Sakrapport

Övervakning av metaller och organiska miljögifter i marin biota, 2011

Överenskommelse 212 1011, dnr 235-3366-10Mm

Report nr 7:2011

Swedish Museum of Natural History
Department of Contaminant Research
P.O.Box 50 007
SE-104 05 Stockholm
Sweden
Comments Concerning the National Swedish Contaminant Monitoring Programme in Marine Biota, 2011

2011-04-15

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1 Introduction

This report summarises the monitoring activities within the National Swedish Contaminant Programme in marine biota. It is the result of joint efforts from the Department of Applied Environmental Science at Stockholm University (analyses of heavy metals, organochlorines, brominated flame retardants and perfluorinated substances), the Department of Chemistry at Umeå University (analyses of PCDD/PCDF), IVL – Swedish Environmental Research Institute (analyses of polycyclic aromatic hydrocarbons) and the Department of Contaminant Research at the Swedish Museum of Natural History (co-ordination, sample collection administration, sample preparation, recording of biological variables, storage of frozen biological tissues in the Environmental Specimen Bank for retrospective studies, data preparation and statistical evaluation). The monitoring programme is financed by the Swedish Environmental Protection Agency (EPA).

Data in this report represent the bioavailable portion of the investigated contaminants i.e. the portion that has effectively passed through biological membranes and may cause toxic effects. The objectives of the monitoring program in marine biota are as follows:

- To estimate the current levels and normal variation of various contaminants in marine biota from several representative sites, uninfluenced by local sources, along the Swedish coasts. The goal is to describe the general contaminant load and to supply reference values for regional and local monitoring programmes.

- To monitor long term time trends and to estimate the rate of changes found. *quantified objective:* to detect an annual change of 10% within a 10 year time period, with a power of 80% at a 5% significance level.

- To estimate the response in marine biota of measures taken to reduce the discharge of various contaminants. *quantified objective:* to detect a 50% decrease within a 10 year time period, with a power of 80% at a 5% significance level.

- To detect incidents of regional impact or widespread incidents of ‘Chernobyl’- character and to act as watchdog monitoring to detect renewed use of banned contaminants. *quantified objective:* to detect an increase of 200% in a single year, with a power of 80% at a 5% significance level.

- To indicate large scale spatial differences. *quantified objective:* to detect differences of a factor of 2 between sites, with a power of 80% at a 5% significance level.

- To explore developmental and regional differences in the composition and pattern of e.g. PCBs, HCHs, DDTs, PCDD/F, PBDE/HBCDD, PAHs and PFCs as well as the ratios between various contaminants.

- Because important commercial fish species like herring and cod are sampled, the time series are also relevant for human consumption of these species from Sweden. A cooperation with the Swedish Food Administration is established. Sampling is also co-
ordinated with SSI (Swedish Radiation Protection Authority) for analysing radionuclides in fish and blue mussels (HELCOM, 1992).

- All analysed samples, and numerous additional specimens, of annual systematically collected material, are stored frozen in the Environmental Specimen Bank. This material enables future retrospective studies of contaminants that are impossible to analyse today, as well as to control analyses of suspected analytical errors.

- Although the programme is focused on contaminant concentration in biota, it also investigates the development of biological variables, e.g. condition factor (CF), liver somatic index (LSI) and fat content, which are monitored at all sites. At a few sites, integrated monitoring of fish physiology and population are run in cooperation with the University of Gothenburg and the Swedish Board of Fisheries.

- Experience from the national programme, which has several time series of greater than 30 years, can be used in the design of regional and local monitoring programmes.

- The unique, high quality material and long time series is further used to explore relationships between biological variables and contaminant concentrations in various tissues, e.g. the effects of changes in sampling strategy, the estimates of variance components and the influence on the concept of power etc.

- The accessibility of high quality data collected and analysed in a consistent manner is an indispensable prerequisite for evaluating the validity of hypotheses and models concerning the fate and distribution of various contaminants. It could furthermore be used as input of ‘real’ data in the ongoing model building activities concerning marine ecosystems in general, and in the Baltic and North Sea environment in particular.

- The contaminant programme in marine biota constitutes an integrated part of the national monitoring activities in the marine environment, as well as of the international programmes within ICES, OSPARCOM, HELCOM and EU.

The present report displays the time series of analysed contaminants in biota and summarises results from the statistical treatment. It does not in general give background or explanations to significant changes found in the time series. Thus, increasing concentrations highlight the need for intensified studies.

Short comments are given for temporal trends as well as for spatial variation and, for some contaminants, differences in geometric mean concentration between various species caught at the same site. Sometimes notes of seasonal variation and differences in concentration between tissues in the same species are given. This information may indicate the relative appropriateness of the sampled matrix and be of help in designing future monitoring programmes. In the temporal trend section, an extract of the relevant findings is summarised in the 'conclusion' paragraph. However, it should be stressed that geographical differences may not reflect anthropogenic influences, but may instead be due to factors such as productivity, temperature, salinity etc.

This report is continuously updated. The date of the latest update can be found at the beginning of each chapter. The creation date of each figure is written in the lower left corner.
2 Summary 2009

The environmental toxicants examined in this report can be classified into five groups – heavy metals, chlorinated compounds, brominated flame retardants, polyaromatic hydrocarbons and perfluorinated compounds. Each of these contaminants has been examined from various sites for up to six different fish species, in blue mussels, and in guillemot eggs, for varying lengths of time. The following summary examines overall trends, spatial and temporal, for the five groups.

Condition and Fat Content
Condition and fat content in different species tended to follow the same pattern at the same sites, with a few exceptions. Most of the fish species generally displayed a decreasing trend in both condition and fat content at most sites examined. Exceptions to this were increases in fat content seen in herring (the last ten years) and cod at Fladen; an increase in condition for herring at Ångskärsklubb in spring; and in perch, a decrease in fat content at Kvädöfjärden but no trend in condition for the same site.

Heavy Metals
Due to a change in methods for metal analysis (not Hg) in 2004, values between 2003 and 2007 should be interpreted with care. From 2009 metals are analyzed at ITM, Stockholm university.

The longer time series in guillemot egg and spring-caught herring from the southern Bothnian Sea and southern Baltic Proper show significant decreases of mercury. The herring site in the southern Bothnian Sea indicates a local Hg-source. The rest of the time series show varying concentrations over the study period, and even increasing trends in e.g. cod muscle and blue mussels, but the concentrations are fairly low compared to measured concentrations in perch from fresh water and coastal sites. However, in most cases, these concentrations are above the newly suggested EU-target level of 20 ng/g wet weight.

Lead is generally decreasing over the study period (in time series of sufficient length), supposedly due to the elimination of lead in gasoline. Elevated lead concentrations between 2003 and 2007 (e.g. Harufjärden) should be viewed with caution (see above regarding change in analysis methods).

Cadmium concentrations show varying non-linear trends over the monitored period. It is worth noting that despite several measures taken to reduce discharges of cadmium, generally the most recent concentrations are similar to concentrations measured 30 years ago in the longer time series.

The reported nickel concentrations show no consistent decreasing trends. Some series begin with two elevated values that exert a strong leverage effect on the regression line and may give a false impression of decreasing trends. Chromium generally shows decreasing trends, possibly explained by a shift in analytical method. The essential trace metals, copper and zinc, show no consistent trends during the monitored period.
**Chlorinated Compounds**
Generally, a decreasing trend was observed for all compounds (DDT’s, PCB’s, HCH’s) in all species examined, with a few exceptions, such as no change in TCDD-equivalents being seen in herring muscle (except at Ånkärsklubb where very high concentrations at the beginning of the sampling period were seen. The longer time-series in guillemot also show a marked decrease from the start in the late 1960s until about 1985 from where no change can be seen in TCDD-equivalent concentrations).

The chlorinated compounds generally show a higher concentration in the Bothnian Sea and/or Baltic Proper when compared to the Bothnian Bay and the Swedish west coast.

**Brominated Flame Retardants**
Elevated levels of HBCDD are seen in sites from the southern Baltic Proper. PBDEs and HBCDD are more evenly distributed among sites compared to e.g. PCBs. Temporally, a significant increase in BDE-47, 99 and 100 has been seen in guillemot eggs since the late 1960s until the early 1990s, where concentrations then began to show a decrease. For herring and cod, HBCDD and BDE-47 decreased at some sites and showed no trend at other sites, whereas a 3% per year increase in HBCDD has been seen in guillemot eggs.

**PAHs**
Only blue mussels have been examined for spatial differences in PAH concentrations. Concentration of sPAH was found to be higher from Kvädöfjärden in the Baltic Proper, but individual PAHs showed varying spatial patterns. Over time, acenaphthalene was rarely found above the detection limit. Significant decreasing trends were observed for sPAH, chrysene, fluoranthene, naphthalene and pyrene at Väderöarna; for naphthalene at Kvädöfjärden; and for chrysene and pyrene at Fladen. Significant increasing trends in the last 10 years were only observed for fluorene at Väderöarna.

