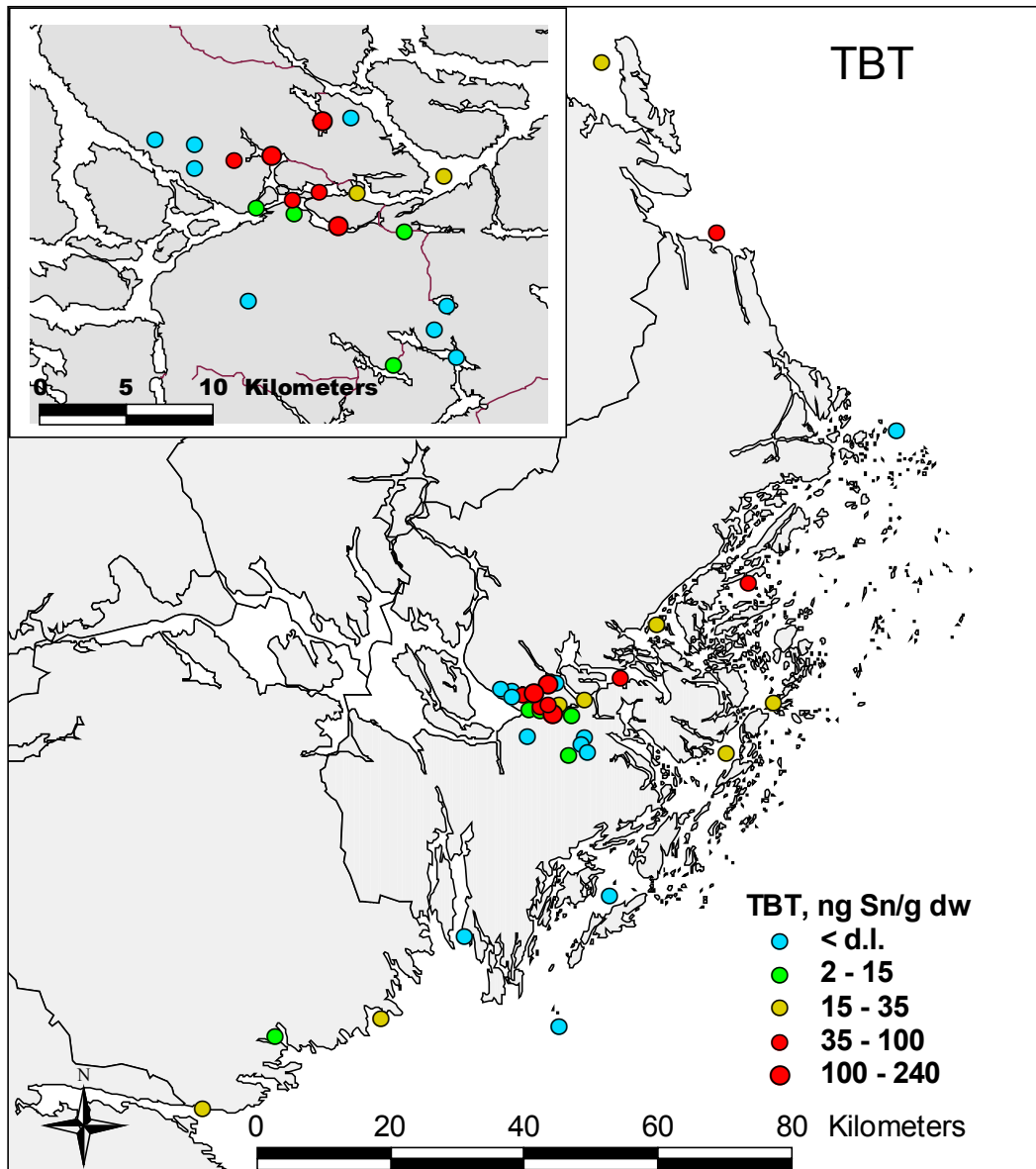


Figure 29. Geographical pattern of TBT. A close-up of Stockholm is provided in the upper left.

4.12 Polychlorinated biphenyls (PCB)

PCB is not on the WFD priority list but is included in this study because it is a suitable reference group of substances. The distribution of $\Sigma 7$ PCB concentrations in the three geographical groups is shown in Figure 31. The individual congeners are presented in the appendix. With respect to $\Sigma 7$ PCB, the lakes and central Stockholm are

heterogeneous. The coastal region is homogeneous except for two stations: södra Kanholmsfjärden and östra Askrikefjärden (Figure 30). The latter value can be explained by the proximity to Stockholm, whereas the former station is discussed in chapter 5.2.3.

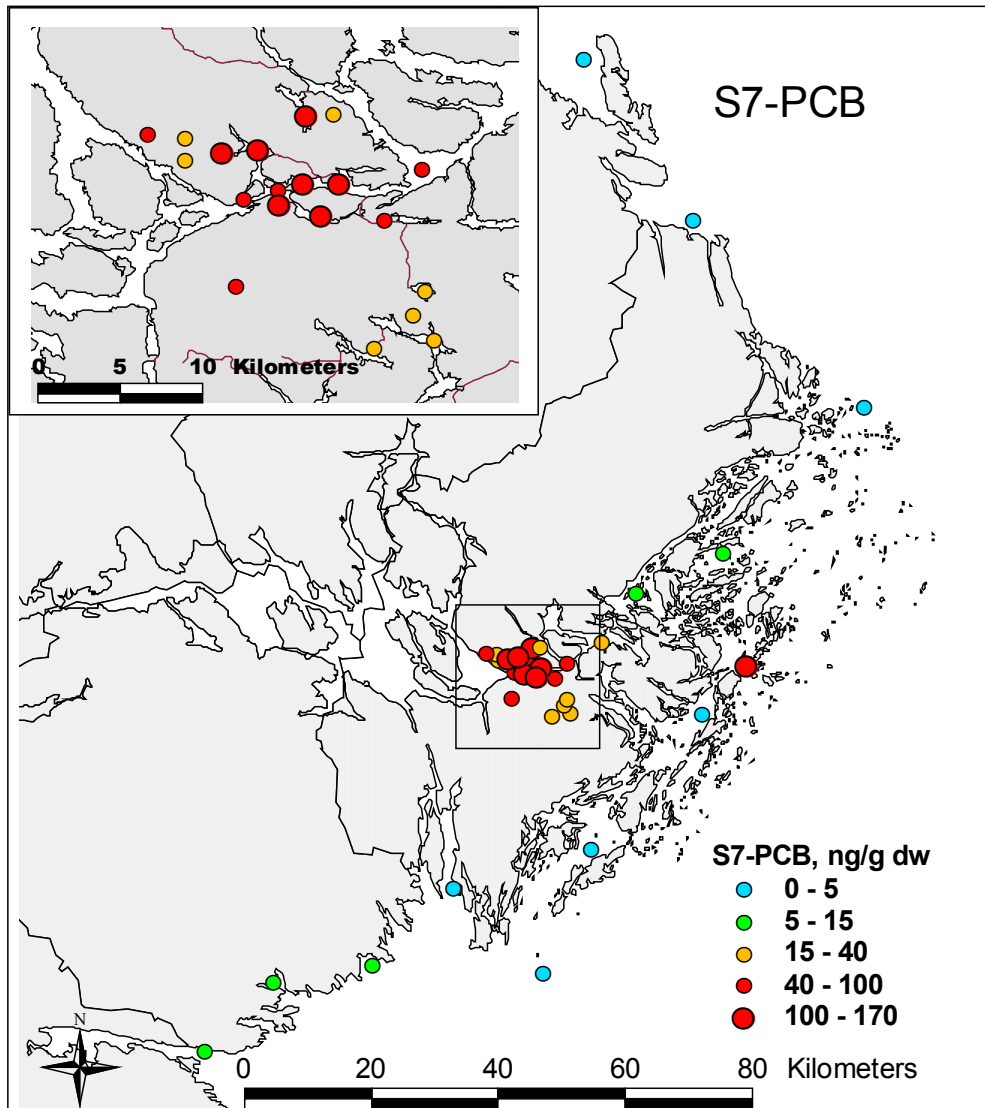


Figure 30. Geographical pattern of $\Sigma 7$ -PCB. A close-up of Stockholm is provided in the upper left.

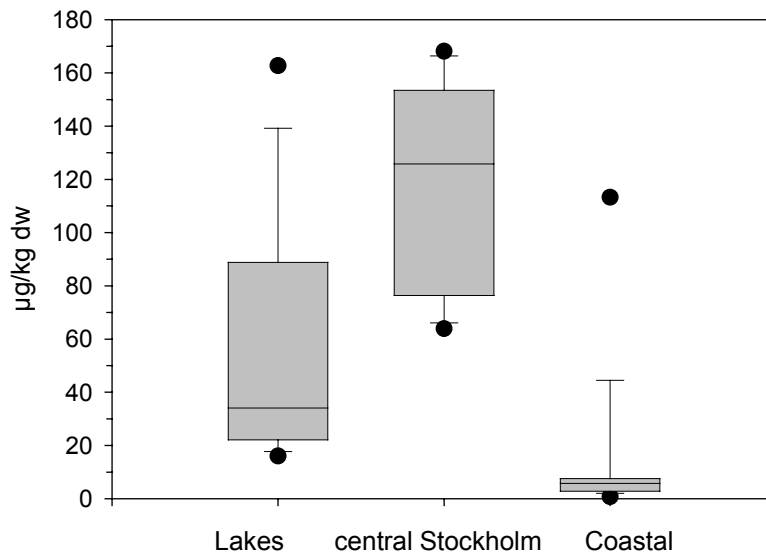


Figure 31. Percentile distribution of Σ 7-PCB in the three groups. For explanation of the plot, see chapter 3.3.

4.13 Non-WFD Pesticides

We also measured some pesticides that are not on the WFD priority list: p,p-DDD, p,p-DDT, p,p-DDE, α -chlordane, γ -chlordane and transnonachlor. Concentrations of Σ -DDT and α -chlordane are shown in Figure 31. Concentrations decrease in the order: central Stockholm > lakes > coast. Certain p,p-DDD values appear relatively high. These values must be regarded as preliminary since they were not verified with GC-MS.

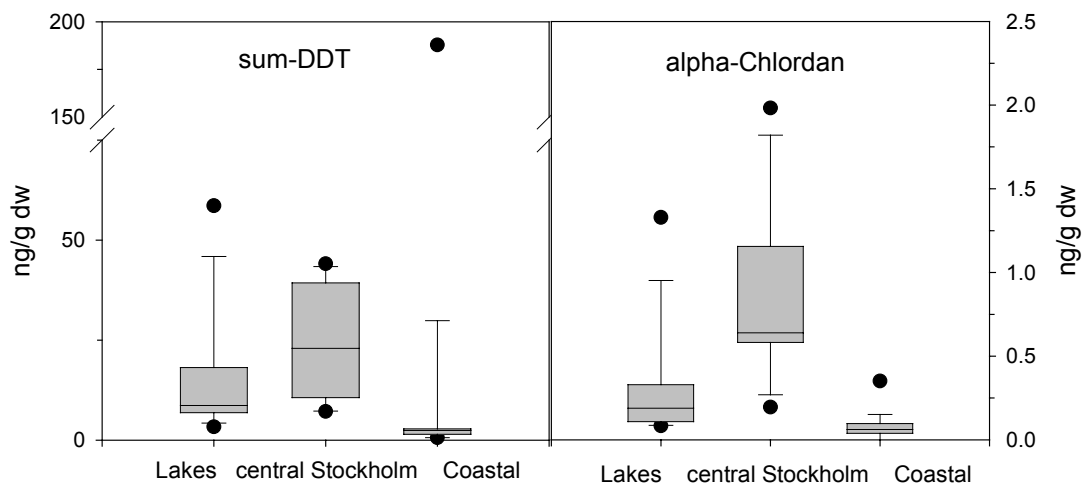


Figure 32. Percentile distribution of Σ -DDT and α -chlordane in the three groups. Please note the break on y-axis for DDT. For explanation of the plot, see chapter 3.3.

4.14 Sewage sludge

Concentrations of all detectable substances are found in Table 6. Detection limits for the remaining substances are shown in appendix. There is generally good agreement between the two samples, which are taken from two of the largest STP's in Sweden: Bromma serving 272 000 pers. equiv. and Henriksdal 621 000 pers. equiv.

Swedish limit values exist for seven metals, and guide values for nonylphenol, $\Sigma 6$ -PAH and $\Sigma 7$ -PCB (Table 6). These values apply for the use of sludge as a fertilizer. No substance (group) exceeds these values. Compared to the averages of Swedish treatment plants during year 2000, only Hg, nonylphenol and PAH are notably higher. National averages are, however, only present for a few substances (Table 6). Concentrations of metals, PAH, PCBs, DEHP and NP are also lower than the preliminary limit values proposed in the European Union (2000), with an exception of Hg for the long term value. However, the value proposed for NP also includes NP-ethoxylates, which we did not analyse.

Nonylphenol is the substance occurring at highest concentrations, demonstrating the strong influence of society on this substance (see chapters 4.7 and 5.2.2). In a longer perspective, concentrations of NP in sludge have decreased in Sweden (e.g., SCB, 2002). Concentrations of DEHP are fairly low compared to other recent Swedish measurements (e.g., Svensson, 2002). Both nonylphenol (-ethoxylates) and DEHP are substances that have been in the focus for many years. Use patterns have changed over time and comparing our data with older data from other STPs is of very limited informative value.

C₁₀₋₁₃ chloroalkanes are among the most abundant organic compounds in these sludge samples. Even higher levels (7-200 mg/kg dw) were recently found in sewage sludge in the UK (Stevens et al., 2003), and it was proposed that leakage from household products could be contributing.

TBT was recently measured in 19 STPs in the southwestern Sweden (Svensson, 2002). Our data are slightly lower than those data that are almost synchronous to our samples. In contrast, their PAH data are generally lower.

PCB in the sludge samples is more enriched in the lighter congeners as compared to the sediments. Probably, this reflects that PCB in sewage sludge is closer to the emission source and that PCB in sediments has had more time to equilibrate.

Table 6. Concentrations in sewage sludge for substances that were above detection limits. The list includes certain substances that are not on the priority list of WFD.

Substance	Unit	Bromma	Henriksdal	Swedish average ¹	Swedish guide value (PAH, PCB, NP) Sw. limit value (metals)
DW	%	29	29		
Σ16-PAH	ng/g dw	3400	4300		
Σ6-PAH	ng/g dw	1600	1800	1200	3000
Benzo(a)pyrene	ng/g dw	200	250		
Σ7- PCB	ng/g dw	150	64	100	400
DDE	ng/g dw	13	12		
p,p-DDD	ng/g dw	15	9.2		
p,p-DDT	ng/g dw	0.37	0.96		
α-HCH	ng/g dw	0.15	0.10		
β-HCH	ng/g dw	0.42	0.26		
γ-HCH	ng/g dw	0.16	0.10		
γ-chlordan	ng/g dw	0.65	0.76		
α-chlordan	ng/g dw	0.45	0.47		
transnonachlor	ng/g dw	0.48	0.50		
α-endosulfan	ng/g dw	≈0.03	0.13		
BDE47	ng/g dw	36	19	54 (large STP)	
BDE100	ng/g dw	9.8	4.8	11 (large STP)	
BDE99	ng/g dw	46	23	62 (large STP)	
Pentachlorophenol	ng/g dw	3.2	1.4		
DEHP	ng/g dw	530	1920		
4-nonylphenol	ng/g dw	43 000	31 000	17 500	50 000
4-tert-octylphenol,	ng/g dw	960	780		
TBT	ng/g dw	18	35		
TBT-Sn	ng/g dw	7.5	15		
C ₁₀₋₁₃ chloroalkanes	ng/g dw	2250	2300		
Cd	µg/g dw	1	1	1.1	2
Hg	µg/g dw	1.4	2.1	1.0	2.5
Pb	µg/g dw	26	28	34	100
Cr	µg/g dw	40	31	31	100
Cu	µg/g dw	380	360	374	600
Zn	µg/g dw	550	540	550	800
As	µg/g dw	1.5	1.7		
Co	µg/g dw	7	8		
Mn	µg/g dw	140	130		
Ni	µg/g dw	23	27	17	50

1. References to Swedish averages: Nylund et al. (2002): PBDE; SCB (2002) PAH, PCB, nonylphenol and metals.

5 Discussion

5.1 General observations

Overall, there is excellent agreement between which substances that we find in sediments and what could be expected on the basis of known uses and emissions, regulation etc. (Table 1). Of the substances known to be used or emitted in Sweden, the following were found, and were also enriched in Stockholm compared to the coast: Cd, Cr, Cu, Hg, Pb, Zn, PAH, PBDE, nonylphenol, octylphenol, C₁₀₋₁₃ chloroalkanes, DEHP and TBT. We did not find the pesticides isoproturon or chlorpyrifos, which actually are used in Sweden. Except for a few samples, these were neither found in sediment monitoring of Swedish agricultural areas (Sundin et al., 2002). The region studied here is no typical agricultural region, and isoproturon and chlorpyrifos are therefore less likely to occur at detectable concentrations. Neither did we find hexachlorobutadiene (HCB) or TBBPA. Although HCB is not regulated in Sweden, there are no known uses either. That the common flame retardant TBBPA was below detection level may be due to anaerobic debromination in sediments. Furthermore, benzene and the chlorinated solvents were not found although most of them are used and emitted in Sweden. This is logical because they do not partition strongly to sediments (chapter 5.4), and are probably mainly emitted to air.

Some substances that are not permitted were also found: chlorobenzenes, HCHs, pentachlorophenol, chlorfenvinphos, α -endosulfan and simazine, and the non-WFD substances PCBs, DDTs, and chlordanes. Most of these substances are known to be persistent in the environment. Their occurrence in surface sediments may therefore be influenced by redistribution from previously released amounts. Resuspension of older contaminated sediments in more shallow areas is one possible explanation, in particular for PCBs that is known to occur at higher concentration in deeper (older) sediments (e.g. Östlund et al., 1998). Leaching from contaminated soils is also possible. Furthermore, emissions of PCBs still occur from certain buildings.

Long-range atmospheric transport (LRT) and deposition is most likely important for the general occurrence of HCHs, p,p-DDE, p,p-DDT and hexachlorbenzene (compare e.g., Beyer et al., 2000; Brorström-Lunden et al., 2003). For HCHs, insignificant differences between the lakes, central Stockholm and coast support this interpretation, although certain stations show evidence of local sources. Also PCP shows minor variance between the geographical groups, indicating low importance of local sources. Long-range atmospheric transport is most likely important also for PCBs and PBDEs in the coastal region, whereas local sources are clearly more important in Stockholm. Chlorobenzenes occurred sporadically, 1,4-DiCB being the most common.

Chlorobenzenes may possibly be unintentionally formed, e.g. during high-temperature processes involving chlorine. Some CBs may also be degradation products of other chemicals, including HCB (e.g., Masunaga et al., 1996). The presence of chlorfenvinphos is a little unexpected, although it was permitted in Sweden very recently. Its irregular occurrence does not support LRT as a source. At least at certain sites, both chlorfenvinphos and endosulfan may derive from former industrial sites

The WFD-PS's were recently investigated in several fish species from two large Swedish lakes, Vänern and Vättern (Öberg, 2003). Fewer compounds were found as compared to the present study, namely: HCB, HCHs, PCP, TBT, chloroform, DEHP, dichloromethane, naphthalene, Ni, Pb och Hg. One PAH, benzo(b)fluoranthene, was detectable in one sample. Of these, we did not detect dichloromethane, probably because it does not partition strongly to sediments.

5.2 Geographical trends

The general trends are illustrated in Figure 33, using the relative distribution of median values between the three geographical groups. Most substances are less abundant in the coastal region than in Stockholm. As pointed out in the results section, only DEHP and the alkylphenols are pronouncedly higher in the lakes than in central Stockholm. Those metals that show little anthropogenic impact are about equally distributed between the groups (As, Cr, Mn, Ni and Co). In the following, we will briefly discuss trends within each group.

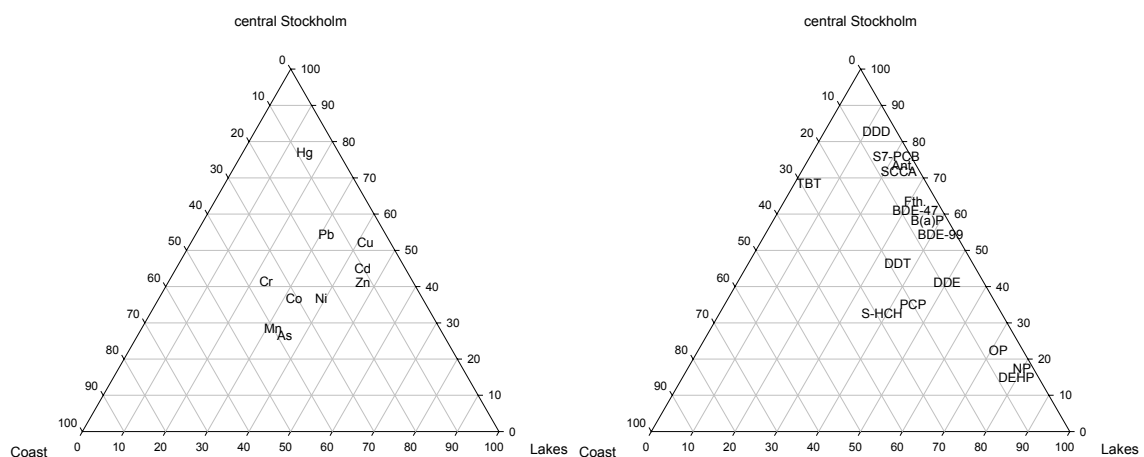


Figure 33. Relative distribution of the median values in the three groups. Fth: flouranthene, BaP: benzo(a)pyrene, Ant: anthracene, OP: octylphenol, NP: nonylphenol, SCCA: C10-C13 chloroalkanes.