**PFCs**
PFHxS and PFOS show a similar spatial pattern, but PFOS concentrations were approximately 45 times higher than PFHxS levels. The distribution of PFOS is quite homogenous along the Swedish coast. PFOS concentrations in guillemot eggs are about 100-200 times higher than in herring liver. A consistently increasing trend of PFOS in guillemot eggs has been observed throughout the whole time period. This has increased at a faster rate since the mid-1990s compared to earlier years.

The temporal trends for all analysed compounds can be seen in Appendix 1.
3 Sampling

3.1 Sampling area
Sampling areas are defined by a central coordinate surrounded by a circumference of three nautical miles. The exact sampling location is registered at collection. General demands on sampling sites within the national contaminant monitoring programme are defined in chapter five.

3.2 Collected specimens
For many species, adult are more active than sub-adults and represent a more recent picture of the contaminant load since many contaminants bioaccumulate. To increase comparability between years, young specimens are generally collected. However, the size of individual specimens has to be big enough to allow individual chemical analysis. Thus, the size and age of specimens vary between species and sites (see chapter four). To avoid possible influences of between-year variance due to sex differences, the same sex (female) is analysed each year in most time series. In the past, both sexes were used and thus, at least for the oldest time series, both sexes appear. To achieve the requested number of individual specimens of the prescribed age and sex range, about 50 - 100 specimens are collected at each site. Only healthy looking specimens with undamaged skin are selected.

The collected specimens are placed individually in polyethylene plastic bags, frozen as soon as possible, and transported to the sample preparation laboratory.

Collected specimens not used for the annual contaminant monitoring programme are stored in the Environmental Specimen Bank (ESB) (see Odsjö 1993 for further information). These specimens are registered. Biological information and notes of the available amount of tissue, together with a precise location in the ESB, are accessible from a database. These specimens are thus available for retrospective analyses or for control purposes.

3.3 Number of samples and sampling frequency
In general, 10 - 12 individual specimens from the old Baltic sites (reported to the Helsinki Convention (HELCOM)) and the old Swedish west coast sites (reported to OSPARCOM) are analysed annually from each site for each species. At the new Baltic and west coast sites, 2 pools of 12 individuals are analysed from each site for each species. For guillemot eggs and perch (old sites), 10 individual specimens are analysed. Organochlorines in blue mussels are analysed in pooled samples containing approximately 20 individuals in each pool. Since 1996, samples from 12 individual specimens are analysed, which is proposed in the revised guidelines for HELCOM and OSPARCOM.

The sampling recommendation prescribes a narrow age range for sampled species. In a few cases it has not been possible to achieve the required number of individuals within that range. In order to reduce the between-year variation due to sampling differences in age composition, only specimens within the age range classes given in brackets after species names in the figures, are selected for this presentation.

Sampling is carried out annually for all time series. Less frequent sampling would result in a considerable loss in statistical and interpretational power.
3.4 Sampling season
Sampling of the various fish species and blue mussels is carried out every autumn, outside the spawning season. However, from two sites, Ångskärsklubb and Utlängan, herring is also sampled in spring. The two spring time series were started in 1972. To begin with, only organochlorines where analysed, but since 1996, metals have been analysed on a yearly basis. This provides the possibility to study seasonal differences and, when possible, to adjust for these differences and improve the resolution of the time series. It also gives an opportunity to study possible changes in the frequencies of spring and autumn spawners.

Guillemot eggs are collected in the beginning to the middle of May. Due to a lost first egg, a second egg is often laid. These second eggs should not be collected. To avoid this, only early laid eggs are sampled (see section 4.6).

3.5 Sample preparation and registered variables
A short description of the various sampling matrices and the type of variables that are registered are given below. See TemaNord (1995) for further details.

3.5.1 Fish
For each specimen, total body weight, total length, body length, sex, age (see chapter four for various age determination methods for different species), reproductive stage, state of nutrition, liver weight and sample weight are registered.

Muscle samples are taken from the middle dorsal muscle layer. The epidermis and subcutaneous fatty tissue are carefully removed. Samples of 10 g muscle tissue are prepared for organochlorine/bromine analysis, 20 g for analysis of PCDD/F and 1.5 g for mercury analysis.

The liver is completely removed and weighed. Samples of 0.5 – 1 g are prepared for metal analyses, and 0.5 g for analysis of perfluorinated substances.

3.5.2 Blue mussels
For each specimen, total shell length, shell and soft body weight are registered. Trace metals are analysed in individual mussels, whereas samples for organochlorine/bromine determination and PAHs are analysed in pools of approximately 20 specimens.

3.5.3 Guillemot egg
Length, width and total weight are recorded. Egg contents are removed (blown out). Embryo tissue is separated from the yolk and white, which are homogenised.

Weight of the empty, dried eggshell is recorded. Egg shell thickness is measured at the blowing hole using a modified micrometer.

Two grams of the homogenised egg content is prepared for mercury analyses, and another 2 g for the other analysed metals. Ten grams is prepared for analyses of organochlorines/bromines, 30 g for analysis of PCDD/F and 1 g for perfluorinated substances.

3.6 Data registration
Data are stored in a flat ASCII file in a hierarchical fashion, where each individual specimen represents one level. Each measured value is coded and the codes are defined in a code list (Danielsson, Gustavsson and Nyberg, 2011). The primary data files are processed through a quality control program. Suspect values are checked and corrected if necessary. Data are retrieved from the primary file into a table format suitable for import to database or statistical programs.
4 Sample matrices

The sample database provides the basic information for this report, and contains data of contaminant concentrations in biota from individual specimens of different species (table 4.1).

Table 4.1. Number of specimens for various species sampled for analysis of contaminants within the base program.

<table>
<thead>
<tr>
<th>Species</th>
<th>N of individual specimen</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring</td>
<td>5640</td>
<td>51</td>
</tr>
<tr>
<td>Cod</td>
<td>1090</td>
<td>10</td>
</tr>
<tr>
<td>Perch</td>
<td>970</td>
<td>9</td>
</tr>
<tr>
<td>Eelpout</td>
<td>530</td>
<td>5</td>
</tr>
<tr>
<td>Dab</td>
<td>350</td>
<td>3</td>
</tr>
<tr>
<td>Flounder</td>
<td>340</td>
<td>3</td>
</tr>
<tr>
<td>Guillemot</td>
<td>600</td>
<td>5</td>
</tr>
<tr>
<td>Blue mussel</td>
<td>1580</td>
<td>14</td>
</tr>
<tr>
<td>Total</td>
<td>11100</td>
<td>100</td>
</tr>
</tbody>
</table>

4.1 Herring (*Clupea harengus*)

Herring is a pelagic species that feeds mainly on zooplankton. It becomes sexually mature at about 2 - 3 years in the Baltic, and 3 - 4 years on the Swedish west coast. It is the most dominant commercial fish species in the Baltic. It is important not only for human consumption but also for several other predators in the marine environment.

Herring is the most commonly used indicator species for monitoring contaminants in biota within the BMP (Baltic Monitoring Programme) in the HELCOM convention area, and is sampled by Finland, Estonia, Poland and Sweden.

Herring muscle tissue is fat and thus very appropriate for analysis of fat-soluble contaminants i.e. hydrocarbons.

Herring samples are collected each year from seventeen sites along the Swedish coasts: Rånefjärden, Harufjärden, Kinnbäcksfjärden (Bothnian Bay), Holmöarna, Örefjärden, Gaviksälvens fjärden, Långvindsfjärden, Ångskärsklubb (Bothnian Sea), Lagnö, Landsort (northern Baltic Proper), Byxelkrok, Abbekås, Hanöbukten, Utlängan (southern Baltic Proper), Kullen, Fladen (Kattegatt) and at Väderöarna (Skagerrak). Herring are also collected from two sites in the open sea, the Baltic Proper and the Bothnian Sea, (by the Swedish Board of Fisheries).

Herring liver tissue is analysed for lead, cadmium, copper, zinc and perflourinated substances. In 1995, analyses of chromium and nickel were added to the programme. Herring muscle tissue is analysed for mercury, organochlorines (DDTs, PCBs, HCHs, HCB and PCDD/PCDF) and polybrominated flame retardants. Herring muscle from spring-caught specimens from Ångskärsklubb and Utlängan are analysed for organochlorines and polybrominated flame retardants. From 1996, herring tissue has also been analysed for the above mentioned metals. Herring samples from various sites within the marine monitoring programme have been analysed for dioxins/dibenzofurans, co-planar CBs, polybrominated diphenyl ethers (Sellström, 1996) and fat composition in pilot studies. Monitoring of Cs-
135 is also carried out on herring from these sites by the Swedish Radiation Protection Institute.

The age of the herring specimens is determined using their scales. The analysed specimens are females, between 2 - 5 years. Total body weight, liver weight, total length and maturity of gonads are recorded (Table 4.2). Growth rate varies considerably at the different sites (Table 4.3).