5.2.1 Central Stockholm

Of the seven stations in central Stockholm, Essingen is located upstream of the city center and may serve as a reference, and Fjäderholmarna is a downstream reference. The following substances show at least a twofold enrichment in the city center relative to Essingen: Cd, Cu, Hg, Pb, Zn, PAHs, NP, OP, PCP, C₁₀₋₁₃ chloroalkanes, HCHs, TBT, PCBs, DDTs and chlordanes. This may be interpreted as an urban influence on these substances. Central Stockholm may also be compared to the surrounding lakes and is clearly enriched in Hg, PAHs, PBDE, TBT and PCBs. Considering the clear enrichment of PBDE, it is surprising that PBDE was not enriched in the city center relative to Essingen. Similar results was, however, also obtained with sediment traps (Broman et al., 2001). Possibly, PBDE in Stockholm is also influenced by unknown sources upstream in Lake Mälaren.

5.2.2 Lakes

Most substances show very wide scatter in the lakes group, suggesting that local factors are important. The lakes differ significantly in residence times and in the character of the catchment (Table 3). Certain lakes have large catchments with a large share of impervious surfaces. Pollutants that are subject to atmospheric transport and deposition will not be retained to the same degree in catchments with impervious surfaces, as compared to catchments covered by vegetation. A slight enrichment of LRT-substances, e.g. HCHs (Figure 16), is therefore to be expected in certain lakes compared to the coastal region, and do not necessarily imply that local sources are present. This does also apply to central Stockholm.

DEHP, nonylphenol (NP) and octylphenol (OP) are clearly higher in the lakes compared to central Stockholm, although the scatter is wide. Actually, the lowest value of DEHP in the entire study is in a lake (Judarn). Several lakes also hold very low levels of NP and OP. Sewage treatment plants have previously been considered as a major source of NP and OP to the environment (e.g., Bennett and Metcalfe, 2000). However, releases from STPs cannot explain the present situation, since the STPs in Stockholm only influence two central Stockholm stations and none of the lakes. The data strongly suggest very local influence, probably by diffuse emissions in the catchments.

The highest concentrations of DEHP in the present study (Trekanten, Brunnsviken and Räcksta Träsk; 20 000, 30 000 and 37 000 ng/g dw, respectively) are similar to what Parkman and Remberger (1994) measured in sediments close to two point sources (33 000 and 47 000 ng/g dw). Road traffic has sometimes been proposed as a DEHP source. Although these three lakes are heavily influenced by road traffic, so are other stations where DEHP levels are low (e.g., Lillsjön). The major use of DEHP is as an additive in various PVC products.

Nonylphenols are not used widely, but are formed in the environment through degradation of NP-ethoxylates. NP-ethoxylates (NPE) have been replaced with other substances in many former diffuse NP sources, e.g., car wash detergents. The latter use ceased during 1990/91 in Stockholm, resulting in rapid decline of NP in sewage sludge from ca 1000 to 200 mg/kg (Wahlberg, pers. comm.). The reported use of NPE in Sweden has decreased by more than 90% since 1990 (KemI, 2001b). Both DEHP and NPE are common in building components and chemical products used in, e.g. plastic floors, roofs, water based paints, wood preservatives etc (e.g. KemI, 2001 b). Many lakes are situated in fairly modern communities, whereas elder buildings probably dominate in central Stockholm. It can be assumed that DEHP, NPE and OPE were much less common in the elder buildings. We suggest that one important factor contributing to the enrichment of alkylphenols and DEHP in lakes could be the relatively modern communities in the catchments. In line with our hypothesis, a study in 120 US homes actually shows that DEHP and NP were among the most abundant substances in indoor air and dust (Rudel et al. 2003).

This hypothesis is supported by detailed analysis. For instance Lake Flaten, which is a forest lake, display a high level of DEHP (11 000 ng/g dw). Actually, Flaten receives storm water from a relatively large community that was built during the early 1980's. Local factors such as character of the catchment, traffic storm water and other sources are superimposed on this pattern, leading to the wide scatter of DEHP and NP/OP in the lakes. Finally, because lake sediments commonly are mixed at the surface (chapter 2), we cannot totally exclude that concentrations are influenced by previous emissions. This will hopefully be investigated in a future study on time trends in sediments.

5.2.3 Coastal region

Except at a few stations, most substances are relatively homogeneous in the coastal region. Södra Kanholmsfjärden deviate from the other coastal stations by showing higher concentrations of Σ 7PCB, Σ -DDT, PCP, α -HCH, γ -HCH, C₁₀₋₁₃ chloroalkanes and PAH. In particular the Σ 7PCB concentration must be regarded as strongly elevated, being similar to the highest values in Stockholm. The congener composition of PCB deviate from all other samples and suggests that the station is close to a source of high-chlorinated PCB. The Σ -DDT concentration at Södra Kanholmsfjärden (ca 190 ng/g dw) is actually highest in the entire dataset and much higher than any measurement on the Swedish west coast (mean 1.3, max 6.6 ng/g dw; Cato, 1997). A well-known dumping site is situated in a deep ca five km north of our station (B. Sundqvist, pers. comm.). Large amounts of ammunition, boats, cables, refrigerators etc have been identified here. However, there is no specific evidence that these dumped materials would contain these pollutants. At present, the cause of these high levels is unresolved. Because the Baltic coast is a sensitive region, it is very important to determine the areal extent of this pollution as well as their causes, through more detailed investigations.

Östra Askrikefjärden, which is the station located nearest Stockholm, displays high levels of PAHs, PBDEs, DEHP, TBT, C₁₀₋₁₃ chloroalkanes, Σ 7-PCB, Σ -DDT, Cd, Cu, Hg, Pb and Zn, relative to most other coastal stations. Also NP and OP are fairly high here. We consider that östra Askrikefjärden is influenced by the proximity to central Stockholm, and possibly to the outlet of a STP nearby. An influence of central Stockholm on the coastal region outside östra Askrikefjärden is not evident, with a possible exception of OP and NP.

This agrees perfectly with a study where sediment traps were placed in a gradient from central Stockholm to the outer archipelago (Broman et al., 1988). Concentrations and fluxes of PAH and lead decreased rapidly downstream central Stockholm. An urban influence was recognized in Östra Askrikefjärden, but not further eastward.

There are no other coastal stations that seem polluted in general, although individual substances are high at certain stations. For instance, PBDE is rather high at Tvären and NP is high in Svärdsfjärden. Actually, it was shown in the chapter 4 that most PBDE levels are similar to the open Baltic Sea, and that γ -HCH is similar to data from the southern Baltic Sea. For PBDE, this conclusion agrees with modelling results, showing that PBDE partitions more strongly to sediments than any other substance in this study (chapter 5.4). The metals As, Co, Cr and Ni are almost identical to preindustrial levels in the Baltic Sea (Anon, 1999). Cadmium, Cu, Hg, Pb and Zn are actually lower than surface sediments from the open Baltic proper sampled during the early 1990s (Borg and Jonsson, 1996).

5.3 Changes since 1997?

Data from central Stockholm and the lakes can be compared with results from the earlier study (Östlund et al., 1998; Sternbeck and Östlund, 2001). Five years have passed since those sediments were sampled. At several stations, the 2-cm interval presently studied should mainly represent sediment that accumulated since then. Some of the smaller lakes will, however, most likely show a pronounced overlap with the older samples (chapter 2). Even if no changes in pollution load would have occurred, one should not expect identical values at each station due to natural variation, analytical uncertainties etc. Comparing the whole dataset is a more reasonable approach, whereby random errors may cancel.

Examples are presented in Figure 34 and Figure 35. It appears that data from the two studies agree very well. When normalising each data point with the corresponding 1997 data, the median values for the 20 Stockholm stations varies between 0.8 and 0.9 for the metals that display anthropogenic influence (i.e., Cd, Cu, Pb, Zn and Hg), indicating that concentrations decline. In particular Cd but also the other metals have previously shown long-term declining trends in dated sediments from the region (Östlund et al.,

1998; Sternbeck and Östlund, 2001). Chromium, As and Ni show median values of 1.0, i.e. no change. Cobolt and Mn show a slight increase (1.1) which very well could be due to redox processes in the surface sediments. At several stations, PAH concentrations are identical at the two occasions, but show larger scatter at higher values. No general trend can be identified.

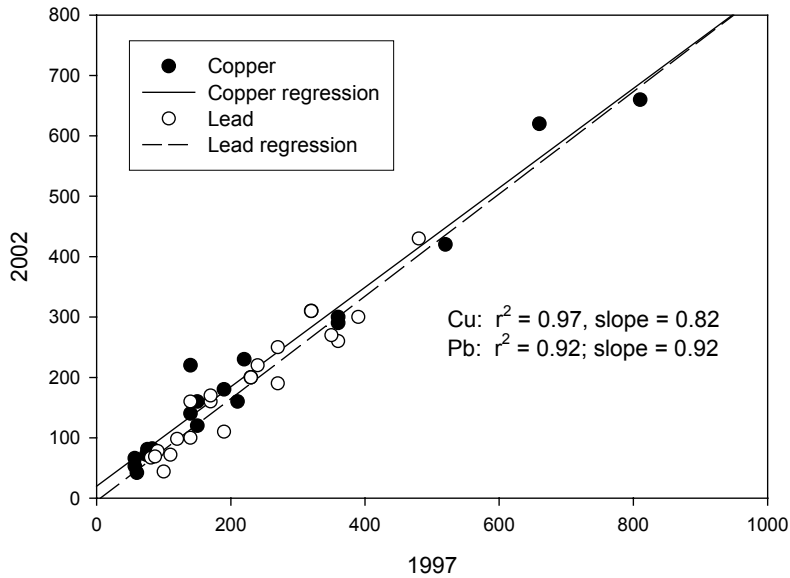


Figure 34. Relationship between Cu and Pb in samples from 1997 and 2002. Concentrations in mg/kg dw.

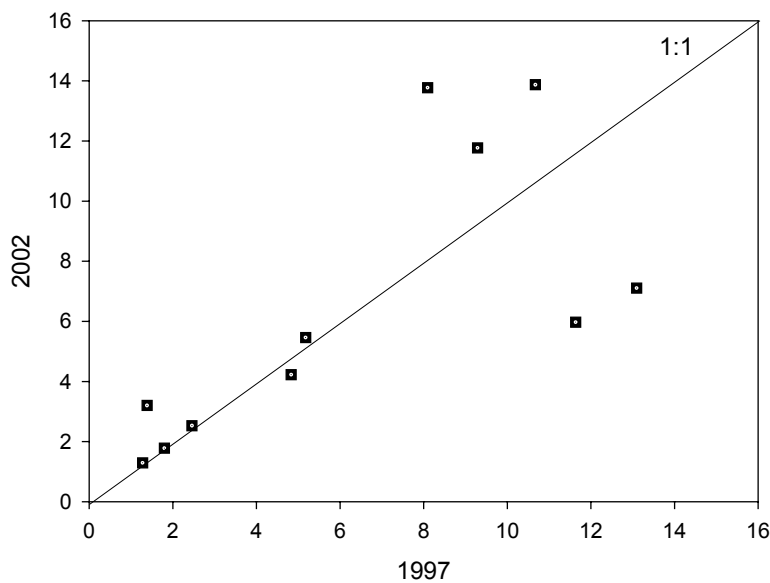


Figure 35. Relationship between Σ16-PAH (µg/g dw) in samples from 1997 and 2002. A line shows where concentrations are equal.

5.4 Are the levels in sediments representative for the substances flow in the aquatic environment?

The fact that concentrations in sediments differ widely between the different chemicals cannot be directly interpreted as a difference in the magnitude of their releases. The WFD-PS represent a wide range of physical-chemical properties which will affect their tendency to partition to sediments.