Table 4.2. Weeks when sample collections have been carried out in all (or most) years at the old locations; selected age classes are presented in the time series below. The 95% confidence intervals for the yearly means of total body weight, total length, liver weight and liver and muscle dry weight are given.

<table>
<thead>
<tr>
<th>Sampling week</th>
<th>age (year)</th>
<th>body weight (g)</th>
<th>length (cm)</th>
<th>liver weight (g)</th>
<th>liver dry weight (%)</th>
<th>muscle dry weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harufjärden</td>
<td>38-42</td>
<td>3-4</td>
<td>28-31</td>
<td>16-17</td>
<td>0.32-0.39</td>
<td>20-35</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>22-23</td>
</tr>
<tr>
<td>Ångskärsklubb</td>
<td>38-42</td>
<td>3-5</td>
<td>33-42</td>
<td>17-18</td>
<td>0.38-0.56</td>
<td>20-35</td>
</tr>
<tr>
<td>- spring</td>
<td>20-24</td>
<td>2-5</td>
<td>25-33</td>
<td>16-17</td>
<td>0.31-0.54</td>
<td>19-23</td>
</tr>
<tr>
<td>Landsort</td>
<td>41-48</td>
<td>3-5</td>
<td>38-50</td>
<td>18-20</td>
<td>0.46-0.66</td>
<td>20-32</td>
</tr>
<tr>
<td>- spring</td>
<td>18-23</td>
<td>2-3</td>
<td>51-65</td>
<td>19-22</td>
<td>0.30-0.55</td>
<td>17-20</td>
</tr>
<tr>
<td>Karlskrona</td>
<td>41-46</td>
<td>2-4</td>
<td>38-48</td>
<td>17-19</td>
<td>0.36-0.51</td>
<td>22-35</td>
</tr>
<tr>
<td>- spring</td>
<td>35-45</td>
<td>2-3</td>
<td>47-61</td>
<td>19-20</td>
<td>0.55-0.70</td>
<td>22-38</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>38-40</td>
<td>2-3</td>
<td>50-90</td>
<td>18-24</td>
<td>0.40-1.0</td>
<td>27-39</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>24-35</td>
</tr>
</tbody>
</table>

Table 4.3. Average length at the age 3 years, and age at 16 cm length at the old sites.

<table>
<thead>
<tr>
<th>Average length (cm) at 3 years</th>
<th>Average age (years) at 16 cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harufjärden</td>
<td>15.91</td>
</tr>
<tr>
<td>Ångskärsklubb</td>
<td>16.87</td>
</tr>
<tr>
<td>- spring</td>
<td>16.79</td>
</tr>
<tr>
<td>Landsort</td>
<td>17.28</td>
</tr>
<tr>
<td>Karlskrona</td>
<td>18.20</td>
</tr>
<tr>
<td>Fladen</td>
<td>20.32</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>21.73</td>
</tr>
</tbody>
</table>

4.2 Cod (Gadus morhua)

The Baltic cod lives below the halocline, feeding on bottom organisms. In Swedish waters, it becomes sexually mature between 2 - 6 years old. Spawning takes place during May - August (occasionally spawning specimens can be found in March or September). Cod require a salinity of at least 11 PSU, and an oxygen content of at least 2 ml/l (Nissling, 1995) to successfully spawn. The population shows great fluctuations and decreased dramatically between 1984 - 1993. Cod fishing for human consumption is economically important.

Cod is among the ‘first choice species’ recommended within the JAMP (Joint Assessment and Monitoring Programme) and BMP (Baltic Monitoring Programme).

Cod is collected in autumn from two sites - south east of Gotland, and from Fladen on the Swedish west coast. Cod age is determined using otoliths. Specimens of both sexes, between 3 - 4 years from Gotland, and between 2 - 4 years from Fladen, are analysed (Table 4.4).
Table 4.4. Weeks when sample collections have been carried out in all (or most) years at a specific location; selected age classes are presented in the time series below. The 95% confidence intervals for the yearly means of total body weight, total length, liver weight and liver dry weight are given.

<table>
<thead>
<tr>
<th>Sampling week</th>
<th>age (year)</th>
<th>body weight (g)</th>
<th>Length (cm)</th>
<th>liver weight (g)</th>
<th>liver dry weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SE Gotland</td>
<td>35-39</td>
<td>310-455</td>
<td>32-35</td>
<td>16-41</td>
<td>53-63</td>
</tr>
<tr>
<td>Fladen</td>
<td>37-42</td>
<td>240-345</td>
<td>29-33</td>
<td>4-10</td>
<td>33-44</td>
</tr>
</tbody>
</table>

The cod liver is fat and organic contaminants are often found in relatively high concentrations. For that reason, it is a very appropriate matrix for screening for ‘new’ contaminants.

Cod liver tissue is analysed for lead, cadmium, copper and zinc, as well as for organochlorines. In 1995, analyses of chromium and nickel were added, and in 1999, analysis for brominated substances and HBCDD were added. Cod muscle tissue is analysed for mercury.

Before 1989, 20 individual samples from south east of Gotland, and 25 samples from the Kattegatt were analysed for organochlorines. Between 1989 - 1993 one pooled sample from each site in, each year was analysed. Since 1994, 10 individual cod samples are analysed at the two sites every year.

4.3 Dab (*Limanda limanda*)

Dab is a bottom living species feeding on crustaceans, mussels, worms, echinoderms and small fish. Males become sexually mature between 2 - 4 years, and females between 3 - 5 years. Spawning takes place during April – June in shallow coastal waters. Dab tend to migrate to deeper water in late autumn.

Dab is among the ‘first choice species’ recommended within the JAMP (Joint Assessment and Monitoring Programme).

Because of reduced analytical capacity, organochlorines in dab were analysed annually in one pooled sample from 1989 - 1995. Since 1995, samples of dab are no longer analysed but are still collected and stored in the Environment Specimen Bank (ESB).

Dab is collected from the Kattegatt (Fladen) in autumn. Liver tissue samples have been analysed for lead, cadmium, copper and zinc, and muscle tissue samples for organochlorines and mercury. Dab age is determined using otoliths. Specimens between 3 - 5 years have been analysed (Table 4.5).

Table 4.5. Weeks when sample collections have been carried out in all (or most) years; selected age classes are presented in the time series below. The 95% confidence intervals for the yearly means of total body weight, total body length, liver weight and liver dry weight are given.

<table>
<thead>
<tr>
<th>Sampling week</th>
<th>age (year)</th>
<th>body weight (g)</th>
<th>length (cm)</th>
<th>liver weight (g)</th>
<th>liver dry weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fladen</td>
<td>37-44</td>
<td>50-250</td>
<td>15-30</td>
<td>0.5-2</td>
<td>20-40</td>
</tr>
</tbody>
</table>
4.4  **Flounder** (*Platichthys flesus*)

Flounder is a bottom-dwelling species that feeds on crustaceans, mussels, worms, echinoderms and small fish. In the Skagerrak, males become sexually mature between 3 - 4 years of age, and females one year later. Spawning in the Skagerrak takes place during January – April in shallow coastal waters. Flounder tend to migrate to deeper waters in late autumn.

Flounder is among the ‘second choice species’ recommended within the JAMP.

Because of reduced analytical capacity, organochlorines in flounder were analysed annually in one pooled sample from 1989 - 1995. Since 1995, flounder samples are no longer analysed but are still collected and stored in the ESB.

Flounder is collected from the Skagerrak (Väderöarna) in autumn. Liver tissue samples have been analysed for lead, cadmium, copper and zinc, and muscle tissue samples for organochlorines and mercury. Flounder age is determined using otoliths. Specimens between 4 - 6 years have been analysed (Table 4.6).

<table>
<thead>
<tr>
<th>Sampling week</th>
<th>age (year)</th>
<th>body weight (g)</th>
<th>length (cm)</th>
<th>liver weight (g)</th>
<th>liver dry weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Väderöarna</td>
<td>37-44</td>
<td>3-6</td>
<td>100-400</td>
<td>20-35</td>
<td>1-5</td>
</tr>
</tbody>
</table>

4.5  **Blue mussels** (*Mytilus edulis*)

Blue mussels are one of the most commonly used organisms for monitoring contaminants in biota. Adult mussels are sessile, hence it is easier to define the area that the samples represent compared to fish.

Blue mussels are among the ‘first choice species’ recommended within the JAMP.

Blue mussels are collected from the Kattegatt (Fladen, Nidingen), the Skagerrak (Väderöarna) and Kvädöfjärden in the Baltic Proper. The mussels are sampled in autumn. Sampling depth varies between the sampling sites (Table 4.7).

Soft body tissue is analysed for lead, cadmium, copper, zinc, mercury and organochlorines. In 1995 analyses of chromium and nickel were added, and in 2000 analysis of brominated substances. From 1995, samples from Kvädöfjärden were included in the analysis. Since 1981, samples from this site had only been collected and stored. Organochlorines in blue mussels are analysed in pooled samples from each site and year, whereas trace metals are analysed in 25 individual samples per year and site (15 from 1996). PAHs have been analysed retrospectively (start 1984/87) in mussels from all three localities and, since 2003, are analysed on a yearly basis in pooled samples (Table 4.7).
Table 4.7. Weeks when collection of samples has been carried out in all (or most) years at a specific location; selected shell length interval are presented in the time series below. The 95% confidence intervals for the yearly means of soft body weight and shell weight are given.

<table>
<thead>
<tr>
<th>Location</th>
<th>Sampling week</th>
<th>Sampling depth</th>
<th>Shell length</th>
<th>Shell weight</th>
<th>Soft body weight</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kvädöfjärden</td>
<td>38-43</td>
<td>2-10</td>
<td>2-3</td>
<td>0.4-0.6</td>
<td>1-2</td>
</tr>
<tr>
<td>Fladen, Nidingen</td>
<td>37-51</td>
<td>0.5</td>
<td>5-8</td>
<td>5-25</td>
<td>2-10</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>42-51</td>
<td>2</td>
<td>6-10</td>
<td>10-30</td>
<td>5-25</td>
</tr>
</tbody>
</table>

4.6 Guillemot (*Uria aalge*)

Guillemots are suitable for monitoring contaminants in the Baltic Sea as most do not migrate further than the southern parts of the Baltic Proper during the winter season. They feed mainly on sprat (*Sprattus sprattus*) and herring (*Clupea harengus*). Guillemot breed for the first time at 4 - 5 years. Eggs hatch after about 32 days.

The egg content is high in fat (11 - 13%), thus very appropriate for analysis of fat-soluble contaminants i.e. hydrocarbons.

Normally the guillemot lay just a single egg but if this egg is lost, another may be laid. It has been shown that guillemot eggs that are laid late tend to contain significantly higher concentrations of organochlorines compared to eggs laid early (Bignert et al. 1995). In this report, only early laid eggs are included, except for dioxins, where the results from all collected eggs are included. Ten guillemot eggs, collected between weeks 19 – 21 (22), are analysed each year.

Guillemot egg contents from St Karlsö are analysed for mercury, organochlorines, perflourinated compounds (Holmström et al. 2005) and polybrominated compounds (Sellström, 1996). From 1996, the concentrations of lead, cadmium, nickel, chromium, copper and zinc have also been analysed. The time series has also been analysed for PCC (Wideqvist et al. 1993). Various shell parameters, for example shell weight, thickness and thickness index, are also monitored. The weight of several hundred fledglings is normally recorded each year at St Karlsö. Eggs have also been collected for some years from Bonden in the northern Bothnian Sea, but so far only results (organochlorines) from 1991 are available.

4.7 Perch (*Perca fluviatilis*)

Perch is an omnivorous, opportunistic feeding predatory fish. Male perch become sexually mature between 2 - 4 years and females between 3 - 6 years. Spawning takes place during April - June when the water temperature reaches about 7 - 8 degrees celcius. Perch muscle tissue is lean and contains only about 0.8% fat.

Integrated monitoring of fish physiology and population development is carried out on perch in cooperation with the University of Gothenburg and the Swedish Board of Fisheries. Perch is also used as an indicator species for contaminant monitoring within the national monitoring programme of contaminants in freshwater biota.

Perch muscle tissue samples from two coastal sites, Holmöarna and Kvädöfjärden in the Baltic (Table 4.8), are analysed for organochlorines and mercury. In 1995, analyses of lead, cadmium, chromium, nickel, copper and zinc in perch liver were added to the programme, and in 2006 PCDD/F was added.
Table 4.8. Weeks when sample collections have been carried out in all (or most) years at the old sites; selected age classes are presented in the time series below. The 95% confidence intervals for the yearly means of total body weight, total body length, liver weight and liver dry weight are given.

<table>
<thead>
<tr>
<th>Perch</th>
<th>Sampling week (year)</th>
<th>age (cm)</th>
<th>body weight (g)</th>
<th>length (cm)</th>
<th>liver weight (g)</th>
<th>liver dry weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Holmöarna</td>
<td>33-42</td>
<td>3-5</td>
<td>77-88</td>
<td>17-21</td>
<td>0.86-1.5</td>
<td></td>
</tr>
<tr>
<td>Kvädöfjärden</td>
<td>31-40</td>
<td>3-5</td>
<td>56-67</td>
<td>15-20</td>
<td>0.50-0.73</td>
<td></td>
</tr>
</tbody>
</table>

4.8 Eelpout, viviparous blenny (Zoarces viviparus)

Eelpout is considered to be a more or less stationary species living close to the bottom, feeding on insect larvae, molluscs, crustaceans, worms, hard roe and small fish. It becomes sexually mature when 2 years old at a length of 16 - 18 cm. Spawning takes place during August - September. After 3 - 4 weeks, eggs hatch inside the mother’s body where the fry stay for about three months. The possibility to measure the number of eggs, fertilised eggs, larvae size and embryonic development makes this species suitable for integrated studies of contaminants and reproduction (Jacobsson et al. 1993). Integrated monitoring of fish physiology and population development is carried out on eelpout in cooperation with the University of Gothenburg and the Swedish Board of Fisheries.

Eelpout specimens have been collected from Väderöarna in the Skagerrak since 1988. In this time series, analyses of various PCB congeners are available. Since 1995, eelpout have also been collected from Holmöarna and Kvädöfjärden (Table 4.9). Liver tissue is analysed for lead, cadmium, chromium, nickel, copper and zinc, whereas muscle tissue is analysed for mercury and organochlorines. Contaminant analysis in eelpout from Holmöarna ended in 2007.

Table 4.9. Weeks when sample collections have been carried out in all (or most) years at a specific location; selected age classes are presented in the time series below. The 95% confidence intervals for the yearly means of total body weight, total body length, liver weight and liver and muscle dry weight are given.

<table>
<thead>
<tr>
<th>Sampling week (year)</th>
<th>age (year)</th>
<th>total weight (g)</th>
<th>length (cm)</th>
<th>liver weight (g)</th>
<th>liver dry weight (%)</th>
<th>muscle dry weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Holmöarna</td>
<td>47</td>
<td>3-6</td>
<td>21-26</td>
<td>18-20</td>
<td>0.20-0.50</td>
<td>13-26</td>
</tr>
<tr>
<td>Kvädöfjärden</td>
<td>46</td>
<td>3-6</td>
<td>28-39</td>
<td>19-22</td>
<td>0.20-0.60</td>
<td>18-25</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>(36), 45-47</td>
<td>3-6</td>
<td>35-70</td>
<td>20-25</td>
<td>0.40-1.00</td>
<td>14-32</td>
</tr>
</tbody>
</table>

26
5 Sampling sites

The location and names of the sample sites are shown in figure 5.1. The sampling sites are located in areas regarded as locally uncontaminated and, as far as possible, uninfluenced by major river outlets or ferry routes and not too close to heavily populated areas.

The Swedish sampling stations are included in the net of HELCOM stations in the Baltic and the Oslo and Paris Commissions’ Joint Monitoring Programme (OSPAR, JMP) station net in the North Sea. Denmark (plaice), Estonia (herring, perch), Finland (herring), Germany (perch, cod, herring), Latvia (perch), Lithuania (herring, cod, flounder) and Poland (herring) all report contaminant data within HELCOM. Within the JMP, the time series of various contaminants in biota are reported from Belgium, Denmark, France, Germany, Iceland, The Netherlands, Norway, Spain, Sweden, Ireland and UK. All of the countries within HELCOM and OSPAR submit the data directly to ICES.

During 2007, the National Swedish marine monitoring programme has been expanded, and herring from 10 new sites have been added. Name and location of these sites are found in figure 5.1. From 2007 onwards, herring has also been collected by the Swedish Board of Fisheries from a number of sites in the open sea (Baltic). Two sites, one from the Baltic Proper and one from the Bothnian Sea (fish from 2008 onwards) have been analysed for various contaminants within the national monitoring programme.
5.1 Rånefjärden, Bothnian Bay, north
Co-ordinates: 65° 45’N, 22° 25’E within a radius of 3’, ICES 60H2 93
County: Norrbottens län

Surface salinity: <3 PSU
Average air temperature: January: -10° / April: -1° / July: 15° / October: 2°

Sampling matrix: Baltic herring and perch (only sampling), autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs.