In order to highlight the differences in partitioning properties between the chemicals investigated, modelling exercises were performed with two different multimedia models. The first model used is the so-called Equilibrium Criterion (EQC) model (Mackay et al., 1996), which is a fugacity-based, steady-state, evaluative model representing a hypothesised environment with a surface area of 100 000 km² and a 10 % water coverage. The model contains four compartments (air, soil, water and sediment) and has been applied to a number of chemicals for evaluative assessment of their environmental fate (MacLeod and Mackay, 1999; Booty and Wong, 1996). It is suggested that the EQC model be used for evaluating the fate of chemicals in a relative perspective, i.e. to highlight partitioning differences, rather than for site-specific, quantitative purposes. The second model used here is the CeStoc model (Palm, 2001), which is based on the same principles as the EQC model, but parameterised to central Stockholm with the specific environmental properties of this region. It contains five media: air, water, soil, sediment and an organic film which covers the impervious surfaces in the city. The CeStoc model has previously been applied to fluoranthene and polybrominated diphenyl ethers (Palm, 2001) and to the flame retardant hexabromocyclododecane (Palm et al., 2002b). The model may be used for quantitative estimations of the steady-state distribution, transported amounts and overall residence times of chemicals, providing that release rates and background inflow concentrations are known. In this study, however, the CeStoc model was applied merely for evaluative and relative purposes.

Physical-chemical properties of the chemicals of interest were mainly collected from the SRC database (Syracuse Research Corporation, 2003). The models used are especially adapted to organic, non-ionic chemicals. Metals, tributyltin and the three pH-dependent chemicals (PCP, simazine and atrazine) were thus not included in this exercise. Degradation half-lives were all estimated with the EPIWIN estimation software (Meylan, 1999). It is a well-known fact that degradation half-lives, together with emission rates, are the most uncertain input parameters in these kinds of modelling exercises. Estimated half-lives often differ by large from measured values (e.g. Palm et al., 2002c). For some of the chemicals investigated here, there may well be more accurate information available on degradation rates. However, these values were considered to be sufficiently accurate for the purpose: to obtain a crude relative ranking of the substances rather than to make quantitative predictions.

In the EQC simulation, the release rate was set to 1000 kg/hour, whereas the emission rate used for the Stockholm model was 100 kg/year. In both cases, only emissions to water were considered. There is a linear relationship between emissions and concentrations in these models. The release rate does therefore not affect the relative ranking of the chemicals.

Table 7 shows the resulting percentage distribution in the sediment compartment. This percentage is calculated as the amount of chemical in sediments in relation to the amount that stays in the system; part of what is released may also be transported out of system. The percentage is also related to the percentage of the most “potent” chemical, i.e. the chemical that has the strongest tendency to partition to the sediment compartment. The chemicals were also ranked with the two models, with rank 1 assigned to the most “potent” chemical. As shown in Table 7, both EQC and CeStoc predict the three brominated diphenyl ethers to be the most “potent” chemicals. The two models also agree about the 13 least potent chemicals. The correlation is worse for the chemicals ranked from 4 to 22. It is also clear that CeStoc predicts much higher percentage distribution in the sediments than the EQC model does, and the physical-chemical properties seem to have a smaller impact on the general tendency to partition to sediments in the CeStoc environment. Even fairly water-soluble and volatile chemicals such as naphthalene, are predicted to prefer this compartment to other compartments.

The large difference between the two model outputs can be explained by the difference in environmental properties between the two model worlds.

It may seem surprising that many chemicals with relatively low log K_{OW} -values is predicted to partition more strongly to sediments in CeStoc than in EQC. One reason for this is likely to be the long residence time in water (14 days) in the Stockholm waters, relative to its volume ($1.8 \times 10^8 \text{ m}^3$), which can be compared to the EQC residence time of 500 days with a water volume of $2 \times 10^{11} \text{ m}^3$. In consequence, the EQC water outflow is about 40 times as fast as the CeStoc outflow rate. Thus, chemicals that are released to the smaller water volume of the CeStoc environment will have longer time to deposit to sediments. In addition, the sediment burial rate is about five times higher in the CeStoc environment, than in the EQC environment, which is another contributing factor.

Furthermore, central Stockholm is covered to about 25 % by water, which consequently increases the relative sediment volume in the Stockholm model, and 50 % is impervious surfaces that are regularly washed off with rainfall. Chemicals deposited here are likely to ultimately end up in the water/sediment compartment. Soil only contributes by 25 % to the total surface area, or by 0.002 % to the total model volume, whereas sediment accounts for 0.07 % of the model volume. It thus seems reasonable, that a larger proportion of the chemicals with high log K_{OW} -values, which are released to the water compartment in central Stockholm will partition to sediment, than the proportion that

will volatilise and partition to soils. In the “EQC world”, only 10 % of the surface area is water and the remaining surface area is soil, which is all available for chemical deposition and sorption to soil solids, from where transport to water is low or negligible.

Table 7. Ranking of chemicals with respect to their tendency to partition to sediments. Results are obtained with two different models: EQC and CeStoc. See text for a fuller explanation. The following chemicals could not be modelled: metals, PCP, atrazine, simazine and chloroalkanes.

CAS-number	Name	% in sediment (EQC level III)	Relative sed distribution	Rank EQC	%in sediment (CeStoc)	Relative sed distribution	Rank CeStoc
	BDE 99	97.4	1.00	1	92.1	1.000	1
	BDE 100	97.4	1.00	1	92.1	1.000	1
	BDE 47	96.0	0.986	3	92.1	1.000	3
193-39-5	Ind	95.7	0.983	4	91.9	0.997	9
191-24-2	B(g,h,i)Per	95.5	0.980	5	91.9	0.997	10
50-32-8	BaP	93.2	0.957	6	91.9	0.997	11
207-08-9	BkF	93.0	0.955	7	91.8	0.997	12
118-74-1	HCB	92.2	0.947	8	92.1	1.000	4
117-81-7	DEHP	89.6	0.920	9	90.5	0.983	18
205-99-2	B(b)F	88.8	0.912	10	91.8	0.997	13
1582-09-8	Trifluralin	86.6	0.889	11	92.1	0.999	5
608-93-5	Pentachlorobenzene	80.1	0.822	12	92.0	0.999	6
2921-88-2	Chloropyriphos	74.7	0.767	13	92.0	0.998	7
206-44-0	Fluoranthene	71.6	0.735	14	91.7	0.996	14
104-40-5	4-nonylphenol	71.1	0.730	15	90.5	0.982	19
140-66-9	4-tert-octylphenol	69.1	0.709	16	91.5	0.993	15
87-68-3	Hexachlorobutadiene	61.4	0.630	17	91.9	0.997	8
120-12-7	Anthracene	34.7	0.356	18	91.3	0.991	16
12002-48-1	Trichlorobenzene	19.7	0.202	19	90.8	0.986	17
115-29-7	Endosulfan	17.0	0.175	20	89.9	0.975	21
959-98-8	(alpha-endosulfan)	16.9	0.174	21	89.9	0.975	22
120-82-1	(1,2,4-trichlorobenzene)	14.3	0.147	22	90.3	0.980	20
58-89-9	(gamma-HCH, lindan)	13.3	0.137	23	89.2	0.968	24
470-90-6	Chlorfenvinphos	11.1	0.114	24	89.4	0.970	23
15972-60-8	Alachlor	6.08	0.062	25	87.2	0.946	25
91-20-3	Naphthalene	2.99	0.031	26	83.9	0.911	26
34123-59-6	Isoproturon	1.35	0.014	27	73.5	0.797	27
330-54-1	Diuron	0.96	0.010	28	66.3	0.719	28
71-43-2	Benzene	0.35	0.004	29	38.9	0.422	29
67-66-3	Trichlorometan	0.28	0.003	30	30.9	0.336	30
107-06-2	1,2-Dichloroetane	0.20	0.002	31	13.1	0.142	31
75-09-2	Dichloromethane	0.18	0.002	32	8.2	0.089	32

Judging from the CeStoc outputs, PBDEs are the chemicals, which are most likely to end up in the sediment compartment. However, the majority of the remaining chemicals (all the way down to alachlor) are predicted to partition here by nearly 90 %. Again, this assessment is oversimplified and must not be interpreted quantitatively. Naphthalene, isoproturon, diuron, benzene, chloroform, 1,2-dichloroethane and dichloromethane are less likely to find in sediments.

It should be noted, however, that the adopted emission scenario has significant implications for the relative distribution in the environment. The most volatile chemicals are probably unlikely to be released to the water compartment, thus their actual partitioning percentage to sediment will be significantly lower in reality. In this exercise, the aim was to highlight the general tendency for various chemicals to partition to sediment, providing the same emission scenario. The exact proportions emitted to the different compartments for each individual chemical are not known, why such a detailed analysis was not possible.

These are the major conclusions of the modelling:

- PBDEs are the chemicals partitioning most strongly to sediments in Stockholm, but also heavier PAHs, DEHP, alkylphenols and certain pesticides partition very strongly.
- Naphthalene, isoproturon, diuron, benzene, chloroform, 1,2-dichloroethane and dichloromethane are least likely to find in sediments, in agreement with the experimental results.
- Stockholm sediments capture large percentage of chemicals due to the long residence time (relative to volume) and high sedimentation rate.
- Environmental properties may be more important than expected for the partitioning properties of a chemical. Thus, site-specific models may be necessary also for ranking purposes, especially if physical-chemical properties are well known!

5.5 Risk for biological effects

5.5.1 Organic substances

The pollutants present in surface sediments may be assimilated by sediment dwelling organisms. They may also be taken up by aquatic organisms, which feed on sediment organisms or by uptake through resuspension of sediment material. If concentrations and bioavailability are sufficiently high, negative effects on individual species can occur. The technical guidance document for risk assessment in the European Union,

states that in order to realistically assess the risk for the sediment compartment, whole-sediment tests are required to derive a “predicted no effect concentration” ($PNEC_{sed}$). Such values can then be compared to measured or estimated environmental concentrations. Data from such tests are rarely available and an alternative approach is suggested, which employs the equilibrium partitioning method. If this alternative method indicates a risk, then it would motivate the performance of more detailed whole-sediment tests (TGD, 2003).

Using the equilibrium partitioning method, $PNEC_{sed}$ -values for the sediment compartment are derived from $PNEC_{water}$ for aquatic organisms and the sediment-water partition coefficient. In this approach it is assumed that: i) benthic organisms are equally sensitive to the chemical as are aquatic species; ii) the concentration of the chemical in sediment, interstitial water and benthic organisms are at thermodynamic equilibrium; iii) the toxicity in the sediment compartment is closely related to the pore water concentration and iv) the partitioning between sediment and water can be described by a partition coefficient measured or estimated from properties of the chemical and the sediment itself. For chemicals of $\log K_{OW} > 5$ it is suggested that a correction factor is introduced, allowing for uptake through sediment ingestion (TGD, 2003). This correction has been applied to the PNEC values where appropriate.