5.2 Harufjärden, Bothnian Bay, north
Co-ordinates: 65° 35’N, 22° 53’E within a radius of 3’, ICES 60H2 93
County: Norrbottens län

Surface salinity: <3 PSU
Average air temperature: January: -10° / April: -1° / July: 15° / October: 2°

Sampling matrix: Baltic herring, autumn
Start: 1978 DDT/PCB; 1980 Hg; 1982 Pb/Cd/Cu/Zn; 1988 HCHs/HCB; 1995 Cr/Ni

5.3 Kinnbäcksfjärden, Bothnian Bay
Co-ordinates: 64° 50’N, 21° 16’E within a radius of 3’, ICES 58H1
County: Norrbottens län

Average air temperature: January: -10° / April: -1° / July: 15° / October: 2°

Sampling matrix: Baltic herring and perch, autumn
Start: Only sampling

5.4 Holmöarna, Bothnian Bay, south, coastal site
Co-ordinates: 63° 41’N, 20° 53’E, ICES 56H0
County: Västerbottens län

Surface salinity: c 4 PSU
Average air temperature: January: -5° / April: 0° / July: 15° / October: 4°

Table 5.1. Start year for various contaminants for perch and eelpout.

<table>
<thead>
<tr>
<th>Contaminant/Species</th>
<th>PCB/DDT</th>
<th>HCH/HCB</th>
<th>Hg</th>
<th>Pb/Cd/Cu/Zn</th>
<th>Cr/Ni</th>
<th>PCDD/F</th>
</tr>
</thead>
<tbody>
<tr>
<td>Perch</td>
<td>1980</td>
<td>19(89)95</td>
<td>19(91)95</td>
<td>1995</td>
<td>1995</td>
<td>2007</td>
</tr>
</tbody>
</table>

Both species are collected during autumn. Since 2007, Baltic herring has also been sampled.

At Holmöarna, the contaminant monitoring is integrated with fish population and physiology monitoring, carried out by the Swedish Board of Fisheries and the University of Gothenburg.

5.5 Örefjärden, Bothnian Bay, south
Co-ordinates: 63° 25’N, 19° 24’E within a radius of 3’, ICES 55G9
County: Västernorrlands län
Average air temperature: January: -10° / April: -1° / July: 15° / October: 2°
Sampling matrix: Baltic herring and perch, autumn
Start: Only sampling

5.6 Gaviksfjärden, Bothnian Bay, south
Co-ordinates: 63° 07’N, 18° 38’E within a radius of 3’, ICES 55G8
County: Västernorrlands län
Average air temperature: January: -10° / April: -1° / July: 15° / October: 2°
Sampling matrix: Baltic herring and perch 8only sampling), autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs

5.7 Långvindsfjärden, Bothnian Sea
Co-ordinates: 61° 46’N, 17° 27’E within a radius of 3’, ICES 52G7
County: Gävleborgs län
Average air temperature: January: -3° / April: 2° / July: 15° / October: 6°
Sampling matrix: Baltic herring and perch (only sampling), autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs

5.8 Ängskärsklubb, Bothnian Sea
Co-ordinates: 60° 44’N, 17° 52’E, ICES 50G7 83
County: Gävleborgs län/Uppsala län
Surface salinity: c 6 PSU
Average air temperature: January: -3° / April: 2° / July: 15° / October: 6°
Sampling matrix: Baltic herring, spring/autumn
Start, spring: 1972 DDT/PCB, 1972-75 Hg, 1988 HCHs/HCB, 1979 PCDD/F, 2005 PFC
In 1996, collection and analyses of herring samples from four other sites in the region were financed by the county board of Gävleborgs län. This investigation is valuable to estimate how representative the well established sample site at Ängskärsklubb is. It also gives information on small scale geographical variation in general.

5.9 Lagnö, Baltic Proper, north
Co-ordinates: 59° 25’N, 18° 37’E, ICES 47G8
County: Stockholms län
Surface salinity: c 6-7 PSU
Average air temperature: January: -1° / April: 3° / July: 16° / October: 7°
Sampling matrix: Baltic herring and perch (only sampling), autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs
5.10 Landsort, Baltic Proper, north
Co-ordinates: 58° 42’N, 18° 04’E, ICES 46G8 23
County: Stockholms län/Södermanlands län

Surface salinity: c 6-7 PSU
Average air temperature: January: -1° / April: 3° / July: 16° / October: 7°

Sampling matrix: Baltic herring, autumn

Herring samples have also been collected to analyse metallothionein concentration and to compare the fat composition in old versus young herring specimen.

5.11 Kvädöfjärden, Baltic Proper, coastal site
Co-ordinates: 58° 2’N, 16° 46’E, ICES 45G6
County: Östergötland / Kalmar

Surface salinity: c 6-7 PSU
Average air temperature: January: -1° / April: 4° / July: 17° / October: 7°

<table>
<thead>
<tr>
<th>Contaminant/Species</th>
<th>PCB/DDT</th>
<th>HCH/HCB</th>
<th>Hg</th>
<th>Pb/Cd/Cu/Zn</th>
<th>Cr/Ni</th>
<th>PAH</th>
<th>PCDD/F</th>
</tr>
</thead>
</table>

All species are collected during autumn.

At Kvädöfjärden, contaminant monitoring is integrated with fish population and physiology monitoring, carried out by the Swedish Board of Fisheries and the University of Gothenburg.

Neuman et al. (1988) reports decreasing Secchi depths during the investigated period, from just below 6 m in 1980, to just above 4 m in the mid-1980s.

5.12 Byxelkrok, Baltic Proper
Co-ordinates: 57° 19’N, 17° 30’E, ICES 43G6
County: Kalmar län

Surface salinity: c 7 PSU
Average air temperature: January: 0° / April: 3° / July: 16° / October: 8°

Sampling matrix: Baltic herring, autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs
5.13 St Karlsö, Baltic Proper
Co-ordinates: 57° 11’N, 17° 59’E, ICES 43G7 County: Gotland

St Karlsö is situated about 7 km west of the island of Gotland and about 80 km east of the Swedish Baltic coast.

Surface salinity: c 7 PSU
Average air temperature: January: 0° / April: 3° / July: 16° / October: 8°

Sampling matrix: Guillemot egg, May
Start: 1968 DDT/PCB; 1969 Hg; 1988 HCHs/HCB

5.14 South east of Gotland, Baltic Proper
Co-ordinates: 56° 53’N, 18° 38’E, ICES 42G8 43 County: Gotland

Surface salinity: c 7-8 PSU
Average air temperature: January: 0° / April: 3° / July: 16° / October: 8°

Sampling matrix: Cod, autumn
Start: 1980 DDT/PCB/Hg; 1982 Pb/Cd/Cu/Zn; 1988 HCHs/HCB; 1995 Cr/Ni; 1999 PBDE/HBCDD

5.15 Utlängan, Karlskrona archipelago, Baltic Proper, south
Co-ordinates: 55° 57’N, 15° 47’E, ICES 40G5 73 County: Blekinge

Surface salinity: c 8 PSU
Average air temperature: January: 0° / April: 4° / July: 16° / October: 8°

Table 5.3. Start year for analysis of various contaminants for herring in spring and autumn.

<table>
<thead>
<tr>
<th>Contaminant/Species</th>
<th>PCB/DDT</th>
<th>HCH/HC B</th>
<th>Hg</th>
<th>Pb/Cd/Cu/Zn</th>
<th>Cr/Ni</th>
<th>PBDE/HBC DD</th>
<th>PCDD/F</th>
<th>PFC</th>
</tr>
</thead>
</table>

In 1997, collection and analyses of herring samples from one site rather close to the reference site, and two sites in Hanöbuken, were financed by the Swedish EPA. This investigation is valuable to estimate how representative the well-established sample site at Utlängan is. It will also give information on small-scale geographical variation in general.
5.16  Västra Hanöbukten, Baltic Proper, south
Co-ordinates: 55° 45’N, 14° 17’E, ICES 40G4
County: Skåne

Surface salinity: c 8 PSU
Average air temperature: January: 0° / April: 4° / July: 16° / October: 8°

Sampling matrix: Baltic herring, autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs

5.17  Abbekås, Baltic Proper, south
Co-ordinates: 55° 18’N, 13° 36’E, ICES 39G3
County: Skåne

Surface salinity: c 8 PSU
Average air temperature: January: 0° / April: 4° / July: 16° / October: 8°

Sampling matrix: Baltic herring, autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs

5.18  Kullen, Kattegatt, Swedish west coast
Co-ordinates: 56° 19’N, 12° 23’E, ICES 41G2
County: Skåne

Surface salinity: c 20-25 PSU
Average air temperature: January: 0° / April: 5° / July: 16° / October: 8°

Sampling matrix: Herring, autumn
Start: 2007 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs

5.19  Fladen, Kattegatt, Swedish west coast
Co-ordinates: 57° 14 N, 11° 50’E, ICES 43G1 83, JMP J34
County: Halland

Surface salinity: c 20-25 PSU
Average air temperature: January: 0° / April: 5° / July: 16° / October: 8°

| Table 5.4. Start year for various contaminants for herring, cod, dab and blue mussels. |
|---------------------------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| Contaminant/Species             | PCB/DDT  | HCH/HCB   | Hg        | Pb/Cd/Cu/Zn | Cr/Ni     | PAH       | PBDE/HBCDD | PCDD/F    | PFC       |
| Dab                             | 1981      | 1988      | 1981      | 1981        | -         |           |            |            |            |

All species are collected during autumn.
5.20 Nidingen, Kattegatt, Swedish west coast

Since 1987, blue mussels have been collected at Nidingen about 10 km NNE of Fladen.

5.21 Väderöarna, Skagerrak, Swedish west coast

Co-ordinates: 58° 31’N, 10° 54’E ICES 46G0 93, JMP J33
County: Göteborgs- o Bohus län

Surface salinity: c 25-30 PSU
Average air temperature: January: 0° / April: 5° / July: 16° / October: 8°

Table 5.5. Start year for various contaminants for herring, eelpout, flounder and blue mussels.

<table>
<thead>
<tr>
<th>Contaminant/Species</th>
<th>PCB/ HCH/ Hg</th>
<th>Pb/Cd/ Cr/Ni</th>
<th>PAH</th>
<th>PBDE/ HBCDD</th>
<th>PCDD/ F</th>
<th>PFC</th>
</tr>
</thead>
<tbody>
<tr>
<td>HCB</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

All species are collected during autumn.

5.22 Fjällbacka, Skagerrak, Swedish west coast

Eelpout and blue mussels are collected at Musön and Fjällbacka on the Swedish coast, about 10 km east of Väderöarna.

5.23 Bothnian Sea, offshore site

Co-ordinates: 61° 22 N, 19° 16’E, ICES 51G9

Average air temperature: January: -3° / April: 2° / July: 15° / October: 6°

Sampling matrix: Herring, autumn
Start: 2008 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs

5.24 Baltic Proper north, offshore site

Co-ordinates: 58° 51’N, 20° 18’E, ICES 46H0

Surface salinity: c 6-7 PSU
Average air temperature: January: -1° / April: 3° / July: 16° / October: 7°

Sampling matrix: Herring, autumn
Start: 2008 DDT/PCB, Hg, Pb/Cd/Cu/Zn/Cr/Ni, HCHs/HCB, PBDE/HBCDD, PCDD/F and PFCs.
5.25  **Boden, northern Bothnian Sea**

Co-ordinates:  63° 25’N, 20° 02’E,  ICES 55H0
County:  Västerbotten

Surface salinity:  c 5 PSU
Average air temperature:  January: -5° / April: 0° / July: 15° / October: 4°

Sampling matrix:  Guillemot egg (only rotten eggs), summer
Start:  1991 DDT/PCB

The collection of egg samples has been sporadic because of low population growth.
6 Analytical methods

6.1 Trace metals

Prior to 2007, metal analyses were carried out at the Department of Environmental Assessment at the Swedish University of Agricultural Sciences (SLU). Due to some inconsistencies in results, the results from the years 2003 up to 2006 should be looked upon with caution. From 2007, the analyses were carried out at the Department of Applied Environmental Science (ITM) at Stockholm University (SU).

Prior to 2007, heavy metal concentrations, except mercury, in fish liver and blue mussel soft body were determined using an atomic absorption spectrophotometer with a graphite furnace at SLU. The quantification limit was estimated to approximately 100 ng/g dry weight for zinc, approximately 10 ng/g dry weight for lead and copper, approximately 5 ng/g dry weight for cadmium and approximately 0.1 μg/g dry weight for nickel and chromium, which implies that the concentrations in herring, flounder and dab are approximately 10 - 20 times above the quantification limit.

Since 2007, ITM has determined heavy metal concentrations in fish liver and fish muscle (mercury), blue mussel soft body and guillemot egg. Analytical methods for metals in liver are performed according to the Swedish standards SS-EN 13805 (Foodstuffs – Determination of trace elements – Pressure digestion) and SS-EN ISO 17294-2 (Water quality – Application of inductively coupled plasma mass spectrometry (ICP-MS) – Part 2: Determination of 62 elements), and for mercury according to the US EPA Method 7473 (mercury in solids and solutions by thermal decomposition, amalgamation and atomic absorption spectrophotometry). The laboratory participates in the periodic QUASIMEME intercalibration rounds.

CRMs (certified reference material) used for mercury are:
DORM-2 and DORM-3 (dogfish muscle)
IAEA-407 (fish tissue)
For all other metals, CRMs used are:
DOLT-2 and DOLT-3 (dogfish liver)
NIST 1566 (oyster tissue).

Due to the change in laboratory and hence analysis methods, an intercalibration has been conducted to provide comparable results for the time series between laboratories.

Results from metal analysis have been compared between the laboratories. For herring from Utlängan, Väderöarna and Fladen, cod from SE Gotland and Fladen and guillemot egg from St. Karlsö the same individuals have been compared. For blue mussel from Fladen, perch from Kvädfjärden and herring from Landsort the comparisons are made on samples from same catch but not the same individuals (due to lack of sufficient sample material).

The metal concentrations analysed by SLU have, in the time series, been recalculated by the ratios between laboratories in cases where these were significantly separated from 1, presented in table 6.1, to make the SLU-data comparable with the results from ITM. No comparison between the laboratories has been done for eelpout. To make the metal results for eelpout analysed at SLU comparable with those analysed at ITM the mean ratio of herring, cod and perch for each metal (table 6.2) have been used for recalculation.
Table 6.1. Ratios of metal concentrations analysed by SLU versus ITM with corresponding standard error. P-values below 0.05 indicate mean ratios separated from 1.

<table>
<thead>
<tr>
<th>Herring</th>
<th>n</th>
<th>Ratio, SLU/ITM</th>
<th>Std.Err.</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>9</td>
<td>0.92</td>
<td>0.16</td>
<td>NS</td>
</tr>
<tr>
<td>Pb</td>
<td>40</td>
<td>1.40</td>
<td>0.12</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Cd</td>
<td>40</td>
<td>1.14</td>
<td>0.06</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Cu</td>
<td>40</td>
<td>0.89</td>
<td>0.03</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Zn</td>
<td>40</td>
<td>1.13</td>
<td>0.03</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Ni</td>
<td>36</td>
<td>1.97</td>
<td>0.20</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Cr</td>
<td>30</td>
<td>2.01</td>
<td>0.30</td>
<td>&lt;0.01</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cod</th>
<th>n</th>
<th>Ratio, SLU/ITM</th>
<th>Std.Err.</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>9</td>
<td>1.19</td>
<td>0.07</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Pb</td>
<td>15</td>
<td>3.06</td>
<td>0.76</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Cd</td>
<td>19</td>
<td>1.85</td>
<td>0.18</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Cu</td>
<td>19</td>
<td>1.35</td>
<td>0.15</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Zn</td>
<td>19</td>
<td>1.30</td>
<td>0.20</td>
<td>NS</td>
</tr>
<tr>
<td>Ni</td>
<td>11</td>
<td>1.38</td>
<td>0.27</td>
<td>NS</td>
</tr>
<tr>
<td>Cr</td>
<td>11</td>
<td>1.42</td>
<td>0.87</td>
<td>NS</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Perch</th>
<th>n</th>
<th>Ratio, SLU/ITM</th>
<th>Std.Err.</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>9</td>
<td>0.95</td>
<td>0.06</td>
<td>NS</td>
</tr>
<tr>
<td>Pb</td>
<td>9</td>
<td>2.31</td>
<td>0.19</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Cd</td>
<td>9</td>
<td>1.50</td>
<td>0.21</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Cu</td>
<td>9</td>
<td>1.35</td>
<td>0.12</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Zn</td>
<td>9</td>
<td>1.21</td>
<td>0.11</td>
<td>NS</td>
</tr>
<tr>
<td>Ni</td>
<td>1</td>
<td>0.76</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Cr</td>
<td>2</td>
<td>8.95</td>
<td>1.05</td>
<td>NS</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mussel</th>
<th>n</th>
<th>Ratio, SLU/ITM</th>
<th>Std.Err.</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>10</td>
<td>1.01</td>
<td>0.15</td>
<td>NS</td>
</tr>
<tr>
<td>Pb</td>
<td>10</td>
<td>1.30</td>
<td>0.20</td>
<td>NS</td>
</tr>
<tr>
<td>Cd</td>
<td>10</td>
<td>0.74</td>
<td>0.07</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Cu</td>
<td>10</td>
<td>0.74</td>
<td>0.05</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Zn</td>
<td>10</td>
<td>0.98</td>
<td>0.08</td>
<td>NS</td>
</tr>
<tr>
<td>Ni</td>
<td>7</td>
<td>0.96</td>
<td>0.15</td>
<td>NS</td>
</tr>
<tr>
<td>Cr</td>
<td>7</td>
<td>2.48</td>
<td>0.47</td>
<td>&lt;0.05</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Guillemot egg</th>
<th>n</th>
<th>Ratio, SLU/ITM</th>
<th>Std.Err.</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>9</td>
<td>1.25</td>
<td>0.07</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Pb</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Cd</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Cu</td>
<td>9</td>
<td>0.95</td>
<td>0.04</td>
<td>NS</td>
</tr>
<tr>
<td>Zn</td>
<td>9</td>
<td>1.15</td>
<td>0.11</td>
<td>NS</td>
</tr>
<tr>
<td>Ni</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Cr</td>
<td>8</td>
<td>9.19</td>
<td>2.05</td>
<td>&lt;0.01</td>
</tr>
</tbody>
</table>
Table 6.2. Herring, cod and perch mean ratios of metal concentrations analysed by SLU versus ITM.