As part of the implementation of the WFD, Water Quality Standards are to be suggested by the European Commission. Personal information from the expert groups says that these will not be made public prior to the end of 2003, and a draft version indicates that such standards will primarily cover water in the first version. Preliminary PNEC-values for the sediment compartment have, however, previously been compiled (Klein et al. 1999), all derived using the equilibrium partitioning method, adopting sediment-water partition coefficients estimated from K_{OW} and allowing for sediment ingestion of chemicals with a $\log K_{OW} > 5$. Consequently, the calculated PNEC-values suffer from the uncertainties described above, and the results should be regarded as screening-level risk ratios rather than absolute measures of risk.

There is an ongoing debate about the sorption mechanism of PAHs, and it has been suggested that the equation currently used for calculating equilibrium concentrations in sediments may be inaccurate (see e.g. Bucheli & Gustafsson, 2000). These studies suggest that PAHs bind strongly to soot carbon in the sediment, and that current models underestimate the partition coefficient typically by a factor of 100-1000. This would significantly reduce their bioavailability in soot-rich sediments, and thus emphasises the importance of the sediment properties associated with bioavailability and risk for effects of PAHs. A recent study by Ghosh et al. (2003) suggests that not only PAH but also PCBs have potential for adsorption to soot-carbon. If true, also PCBs will experience reduced bioavailability in soot-rich material.

Despite the uncertainties discussed above, risk is regarded as a more accurate parameter to use when prioritising the monitoring data, than mere concentrations. Derived risk ratios should provide useful information about the current quality of the aquatic system with respect to the WFD-chemicals and indicate which chemicals that might be of concern and which pose no significant problem. They should not be interpreted as absolute risks, but rather do they indicate a *relative* risk ranking of the chemicals involved. We have mainly used the PNEC-values suggested by Klein et al. (1999). Because PNEC values can be derived using different methods, it is a great advantage to mainly use one compilation. For DEHP, a value of 100 mg/kg dw was used. In the EC draft risk assessment of DEHP, however, it was concluded that a PNEC in the aquatic compartment could not be derived because values were higher than solubility (KemI, 2001a). For certain pesticides and TBT, where PNEC-values were lacking, we used “maximal permissible concentrations” (MPC) as suggested by Crommentuijn et al. (1997), also adopting the equilibrium partitioning method. For C₁₀₋₁₃ chloroalkanes, the PNEC value was taken from ECB (2000).

The risk was calculated for the median concentration of a chemical in each geographical group, by calculating the ratio PEC/PNEC alternatively PEC/MPC, where PEC (predicted environmental concentration) here indicates the measured concentration. In this way, it could be determined whether a chemical was *generally* above or below the proposed risk level. Table 8 shows a compilation of the outcome of the risk assessment. We have split the risk ratios into five subgroups, where group A indicates a ratio > 100, group B indicates a ratio between 10 and 100, group C between 1 and 10 and group D a ratio < 1, i.e. no risk. Group E denotes substances for which the risk could not be assessed, due to missing PNEC (MPC data) or a higher limit of detection (LOD) than the PNEC (MPC) value. The majority of the chemicals in risk group E were below the detection limits.

As shown in Table 8, chlorfenvinphos and PAHs of 4 to 6 rings are predicted to pose the greatest risks to the aquatic ecosystem, in all geographical groups. This is interesting, since chlorfenvinphos has, to our knowledge, not previously been found in sediments. As discussed above, the risk associated with PAHs may be overestimated. However, the calculated risk ratios for the PAHs in group A are generally > 1000 in lakes and central Stockholm, and would thus pose a risk even if the bioavailability is overestimated by as much as a factor of 1000.

The alkylphenols (4-nonylphenol and 4-tert-octylphenol) show median risk ratios between 10 and 100 in the lakes and in central Stockholm, whereas they are only slightly above 1 at the coast. These compounds are highly toxic and disturb the endocrine system, they are not readily degradable in sediments, and our data show that they are widespread in the aquatic environment. Emission pathways are not sufficiently known. Alkylphenols are thus considered to be chemicals of high concern to this region.

Table 8. Relative risk ranking using the median concentrations in each geographical group: Risk group A:>100 B: 10-100 C: 1-10 D: <1, E: Risk could not be calculated (e.g. LOD lower than PNEC).

Risk group	Lakes	central Stockholm	Coast
A	Chlorfenvinphos most 4-6 ringed PAHs	Chlorfenvinphos p,p-DDD most 4-6 ringed PAHs	Chlorfenvinphos most 4-6 ringed PAHs
B	alkylphenols PCB ## 28, 52 p,p-DDT/DDE/DDD 3-ringed PAHs	alkylphenols TBT p,p-DDT/DDE most PCBs 3-ringed PAHs	TBT p,p-DDD Some 3-4-ringed PAHs
C	TBT PCP most PCBs 2-3-ringed PAHs	PCP C ₁₀₋₁₃ chloroalkanes PCB ## 153, 180 2-3-ringed PAHs	PCP alkylphenols p,p-DDT/DDE some PCBs
D	Chlorobenzenes PBDE ¹ C ₁₀₋₁₃ chloroalkanes DEHP PCB ## 101, 180 acenaphthene and chrysene HCHs	Chlorobenzenes PBDE ¹ DEHP PCB 101 acenaphthene HCHs	Chlorobenzenes PBDE ¹ C ₁₀₋₁₃ chloroalkanes DEHP some PCBs 2-3 ringed PAHs and chrysene HCHs
E	Most pesticides, HCBd, Chlorinated solvents, Benzene, chlordanes	Most pesticides, HCBd, Chlorinated solvents, Benzene, chlordanes	Most pesticides, HCBd, Chlorinated solvents, Benzene, chlordanes

1. PBDEs are known to biomagnify, which is not considered in this assessment. As explained below, PBDEs do probably pose a larger risk than this table indicate.

DEHP achieves a low rank, due to the apparently low toxicity of this chemical to aquatic organisms. Nevertheless, DEHP is considered as toxic to humans and our data indicate that DEHP releases are significant in the Stockholm region.

In this assessment, TBT-Sn has been assigned to risk group B, using MPC values suggested by Crommentuijn et al. (1997). However, no well-established PNEC value

exists for this compound and adopting suggested values by OSPAR (see Jansson, 2000), would place this compound in risk group A.

The PBDEs do not pose a risk in the aquatic environment, according to this assessment (group D). These chemicals, together with similar substances like DDTs and PCBs, are likely to biomagnify in the aquatic food web (e.g., Boon et al., 2002). Effects of such substances commonly manifest themselves in the top of the food web, an effect that is not considered in the risk assessment methodology. Thus, we still consider PBDE to be a high-risk substance group.

Simazine and α -endosulfan were not included in the risk ranking, since the results were not verified with GC-MS. The preliminary data, however, give risk ratios > 1 at most stations where concentrations were above detection level.

It cannot be assessed whether benzene and the chlorinated solvents pose a risk to the aquatic environment, since they do not partition sufficiently strongly to sediments (chapter 5.4). However, it is apparent that they do not pose a risk to sedimentary life.

In addition, certain “old sins” are worth mentioning. Pentachlorophenol (PCP) and PCBs, which have been banned or severely restricted for a long time, still stay highly ranked in this relative risk assessment. Consequently, measures to reduce the pollution of chemicals such as these are likely to be difficult to initiate on a local and regional scale. It clearly exemplifies that persistent chemicals may pose a risk to the environment for a long time after reduction measures have been introduced.

5.5.2 Metals

This risk assessment methodology relies on the assumption that concentrations in biota are linearly related to exposure. It has been demonstrated the bioaccumulation of many metals in aquatic species is not linearly related to exposure, but rather shows a weak response to increasing exposure. Actually, the bioaccumulation factor decreases when exposure increases (McGeer et al., 2003). Presumably, this reflects the capability of many organisms to control their internal metal concentrations within a limited range, and also the very complex and dynamic aquatic chemistry that relates metal levels to bioavailability and effects (e.g., Sternbeck, 2000a, and references therein). This is not to say that metals cannot give rise to toxic effects in aquatic biota, but that it is difficult to predict such effects from exposure concentrations.

The relation between toxicity and metal concentrations in sediments is even weaker, because the interactions between all soluble (\approx bioavailable) metal species and the variety of solid phases occurring in sediments are not described by a single partition coefficient. Klein et al. (1999) also pointed this out. Considering these facts, it is

probably better to classify metal concentrations in relation to the local background values or to preindustrial values. To assess whether the metals present are likely to cause biological effects, another research strategy is required. The choice of representative preindustrial values for a certain region is not straight-forward and were previously discussed for Stockholm (Sternbeck, 2000b). Those recommendations were followed for the lakes and central Stockholm. For the coastal samples, preindustrial values for the Baltic Sea were taken from Anon. (1999). Enrichments were calculated as the ratio of the median concentrations and the preindustrial levels.

The relative enrichment in the three geographical groups are shown in Figure 36. The coastal stations show fairly low impact with As, Cr and Ni being almost identical to the preindustrial levels. Although Cd, Cu, Hg, Pb and Zn are slightly elevated in the coastal zone, the levels are actually lower than in surface sediments from the open Baltic proper sampled during the early 1990s (Borg and Jonsson, 1996). The lakes and central Stockholm show a pronounced enrichment of Cd, Cu, Hg, Pb and Zn, but not of As, Cr or Ni. This pattern agrees with previous studies in the Stockholm region (e.g., Sternbeck and Östlund, 2001). Only Hg is significantly more enriched in the urban area (central Stockholm) compared to the lakes. The enrichment of Cd is not typical for the urban/suburban region, because the background level of Cd in S Sweden is 1.4 mg/kg dw, fairly close to the observed levels.

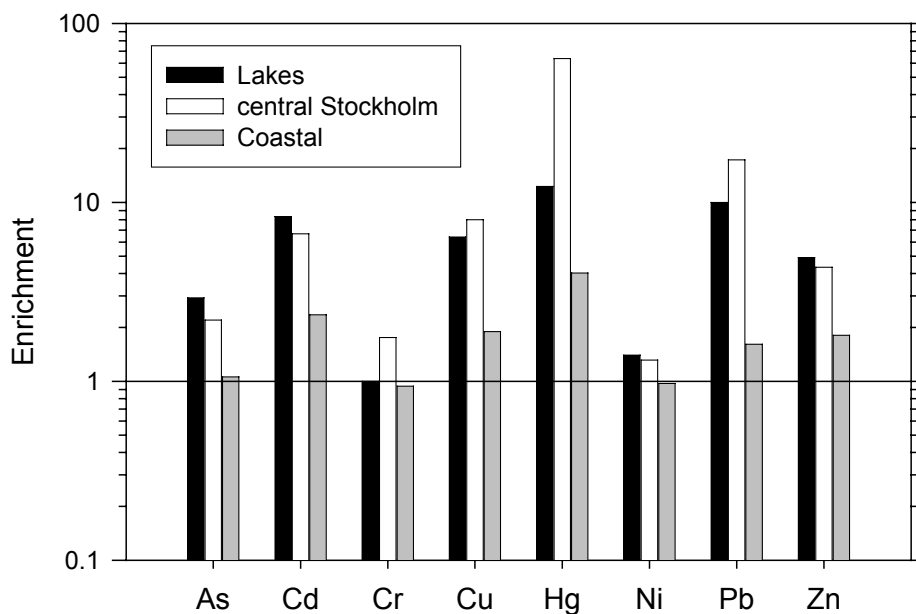


Figure 36. Enrichment of metals in the three geographical groups. The enrichment factors are calculated as the ratio between median concentrations and the estimated preindustrial levels.

6 Conclusions

A number of observations and conclusions can be drawn from this vast dataset, as emphasized in the preceding chapters. We would like to stress the following points:

- ✓ Of the chemicals investigated, most chemicals that are known to be used or unintentionally emitted in Sweden were found.
- ✓ Several chemicals that were investigated are banned in Sweden. Of these, mainly those known to be persistent or to undergo long-range atmospheric transport were found.
- ✓ Many chemicals are apparently released in the urban and suburban region. However, only the innermost coastal region appears to be influenced by these releases.
- ✓ Except for a few stations, concentrations of e.g. PAHs, DEHP, PBDEs and PCBs are fairly homogenous in the coastal region.
- ✓ Södra Kanholmsfjärden deviates from other coastal stations, being strongly enriched in PCBs, DDTs, C₁₀₋₁₃ chloroalkanes, PCP, α -HCH, γ -HCH, and PAHs.
- ✓ Alkylphenols are of high concern. The concentrations are fairly high and strongly irregular, indicating strong local influence. Concentrations are generally highest in the lakes. Furthermore, both nonylphenol and octylphenol obtain high risk ratios.
- ✓ There are indications that Cd, Cu, Hg, Pb and Zn have decreased slightly over the last five years in central Stockholm and the lakes. This is in agreement with general long-term trends in the region.
- ✓ The concentrations of several chemicals span over wide ranges in the lakes. This is interpreted as strong influence of local sources in the catchments.
- ✓ Whether benzene and the chlorinated solvents pose a risk to the aquatic environment cannot be assessed, because these chemicals do not partition sufficiently strongly to sediments.
- ✓ We suggest that future studies focus on chemicals of high concern, generally following the results of the risk ranking. Future studies may include 1) establishing time trends to assess whether these problems are likely to increase and in order to relate pollution trends to changes in the use pattern of chemicals; 2) identification of the emissions sources through material flow analyses and detailed multimedia monitoring programmes; 3) investigate other urban areas, to assess whether the conclusions are generally applicable in Sweden.

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Appendix – analytical data

Metals. Concentrations are presented in mg/kg dw

Sampling station	As	Cd	Co	Cr	Cu	Hg	Mn	Ni	Pb	Zn	DW (%)	LOI %
Lakes												
Brunnsviken	10	7	16	110	230	1.9	430	52	220	890	4.0	25
Drevviken	9.5	1	22	54	81	0.23	2400	52	78	400	5.8	22
Flaten	12	1	9	35	72	0.15	1100	31	44	710	0.77	64
Judarn	4.7	2	15	45	66	0.28	1360	37	160	310	3.6	42
Kyrksjön	6.2	1	10	35	42	0.25	560	30	72	280	1.8	46
Laduviken	8.8	1	13	42	160	0.27	550	38	100	440	3.2	42
Lillsjön	5.2	5	19	83	180	0.36	530	63	200	630	7.0	23
Långsjön	9.1	2	15	42	420	0.31	900	52	110	590	2.5	53
Magelungen	6.7	<1	21	50	82	0.15	1500	50	67	320	5.7	21
Räcksta	7.4	3	21	78	660	0.23	540	43	260	890	7.7	22
Sicklasjön	8.8	4	18	110	310	1.4	710	49	310	1400	4.8	36
Trekanten	16	4	18	89	620	2	590	50	430	1800	5.0	31
Ältasjön	6.2	1	13	44	53	0.25	1200	34	69	360	4.1	34
Central Stockholm												
Essingen	5.4	1	15	77	120	0.55	760	47	98	320	10	14
Fjärderholmarna	8.6	1	18	83	140	1.6	490	33	160	310	6.4	17
Reimersholme	6.6	1	17	87	160	1.2	920	46	190	480	9.6	14
Riddarfjärden	6.5	2	17	98	200	1.4	810	52	270	520	10	14
Strömmen	10	3	19	88	300	3.2	470	34	300	620	5.5	21
Ulvsundasjön	5.4	2	18	110	220	1.1	640	45	170	570	11	14
Årstaviken	6.7	3	20	110	290	2.5	530	47	250	900	9.8	14

Metals, continued.

Sampling station	As	Cd	Co	Cr	Cu	Hg	Mn	Ni	Pb	Zn	DW (%)	LOI %
Coast												
Bråviken	11	0.36	19	85	48	0.21	4600	41	50	170	13	13
Gälnan	6.9	0.53	15	74	<6	0.093	500	<12	50	150	7.5	17
Mysingen	8.3	0.46	12	73	7.4	0.13	780	32	40	150	22	19
Nyköping											19	
Nämndöfjärden	16	0.76	18	68	45	0.15	4000	36	54	180	13	18
Saxarfjärden	9.5	0.64	12	75	23	0.18	1000	24	41	150	11	22
Singö	13	0.47	15.6	104	26	0.16	1300	<10	64	175	4.8	
Svärdsfjärden	8.3	0.36	18	86	31	0.16	2300	36	44	170	18	12
Söderarm	16	0.40	21	100	<6	0.15	1700	33	60	150	17	160
Södra Kanholmsfjärden	17	0.59	8.8	75	19	0.23	3500	15	54	150	20	24
Tvären	11	0.38	12	82	<6	0.085	670	<10	40	140	8.8	19
Öregrundsgrepen	15	0.65	12	91	51	0.18	610	35	48	160	19	
Östra Landsort	5.2	0.2	7.6	63	24	0.07	450	21	29	77	43	6.5
Östra Askrikefjärden	7.9	1.3	21	60	48	1.1	650	30	87	260	7.0	
Sewage sludge												
Bromma	1.5	1	7	40	380	1.4	140	23	26	550	29	
Henriksdal	1.7	1	8	31	360	2.1	130	27	28	540	29	

Concentrations of PAH (µg/g dw)

Sampling station	Naphthalene	Acenaphthene	Fluorene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Benzo(a)anthracene	Chrysene	Benzo(b)fluoranthene	Benzo(k)fluoranthene	Benzo(a)pyrene	Dibenz(ah)anthracene	Benzo(g,h,i)perylene	Indeno(1,2,3-cd)pyrene	Sum (16) PAH
Lakes																
Brunnsviken	0.053	0.011	0.021	0.12	0.038	0.40	0.39	0.25	0.20	0.50	0.23	0.38	0.089	0.57	0.43	3.7
Drevviken	0.040	0.004	0.012	0.077	0.017	0.20	0.18	0.088	0.099	0.27	0.11	0.14	0.042	0.27	0.23	1.8
Flaten	0.15	<0.001	0.022	0.088	0.012	0.14	0.19	0.088	0.14	0.33	0.13	0.17	0.070	0.47	0.38	2.4
Judarn	0.039	0.028	0.043	0.30	0.058	1.1	0.79	0.43	0.49	1.0	0.50	0.68	0.17	1.0	0.81	7.5
Kyrksjön	0.076	<0.001	0.036	0.086	0.017	0.30	0.16	0.085	0.10	0.22	0.10	0.11	0.031	0.21	0.18	1.7
Laduviken	0.078	0.018	0.023	0.16	0.031	0.41	0.36	0.16	0.19	0.33	0.15	0.24	0.049	0.33	0.28	2.8
Lillsjön	0.096	0.008	0.037	0.22	0.041	0.52	0.61	0.21	0.35	0.49	0.21	0.33	0.10	0.63	0.36	4.2
Långsjön	<0.002	0.020	0.032	0.23	0.031	0.62	0.52	0.24	0.30	0.45	0.22	0.34	0.075	0.48	0.33	3.9
Magelungen	0.036	0.007	0.013	0.056	0.010	0.16	0.14	0.068	0.084	0.15	0.064	0.099	0.027	0.17	0.16	1.2
Räcksta	0.042	0.013	0.042	0.18	0.048	0.73	1.0	0.27	0.54	0.72	0.28	0.44	0.18	1.0	0.45	6.0
Sicklasjön	0.034	0.006	0.018	0.12	0.025	0.35	0.35	0.20	0.23	0.46	0.22	0.32	0.077	0.43	0.36	3.2
Trekanten	0.12	0.031	0.086	0.69	0.14	2.1	2.0	0.95	0.93	1.6	0.77	1.3	0.27	1.57	1.2	14
Åltasjön	0.039	0.009	0.020	0.062	0.010	0.18	0.029	0.065	0.082	0.16	0.070	0.098	0.030	0.21	0.22	1.3
Central Stockholm																
Essingen	0.041	0.008	0.017	0.13	0.030	0.35	0.32	0.18	0.18	0.29	0.13	0.23	0.049	0.32	0.25	2.5
Fjäderholmarna	0.11	0.034	0.054	0.31	0.095	0.64	0.54	0.33	0.31	0.52	0.25	0.47	0.10	0.60	0.45	4.8
Reimersholme	0.067	0.029	0.053	0.39	0.095	0.81	0.70	0.40	0.39	0.56	0.28	0.51	0.10	0.62	0.46	5.5
Riddarfjärden	0.094	0.073	0.10	0.79	0.21	1.9	1.6	0.99	0.92	1.2	0.60	1.2	0.22	1.2	0.89	12

Sampling station	Naphthalene	Ace-naph-thene	Fluorene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Benzo(a)anthracene	Chrysene	Benzo(b)fluoranthene	Benzo(k)fluoranthene	Benzo(a)pyrene	Dibenz(ah)anthracene	Benzo(g,h,i)perylene	Indeno(1,2,3-cd)pyrene	Sum (16) PAH
Strömmen	0.18	0.097	0.16	1.2	0.29	2.2	1.8	1.1	1.1	1.4	0.68	1.3	0.23	1.3	0.95	14
Ulvsundasjön	0.048	0.015	0.027	0.23	0.049	0.48	0.56	0.25	0.25	0.47	0.21	0.38	0.12	0.73	0.42	4.2
Årstaviken	0.065	0.036	0.061	0.37	0.10	0.87	0.90	0.52	0.47	0.80	0.40	0.76	0.16	0.89	0.70	7.1
Coast																
Bråviken	<0.004	0.006	0.006	0.031	0.006	0.083	0.060	0.047	0.067	0.096	0.045	0.061	0.015	0.092	0.092	0.71
Gälnan	0.022	<0.001	<0.001	0.019	0.004	0.067	0.043	0.026	0.035	0.098	0.042	0.046	0.015	0.10	0.13	0.65
Mysingen	0.005	0.002	0.002	0.013	0.002	0.043	0.027	0.018	0.022	0.064	0.028	0.029	0.010	0.070	0.076	0.41
Nyköping	<0.004	0.004	0.010	0.029	0.008	0.081	0.068	0.037	0.031	0.072	0.031	0.047	0.010	0.054	0.049	0.53
Nämndöfjärden	<0.004	0.003	0.006	0.023	0.005	0.066	0.047	0.037	0.052	0.13	0.055	0.051	0.017	0.12	0.14	0.75
Saxarfjärden	0.018	0.003	0.003	0.019	0.004	0.058	0.046	0.027	0.034	0.069	0.031	0.039	0.009	0.068	0.072	0.50
Singö	<0.004	<0.001	<0.001	0.018	0.003	0.056	0.035	0.019	0.029	0.063	0.027	0.031	0.008	0.059	0.073	0.42
Svärdsfjärden	0.009	0.002	0.003	0.010	0.002	0.029	0.019	0.015	0.018	0.055	0.024	0.024	0.009	0.060	0.060	0.34
Söderarm	0.010	0.002	0.003	0.020	0.003	0.047	0.033	0.024	0.033	0.080	0.034	0.030	0.011	0.075	0.085	0.49
södra Kanholmsfjärden	0.051	0.002	0.008	0.061	0.020	0.21	0.20	0.18	0.18	0.64	0.20	0.33	0.12	0.33	0.32	2.9
Tvären	0.020	<0.001	<0.001	0.026	0.005	0.073	0.056	0.036	0.047	0.11	0.052	0.066	0.018	0.12	0.15	0.78
Öregrundsgrepen	0.010	0.003	0.003	0.021	0.003	0.066	0.039	0.024	0.038	0.068	0.027	0.029	0.008	0.052	0.055	0.45
öst Landsort	0.005	0.002	0.002	0.009	0.002	0.022	0.015	0.012	0.013	0.031	0.014	0.017	0.005	0.031	0.035	0.21
östra Askrikefjärden	0.044	0.011	0.023	0.12	0.032	0.27	0.23	0.14	0.14	0.24	0.12	0.20	0.043	0.27	0.23	2.1
Sewage sludge																
Bromma	0.060	0.021	0.060	0.39	0.058	0.55	0.62	0.21	0.28	0.25	0.11	0.20	0.061	0.33	0.16	3.4
Henriksdal	0.039	0.036	0.45	0.59	0.064	0.63	0.73	0.21	0.27	0.28	0.13	0.25	0.059	0.34	0.19	4.3