<table>
<thead>
<tr>
<th>mean fish ratios</th>
<th>n</th>
<th>Ratio SLU/ITM</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg</td>
<td>27</td>
<td>1.02</td>
</tr>
<tr>
<td>Pb</td>
<td>64</td>
<td>2.26</td>
</tr>
<tr>
<td>Cd</td>
<td>68</td>
<td>1.50</td>
</tr>
<tr>
<td>Cu</td>
<td>68</td>
<td>1.20</td>
</tr>
<tr>
<td>Zn</td>
<td>68</td>
<td>1.21</td>
</tr>
<tr>
<td>Ni</td>
<td>48</td>
<td>1.67</td>
</tr>
<tr>
<td>Cr</td>
<td>41</td>
<td>1.71</td>
</tr>
</tbody>
</table>

Organochlorines and brominated flame retardants

The analyses of organochlorines and brominated flame retardants are carried out at the Institute of Applied Environmental Science (ITM) at Stockholm University. Specific analytical methods applied are described in the respective chapters where applicable. Before 1988, organochlorines were analysed by a packed column gas chromatography (GC). During 1988, analysis on a capillary column was introduced, allowing analysis of individual congeners (Eriksson et al. 1994). The extraction method originates from the method described by Jensen et al. (Jensen et al. 1983) where wet tissues are extracted with a mixture of polar and non-polar solvents. The organochlorines are analysed on a gas chromatograph (GC) equipped with a μ-electron capture detector (Eriksson et al. 1994). The BFRs are analysed by a GC connected to a mass spectrometer operating in electron capture negative ionization mode (NICI) (Sellström et al. 1998).

Quality assurance

Quality control for organochlorines has continuously improved over the last 20 years, resulting in accreditation in 1999. Assessment is performed once a year by the accreditation body SWEDAC. The laboratory is fulfilling the obligations in SS-EN ICO/IEC 17025:2005. The accreditation is valid for CB28, 52, 101, 118, 153, 138, 180, HCB, p,p′-DDE, p,p′-DDD, p,p′-DDT and α, β- and γ–HCH in biological tissues. So far the BFRs are not accredited but the analysis of BDE-47, 99,100, 153, 154 and HBCD are in many ways performed with the same quality aspects as the organochlorines.

The Quality Assurance program is based on the Quality Manual, standard operation procedures (SOPs) and supplements. The annual audit includes a review of the SOPs, reference materials, proficiency testing, filing system, qualifications of the staff, up-to-date record of the training of the staff (to be able to perform their assigned tasks), accredited methods and audit of the quality program.

Standards

The original of all standards are well documented with known purity and certified concentration with uncertainty for the solutions.

Selectivity

To have the possibility to control impurities in solvents, equipments and glassware, one blank sample is extracted together with each batch of environmental samples.
Coelution of PCB congeners and pesticides in GC analysis is dependent upon instrumental conditions such as column type, length, internal diameter, film thickness and oven temperature. To minimize possible coelutions, two 60 m columns are used in parallel, the commonly used DB-5 and the more polar DB-1701. The only remaining known coelution is for CB-138, which coelutes with CB-163 (Larsen et al. 1990). Therefore CB-138 is reported as CB138+163. PBDE and HBCD are analysed on a 30 m DB-5 MS column, monitoring \( m/\delta \) 79 and 81.

When introducing a new matrix one of the samples is re-extracted with a mixture of more polar solvents for control of no remaining contaminants in the matrix residual.

Samples from new matrixes and samples from already established matrixes from new sampling location are also examined for suitable internal standards.

**Reference Material**

Two laboratory reference materials (LRM) are used as extraction controls, chosen with respect to their lipid content and level of contaminants. The controls consist of herring respectively salmon muscle, homogenised in a household mixer and stored in aliquots in airtight bags of aluminium laminate at -80°C. At every extraction event one extraction control is extracted as well.

The certified reference material CRM 718 (herring muscle) is analysed for PCB once a year.

### 6.1.1 Proficiency testing

Concerning PCBs and pesticides, the laboratory has participated in the periodic QUASIMEME proficiency testing since 1993, with two rounds every year, each one containing two samples. Around 95% of all reported values have been satisfactory according to QUASIMEME, meaning they have been within +/- 2 standard deviation of the assigned value. In 2000, the laboratory participated in the first interlaboratory study ever performed for PBDEs and HBCD, contaminants that since 2001 are incorporated in the QUASIMEME proficiency testing scheme. Around 80% of the values the laboratory has produced during the years have been satisfactory according to QUASIMEME.

### Quantification limits and uncertainty in the measurements

Calculation of the uncertainty in the measurement is based on the Nordtest Report TR 537 “Handbook for calculation of measurement uncertainty in environmental laboratories”, where the within-laboratory reproducibility is combined with estimate of the method and laboratory bias. The within-laboratory reproducibility is calculated from LRM from more than 7000 PCB- and pesticide values during a period of nearly 20 years and around 1500 BDE- and HBCD values during 10 years. The bias is estimated from proficiency testing of more than 8 samples during at least 4 years. Only within-laboratory reproducibility is used for HBCD since no reliable proficiency testing (or certified reference material) exists today. Finally, the expanded uncertainty is calculated, using a coverage factor of 2 to reach approximately 95% confidence level (table 6.3). The reproducibility follows the theory stated by Horwitz where the relative standard deviation increase when the concentration level decrease (Horwitz and Albert, 2006).
Table 6.3. Expanded uncertainty

<table>
<thead>
<tr>
<th>ng/g lw</th>
<th>PBDEs rsd%</th>
<th>HBCD rsd%</th>
<th>PCBs rsd%</th>
<th>HCHs rsd%</th>
<th>DDTs rsd%</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2-1</td>
<td>63</td>
<td>3-10</td>
<td>35</td>
<td>43</td>
<td>40</td>
</tr>
<tr>
<td>1-10</td>
<td>62</td>
<td>10-100</td>
<td>32</td>
<td>38</td>
<td>37</td>
</tr>
<tr>
<td>&gt;10</td>
<td>28</td>
<td>&gt;100</td>
<td>14</td>
<td>10</td>
<td>30</td>
</tr>
<tr>
<td>&gt;4</td>
<td>56</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Quantification limits and other comments are reported under each contaminant description.

6.2 Dioxins, dibenzofurans and dioxin-like PCBs

The analyses of dioxins and dioxin-like PCBs are carried out at the Department of Chemistry, Umeå University. The extraction method is described by Wiberg et al. (1998), the clean-up method by Danielsson et al. (2005), and the instrumental analysis (GC-HRMS) by Liljelind et al. (2003). The laboratory participates in the annual FOOD intercallibration rounds, including laboratory reference material (salmon tissue) with each set of samples.

6.3 Polycyclic Aromatic Hydrocarbons

The analysis of PAHs are carried out at IVL, the Swedish Environmental Research Institute. The extraction and analysis of the samples were performed according to IVLs accredited method for PAHs.

The biota samples were spiked with recovery standard, homogenised in acetone and extracted in an ultrasonic bath. The extract was safeguarded and the samples were extracted once more with acetone and twice with pentane/ether. The extracts were combined and the organic compounds were extracted to an organic phase by liquid/liquid extraction with water and pentane, and further concentrated under nitrogen.

The samples were hydrolysed and pre-treatment procedures, such as fractionation of the organic compounds on silica, were performed as additional "clean-up" procedures. Laboratory blanks followed the same procedures as samples in the analytical work.

Determination of PAH components was carried out using a high performance liquid chromatograph (HPLC, type Varian Prostar 240, M410) with a 5 μm C_{18}-column (Chromosphere PAH 100* 3 mm, Chrompack). A linear gradient elution program was used, starting with acetonitrile/water 50:50 and ending with 100% acetonitrile (Rathburne HPLC-grade) at a flow rate of 1 ml min^{-1}. A fluorescence detector (Varian Prostar 363) with a wavelength program optimised for each PAH was used for quantification. The peak heights were registered with a chromatographic system from Varian (Star). The concentrations of 16 different PAH compounds were calculated by comparison to a certified standard, NIST, SRM 1647.

All of the standards used (both internal standard and quantification standards) are certified with known purity and precision.
6.4 Perfluorinated substances
The analyses of perfluorinated substances are carried out at the Analytical Environmental Chemistry Unit at ITM, University of Stockholm.