Benzene, chlorobenzenes (CBz), and chlorinated solvents ng/g dw. Data for Söderarm are missing for some compounds, due to analytical problems

Sampling station	Benzene	1,2-CBz	1,3-CBz	1,4-CBz	1,2,3-CBz	1,2,4-CBz	1,3,5-CBz	Penta-CBz	HCB	Chloroform	1,2-dichloroethane	Dichloromethane
Lakes												
Brunnsviken	<5200	<2	<2	<3	<3	5	<2	<3	<4	<9	<890	<17000
Drevviken	<3100	<3	<2	<4	<4	<3	<3	<2	<4	<5	<520	<10000
Flaten	<17000	<7	<6	<9	<10	<8	<8	<10	<13	<28	<2800	<57000
Judarn	<7500	<2	<2	<2	<3	<2	<2	<3	<4	<13	<1300	<25000
Kyrksjön	<9800	<3	<2	<4	<4	<3	<3	<4	<6	<16	<1600	<33000
Laduviken	<14500	<4	<4	<6	<6	<5	<5	<6	<9	<24	<2400	<48000
Lillsjön	<3400	<1	<1	<2	<2	<1	<1	6	<3	<6	<570	<11000
Långsjön	<4400	<5	<4	<7	<7	<5	<5	<6	<10	<7	<730	<15000
Magelungen	<3400	<2	<2	<3	<3	<2	<2	<2	<3	<6	<570	<11000
Räcksta	<3700	<2	<2	<3	<3	<3	<2	<3	<3	<6	<610	<12000
Sicklasjön	<1800	<1	<1	<1	<1	<1	<1	<1	<1	<3	<300	<6000
Trekanten	<7000	16	<2	151	<2	6	<2	<2	<3	<12	<1200	<23000
Åltasjön	<4800	<5	<4	<7	<6	<5	<5	<5	<7	<8	<790	<16000
Central Stockholm												
Essingen	<1500	<1	<1	<1	<1	<1	<1	<1	<1	<3	<250	<5100
Fjäderholmarna	<4400	<4	<3	<5	<5	<4	<4	<4	<7	<7	<730	<15000
Reimersholme	<2100	<1	<1	26	<1	3	<1	<1	<2	<4	<350	<7000

Central Stockholm	Benzene	1,2-CBz	1,3-CBz	1,4-CBz	1,2,3-CBz	1,2,4-CBz	1,3,5-CBz	Penta-CBz	HCB	Chloroform	1,2-dichloroethane	Dichloromethane
Riddarfjärden	<2200	<1	<1	12	<1	6	<1	3	3	<4	<360	<7200
Strömmen	<4700	<2	<1	<2	<2	<2	<2	<2	<3	<8	<770	<15000
Ulvsundasjön	<2600	<1	<1	<1	<1	6	<1	4	<2	<4	<430	<8500
Årstaviken	<2600	<1	<1	24	<1	<1	<1	<1	<3	<4	<430	<8500
Coast												
Bråviken	<1800	<0.3	<0.3	0.89	<0.7	<0.4	<0.4	<0.4	<0.7	<3	<300	<6000
Gälnan	<4400	<1	<0.7	<1	<2	<1	<1	<1	<1	<7	<730	<15000
Mysingen	<1400	<0.3	<0.2	0.92	<0.5	<0.3	<0.3	<0.3	<0.5	<2	<230	<4600
Nyköping	<1000	<0.2	<0.2	1.3	<0.5	<0.3	<0.3	<0.3	<0.5	<2	<170	<3500
Nämndöfjärden	<2000	<0.2	<0.2	1.2	<0.5	<0.3	<0.3	<0.2	<0.3	<3	<340	<6800
Saxarfjärden	<2100	<0.5	<0.4	<0.7	<1.0	<0.6	<0.6	<0.6	<1	<4	<350	<6900
Singö	<4500	<0.7	<0.6	<1	<1.7	<1.0	<1.0	<1	<1	<8	<760	<15000
Svärdsfjärden	<1300	<0.2	<0.2	0.72	<0.5	<0.3	<0.3	<0.2	<0.4	<2	<210	<4200
Söderarm	<1500	-	-	-	-	-	-	-	-	<2	<240	<4900
Södra Kanholmsfjärden	<1100	<0.2	<0.2	1.2	<0.5	<0.3	<0.3	<0.3	<0.4	<2	<190	<3700
Tvären	<3200	<0.4	<0.4	<0.7	<1	<0.5	<0.6	<0.6	<1	<5	<540	<11000
Öregrundsgrepen	<1200	<0.2	0.2	0.68	<0.5	<0.3	<0.3	<0.2	<0.4	<2	<200	<3900
öst Landsort	<570	<0.1	<0.1	0.28	<0.2	<0.1	<0.1	<0.2	<0.2	<1	<100	<1900
Östra Askrikefjärden	<1800	<1	<0.8	2.2	<2	<1	<1	<1	<1	<3	<310	<6200
Sewage sludge												
Bromma	<1900	<10	<10	<10	<10	<10	<10	<10	<10	<3	<300	<6200
Henriksdal	<1700	<10	<10	<10	<10	<10	<10	<10	<10	<3	<300	<5700

Pesticides, concentrations are presented in ng/g dw. Results for simazine and endosulfan have not been verified with GC-MS, and should be regarded as preliminary.

Sampling station	Isoproturon	Diuron	Trifluralin	Simazine	Atrazine	Alachlor	Chlorpyrifos	Chlorfenvinphos	PCP	α -endosulfan	β -endosulfan
Lakes											
Brunnsviken	<9	<7	<3	<1	<1	<9	<1	236	5.5	0.89	<0,04
Dreviken	<9	<7	<3	<1	<1	<9	<1	<6	1.5	<0,04	<0,04
Flaten	<14	<16	<3	<1	<1	<10	<1	<7	3.5	<0,04	<0,04
Judarn	<9	<7	<3	<1	<1	<9	<1	15	1.7	<0,04	<0,04
Kyrksjön	<14	<16	<3	<1	<1	<10	<1	<7	5.1	<0,04	<0,04
Laduviken	<14	<16	<3	<1	<1	<10	<1	82	6.4	<0,04	<0,04
Lillsjön	<14	<16	<3	<1	<1	<10	<1	11	5.3	<0,04	<0,04
Långsjön	<14	<16	<3	<1	<1	<10	<1	<7	4.7	<0,04	<0,04
Magelungen	<9	<7	<3	<1	<1	<9	<1	<6	1.8	<0,04	<0,04
Räcksta	<14	<16	<3	<1	<1	<10	<1	<7	<0.5	<0,04	<0,04
Sicklasjön	<9	<7	<3	<1	<1	<9	<1	<6	7.8	<0,04	<0,04
Trekanten	<14	<16	<3	<1	<1	<10	<1	<7	5.8	<0,04	<0,04
Ältasjön	<14	<16	<3	<1	<1	<10	<1	<7	2.5	<0,04	<0,04
Central Stockholm											
Essingen	<9	<7	<3	<1	<1	<9	<1	50	<0.5	<0,04	<0,04
Fjäderholmarna	<9	<7	<3	<1	<1	<9	<1	182	1.2	<0,04	<0,04
Reimersholme	<14	<16	<3	<1	<1	<10	<1	93	7.8	<0,04	<0,04
Riddarfjärden	<9	<7	<3	<1	<1	<9	<1	11	2.9	<0,04	<0,04

Sampling station	Isoproturon	Diuron	Trifluralin	Simazine	Atrazine	Alachlor	Chlorpyrifos	Chlorfenvinphos	PCP	α -endosulfan	β -endosulfan
Strömmen	<14	<16	<3	<1	<1	<10	<1	<7	9.4	0.38	<0,04
Ulvsundasjön	<14	<16	<3	<1	<1	<10	<1	188	4.1	0.13	<0,04
Årstaviken	<14	<16	<3	<1	<1	<10	<1	183	3.7	<0,04	<0,04
Coast											
Bråviken	<20	<20	<6	17	<2	<10	<2	<15	<0.5	<0,04	<0,04
Gälnan	<20	<20	<6	23	<2	<10	<2	51	3.8	0.38	<0,04
Mysingen	<20	<20	<6	11	<2	<10	<2	<15	2.1	<0,04	<0,04
Nyköping	<20	<20	<6	<2	<2	<10	<2	<15	0.58	>0.16	<0,04
Nämndöfjärden	<20	<20	<6	<2	<2	<10	<2	25	0.79	<0,04	<0,04
Saxarfjärden	<20	<20	<6	<2	<2	<10	<2	36	2.4	0.13	<0,04
Singö	<20	<20	<6	27	<2	<10	<2	34	3.6	0.46	<0,04
Svärdsfjärden	<20	<20	<6	14	<2	<10	<2	<15	2.3	<0,04	<0,04
Söderarm	<20	<20	<6	15	<2	<10	<2	<15	2.1	0.33	<0,04
södra Kanholmsfjärden	<20	<20	<6	9.3	<2	<10	<2	120	10	0.07	<0,04
Tvären	<20	<20	<6	17	<2	<10	<2	32	2.1	0.33	<0,04
Öregrundsgrepen	<20	<20	<6	13	<2	<10	<2	<15	0.70	0.18	<0,04
öst Landsort	<20	<20	<6	7.1	<2	<10	<2	<15	0.40	0.14	<0,04
östra Askrikefjärden	<20	<20	<6	<2	<2	<10	<2	190	1.2	<0,04	<0,04
Sewage sludge											
Bromma	<15	<15	<6	<2	<2	<13	<1.4	<9	3.2	> 0,04	<0,04
Henriksdal	<15	<15	<6	<2	<2	<13	<1.4	<9	1.4	> 0,04	<0,04

Concentrations of chemical additives (ng/g dw). Data for Söderarm is missing for HCBd due to analytical problems.

Sampling station	BDE 47	BDE 99	BDE 100	4-nonylphenol	4-tert-octylphenol	HCBd	DEHP	TBT (ng Sn/g dw)	C10-C13- chloroalkanes
Lakes									
Brunnsviken	0.45	1.0	0.24	5300	82	<1	30000	210	300
Drewiken	0.25	0.58	<0.07	1500	36	<2	550	<7	54
Flaten	0.90	1.1	<0.07	1100	40	<5	11000	<11	99
Judam	0.11	0.18	<0.07	100	3	<1	24	<3	34
Kyrksjön	<0.07	<0.07	<0.07	67	2	<2	1600	<2	8.1
Laduviken	0.46	0.99	<0.07	2900	<2	<3	3900	<4	180
Lillsjön	0.33	0.93	0.40	2200	15	<1	130	88	85
Långsjön	0.59	1.3	0.20	1000	8	<3	1900	<4	50
Magelungen	0.31	0.24	<0.07	1400	41	<1	2700	2.7	63
Räcksta	1.1	2.7	0.69	3900	140	<2	37000	<4	2650
Sicklasjön	0.14	0.34	0.32	1800	14	<1	11000	13	140
Trekanten	0.40	0.90	0.58	3200	30	<1	20000	3.1	530
Åltasjön	0.19	0.25	<0.07	210	2.6	<3	1500	<2	37
Central Stockholm									
Essingen	1.1	1.4	0.33	12	0.3	<1	2300	15	170
Fjäderholmarna	0.62	1.1	0.25	310	3.9	<2	480	31	440
Reimersholme	0.54	1.1	0.35	170	2.0	<1	140	48	230
Riddarfjärden	0.49	1.2	0.54	210	4.4	<1	370	77	550
Strömmen	1.3	1.6	0.84	510	6.2	<1	13000	27	3300

Sampling station	BDE 47	BDE 99	BDE 100	4-nonylphenol	4-tert-octylphenol	HCBd	DEHP	TBT (ng Sn/g dw)	C10-C13- chloroalkanes
Ulvsundasjön	1.2	2.6	0.75	610	7.3	<1	1200	140	190
Årstaviken	0.55	1.2	0.76	470	4.6	<1	510	230	260
Coast									
Bråviken	0.079	0.087	<0.03	<20	<1	<0.57	120	25	<0.3
Gälnan	0.10	0.11	<0.03	<20	9.1	<1.68	160	55	13
Mysingen	0.041	0.069	<0.03	68	2.8	<0.44	140	<2	63
Nyköping	0.055	0.074	0.042	45	<1	<0.40	330	10	19
Nämndöfjärden	0.092	0.18	0.041	250	8.0	<0.38	170	16	19
Saxarfjärden	0.089	0.12	0.034	75	3.1	<0.80	280	21	39
Singö	0.068	0.12	<0.03	42	<1	<1.36	250	36	16
Svärdsfjärden	0.045	0.061	<0.03	380	2.9	<0.41	280	<2	16
Söderarm	0.055	0.071	<0.03	<20	<1	-	140	<4	10
Södra Kanholmsfjärden	0.061	0.048	0.055	140	3.5	<0.41	270	24	1000
Tvären	0.39	0.40	<0.03	50	2.4	<0.74	190	19	48
Öregrundsgrepen	0.045	0.059	0.034	31	<1	<0.38	56	20	15
öst Landsort	0.015	<0.04	<0.03	<20	<0.5	<0.18	130	<2	0.6
Östra Askrikefjärden	0.28	0.48	0.18	100	<1	<1.41	3800	92	350
Sewage sludge									
Bromma	36	46	9.8	44	0.96	<10	530	7.5	2250
Henriksdal	19	23	4.8	31	0.78	<10	1900	15	2300

Concentrations of PCB congeners (ng/g dw).

Sampling station	#28	#52	#101	#118	#153	#138	#180	Σ7PCB
Lakes								
Brunnsviken	3.1	9.5	22	19	30	33	16	130
Drevviken	0.77	1.6	4.1	3.0	7.4	8.1	4.5	29
Flaten	<0.4	1.4	3.5	2.0	5.3	5.9	3.2	21
Judarn	1.4	2.2	4.8	3.8	7.9	9.0	4.9	34
Kyrksjön	0.88	1.6	2.1	1.7	2.4	3.9	3.6	16
Laduviken	0.73	1.8	4.4	3.2	7.0	7.1	3.4	28
Lillsjön	12	20	32	23	30	32	13	160
Långsjön	1.2	3.0	7.0	5.3	11	13	7.6	48
Magelungen	0.42	0.98	3.1	2.3	5.6	6.3	3.6	22
Räcksta	2.4	4.6	10	6.7	16	17	4.5	61
Sicklasjön	1.8	5.9	16	12	18	21	9.7	84
Trekanten	3.2	8.7	18	12	23	24	12	100
Ältasjön	0.50	1.2	2.3	1.6	4.4	4.9	3.3	18
Central Stockholm								
Essingen	5.1	5.7	10	9.5	13	14	6.1	64
Fjäderholmarna	3.8	5.4	13	8.9	17	18	8.9	75
Reimersholme	4.8	6.3	14	10	17	19	9.0	81
Riddarfjärden	5.6	8.9	22	14	29	31	16	130
Strömmen	17	17	26	19	31	33	17	160
Ulvsundasjön	4.2	12	27	20	28	31	13	140
Årstaviken	19	20	27	18	32	35	18	170
Coast								
Bråviken	0.40	0.34	1.0	0.70	1.7	1.7	0.78	6.6
Gälnan	0.20	0.61	1.0	0.71	1.7	1.7	0.74	6.7
Mysingen	0.09	0.17	0.37	0.33	0.61	0.67	0.29	2.5
Nyköping	0.30	0.38	0.86	0.55	2.8	2.1	3.2	10
Nämndöfjärden	0.13	0.30	0.65	0.66	1.2	1.4	0.65	5.0
Saxarfjärden	0.22	0.50	1.2	0.89	1.9	2.0	0.85	7.6
Singö	<0.1	<0.2	0.67	0.43	1.3	1.4	0.78	4.5

Svärdsfjärden	0.06	0.15	0.37	0.35	0.81	0.78	0.32	2.8
Söderarm	0.09	0.14	0.41	0.31	0.75	0.75	0.36	2.8
södra Kanholmsfjärden	0.24	7.0	23	19	24	28	13	110
Tvären	0.22	0.45	0.98	0.81	1.6	1.7	0.70	6.5
Öregrundsgrepen	<0.03	0.09	0.22	0.20	0.57	0.61	0.44	2.1
Öst Landsort	0.03	0.07	0.12	0.08	0.19	0.18	0.09	0.77
Östra Askrikefjärden	1.4	2.0	5.7	4.1	8.8	9.9	4.9	37
Sewage sludge								
Bromma	67	35	12	7.9	13	13	5.3	150
Henriksdal	13	8.6	8.3	4.8	12	11	6.1	64

Concentrations of non-WFD pesticides (ng/g dw). Certain p,p-DDD values appear relatively high and marked with an *. These values must be regarded as preliminary since they were not verified with GC-MS. γ -chlordane could not be quantified in certain samples, and are noted with an *.

Sampling site	γ -chlordane	α -chlordane	Trans-nonachlor	p,p-DDD	p,p-DDT	p,p-DDE	α -HCH	β -HCH	γ -HCH
Lakes									
Brunnsviken	0.74	1.1	0.91	30*	0.56	12	0.09	0.74	0.31
Drevviken	0.28	0.19	0.29	4.9	0.25	5.2	0.2	1.2	0.28
Flaten	0.40	0.23	0.23	2.5	1.1	4.8	0.15	1	0.18
Judarn	0.44	0.16	0.31	1.9	<0.05	4.7	0.05	0.87	0.13
Kyrksjön	0.15	0.11	0.06	0.97	<0.05	3.5	0.07	0.72	<0.05
Laduviken	0.12	0.088	0.088	1.9	0.13	7.4	0.1	0.6	0.35
Lillsjön	*	0.86	0.30	2.4	0.27	15	<0.04	0.47	0.22
Långsjön	*	0.12	0.12	1.49	<0.05	5.5	0.37	1.4	0.41
Magelungen	*	0.091	0.091	0.95	<0.05	2.4	0.11	0.29	0.12
Råcksta	0.17	0.23	0.26	4.8	0.48	3.4	0.12	0.5	0.2
Sicklasjön	*	0.20	0.16	11	0.21	8.8	<0.04	0.36	<0.05
Trekanten	*	0.62	0.44	41*	1.6	16	0.95	11	0.59
Ältasjön	0.13	0.08	0.08	2.2	0.42	5.3	0.05	0.36	<0.05
Central Stockholm									
Essingen	*	0.20	0.16	4.2	0.072	3.4	<0.04	0.27	0.07
Fjärderholmarna	1.0	0.64	0.62	16	1.0	6.2	0.32	1.9	0.7
Reimersholme	2.0	1.1	0.92	32*	3.5	5.0	0.56	3.2	0.28

Riddarfjärden	1.9	0.63	0.57	15*	0.32	4.1	0.14	0.52	0.14
Strömmen	4.2	2.0	2.2	29*	3.5	12	0.31	1.5	0.27
Ulvsundasjön	*	0.57	0.31	3.5	<0.05	3.7	<0.04	0.38	0.08
Årstaviken	2.0	1.17	0.74	31*	0.30	4.4	0.1	0.64	0.09
Coast									
Brävikens	0.076	0.039	<0.04	1.6	0.068	1.2	0.089	0.5	0.14
Gälnan	0.093	0.13	0.12	1.1	0.16	1.3	0.23	0.81	0.44
Mysingen	0.053	0.071	0.056	0.72	0.12	0.60	0.1	0.39	0.093
Nyköping	0.051	<0.03	<0.04	0.79	<0.05	0.93	0.029	0.098	0.11
Närmdöfjärden	0.066	0.097	0.090	1.2	0.27	1.1	0.19	0.73	0.14
Saxarfjärden	0.053	0.054	0.063	1.3	0.14	0.93	0.091	0.52	0.13
Singö	<0.03	0.067	0.083	0.59	0.096	0.83	0.18	0.51	0.11
Svärdsfjärden	0.044	0.057	0.054	0.63	0.14	0.71	0.15	0.41	0.17
Söderarn	0.057	0.073	0.070	0.73	1.2	0.68	0.13	0.4	0.093
Södra Kanholmsfjärden	0.47	0.040	0.062	72*	98*	18	0.88	0.85	0.36
Tvären	0.075	0.11	0.098	1.5	0.051	1.6	0.2	1.3	0.083
Öregrundsgrepen	0.051	<0.03	<0.04	0.24	<0.05	0.40	0.078	0.29	0.12
Östra Landsort	<0.03	<0.03	<0.04	0.42	<0.05	0.22	0.055	0.17	0.057
Östra Askrikefjärden	0.58	0.35	0.37	7.9	0.63	3.8	0.2	0.63	0.31
Sewage sludge									
Bromma	0.83	0.45	0.48	15	0.37	13	0.15	0.42	0.16
Henriksdal	0.99	0.47	0.50	9.2	0.96	12	0.10	0.26	0.10

*Interfering peak cannot be disregarded



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