6.4.1 Sample preparation and instrumental analysis
A sample aliquot of approximately 1.2 g homogenized tissue in a polypropylene (PP)-centrifuge tube was spiked with 10 ng each of a suite of mass-labelled internal standards (\(^{18}\)O- or \(^{13}\)C-labelled perfluorinated sulfonates and carboxylic acids). The samples were extracted twice with 5 mL of acetonitrile in an ultrasonic bath. Following centrifugation, the supernatant extract was removed and the combined acetonitrile phases were concentrated to 1 mL under a stream of nitrogen. The concentrated extract underwent dispersive clean-up on graphitised carbon and acetic acid. A volume of 0.5 mL of the cleaned-up extract was added to 0.5 mL of aqueous ammonium acetate. Precipitation occurred and the extract was centrifuged before the clear supernatant was transferred to an autoinjector vial for instrumental analysis and the volume standards BTPA and bPFDcA were added.

Aliquots of the final extracts were injected automatically on a high performance liquid chromatography (HPLC) system (Alliance 2695, Waters) coupled to a tandem mass spectrometer (MS-MS; Quattro II, Micromass). Compound separation was achieved on an Ace 3 C\(^{18}\) column (150 x 2.1 mm, 3 \(\mu\)m particles, Advanced Chromatography Technologies) with a binary gradient of ammonium acetate buffered methanol and water. The mass spectrometer was operated in negative electrospray ionisation mode with the following optimised parameters: Capillary voltage, 2.5 kV; drying and nebuliser gas flow (N\(_2\)), 300 and 15 L/h, respectively; desolvation and source temperature, 150 and 120 \(^\circ\)C, respectively. Quantification was performed in selected reaction monitoring chromatograms using the internal standard method.

6.4.2 Quality control
The extraction method employed in the present study (with the exception of the concentration step) has previously been validated for biological matrices and showed excellent analyte recoveries ranging between 90 and 110% for PFCAs from C6 to C14 (Powley & Buck 2005). Including extract concentration, we determined recoveries between 70 and 90% for C6- to C10-PFCAs and 65 – 70% for C11-C14 PFCAs. Extraction efficiencies for perfluorosulfonates (PFSAs), including perfluorooctane sulfonamide (PFOSA), were determined to 70 – 95%. Furthermore, mean method recoveries of the mass labelled internal standard compounds were between 53% and 106%. Method quantification limits (MQLs) for all analytes were determined on the basis of blank extraction experiments and ranged between 0.15 and 0.8 ng/g wet weight for the different compounds. A fish tissue sample used in an international inter-laboratory comparison (ILC) study in 2007 (van Leeuwen et al. 2009) was analysed along with the samples. The obtained concentrations deviated from the mean concentration from the ILC study by less than 11% for all seven compounds quantified in the ILC.
7 Statistical treatment, graphical presentation

7.1 Trend detection
One of the main purposes of the monitoring programme is to detect trends. The trend detection is carried out in three steps.

7.1.1 Log-linear regression analyses
Log-linear regression analyses are performed for the entire investigated time period and also for the most recent 10 years for the longer time series.

The slope of the line describes the yearly percentage change. A slope of 5% implies that the concentration is halved in 14 years, whereas a slope of 10% corresponds to a similar reduction in 7 years, and 2% in 35 years. (Table 7.1).

<table>
<thead>
<tr>
<th>Increase</th>
<th>1%</th>
<th>2%</th>
<th>3%</th>
<th>4%</th>
<th>5%</th>
<th>7%</th>
<th>10%</th>
<th>12%</th>
<th>15%</th>
<th>20%</th>
</tr>
</thead>
<tbody>
<tr>
<td>70</td>
<td>35</td>
<td>24</td>
<td>18</td>
<td>14</td>
<td>10</td>
<td>7</td>
<td>6</td>
<td>5</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>Decrease</td>
<td>69</td>
<td>35</td>
<td>23</td>
<td>17</td>
<td>14</td>
<td>10</td>
<td>7</td>
<td>6</td>
<td>4</td>
<td>3</td>
</tr>
</tbody>
</table>

7.1.2 Non-parametric trend test
The regression analysis assumes, among other things, that the regression line gives a good description of the trend. The leverage affect of points at the end of the line is also a well-known fact. An exaggerated slope, caused 'by chance' by a single or a few points at the end of the line, increases the risk of a false significant result when no real trend exist. A non-parametric alternative to the regression analysis is the Mann-Kendall trend test (Gilbert 1987; Helsel & Hirsch 1995; Swertz 1995). This test generally has lower power than the regression analysis, and does not take into account differences in magnitude of concentrations; it only counts the number of consecutive years where the concentration increases or decreases compared with the year before. If the regression analysis yields a significant result but the Mann-Kendall test does not, the explanation could be either that the latter test had lower power, or that the influence of end points in the time series has become unjustifiably high on the slope. Hence, the eighth line reports Kendall’s ‘τ’, and the corresponding p-value. The Kendall’s ‘τ’ range from 0 to 1 like the traditional correlation coefficient ‘r’, but will generally be lower. ‘Strong’ linear correlations of 0.9 or above correspond to τ-values of about 0.7 or above (Helsel & Hirsch 1995, p. 212). This test was recommended by the US Environmental Protection Agency (EPA) for use in water quality monitoring programmes with annual samples, in an evaluation comparing several other trend tests (Loftis et al. 1989).
7.1.3  Non-linear trend components
In order to describe development over time, an alternative to the regression line is a type of smoothed line. The smoother applied here is a simple 3-point running mean smoother fitted to the annual geometric mean values. In cases where the regression line is a poor fit, the smoothed line may be more appropriate. The significance of this line is tested by means of an Analysis of Variance, where the variance is explained by the smoother line, and the regression line is compared with the total variance. This procedure has been used in assessments at ICES and is described by Nicholson et al. (1995).

7.2  Adjustments for covariables
It has been shown that metal concentrations in cod liver are influenced by fat content (Grimås et al. 1985). Consequently, the metal concentrations in cod liver are adjusted for fat content. On some occasions (when the average fat content differs between years) this is of major importance and might change the direction of the slope and decrease the between-year variation considerably. For the same reasons, organochlorines in spring-caught herring muscle tissue are adjusted for fat content (Bignert et al. 1993) where appropriate (indicated in the header text of the figures).

7.3  Outliers and values below the quantification limit
Observations further from the regression line than expected from the residual variance around the line are subject to special concern. These deviations may be caused by an atypical occurrence of something in the physical environment, a change in pollution load, or errors in the sampling or analytical procedure. The procedure used to detect suspected outliers in this report is described by Hoaglin and Welsch (1978). It makes use of the leverage coefficients and the standardised residuals. The standardised residuals are tested against a \(t_{0.05}\) distribution with \(n-2\) degrees of freedom. When calculating the \(i\)th standardised residual the current observation is left out, implying that the \(i\)th observation does not influence the slope or the variance around the regression line. The suspected outliers are merely indicated in the figures and are included in the statistical calculations except in a few cases, as indicated in the figures.

Values reported that are below the quantification limit are substituted using the reported LOQ (or in case when this information is missing the minimum value for the current year) divided by the square root of 2.

7.4  Plot Legends
The analytical results from each of the investigated elements are displayed in figures. A selection of sites and species are presented in the plots; no time series are shorter than four years.

The plot displays the geometric mean concentration of each year (circles) together with the individual analyses (small dots) and the 95% confidence intervals of the geometric means.

The overall geometric mean value for the time series is depicted as a horizontal, thin line.

The trend is presented by one or two regression lines (plotted if \(p < 0.10\), two-sided regression analysis); one for the whole time period in red and one for the last ten years in
pink (if the time series is longer than ten years). Ten years is often too short a period to statistically detect a trend unless it is of considerable magnitude. Nevertheless, the ten year regression line will indicate a possible change in the direction of a trend. Furthermore, the residual variance around the line compared to the residual variance for the entire period will indicate if the sensitivity has increased as a result of, for example, improved sampling techniques or that problems in the chemical analysis have disappeared.

A smoother is applied to test for non-linear trend components (see section 7.1.3). The smoothed line in blue is plotted if \( p < 0.10 \). A broken or dashed line segment indicates a gap in the time series with a missing year.

The log-linear regression lines fitted through the geometric mean concentrations follow smooth exponential functions.

A cross inside a circle, indicates a suspected outlier (see section 7.3). Suspected outliers are indicated in the figures and are included in the statistical calculations, except in a few cases, and pointed out in the figures.

Each plot has a header with species name, age class and sampling locality. Age class may be replaced by shell length for blue mussels. Sampling locality is in a few cases in coded form to save space; C1=herring, Harufjärden; C2=herring, Ångskärsklubb; C3=herring, Landsort; C4=herring, Utlängan; C6=herring, Fladen; V2=spring caught herring, Ångskärsklubb; V4=spring caught herring, Karlskrona archipelago; U8=guillemot egg, St Karlsö; G5=cod, south east of Gotland; G6=cod, Fladen; P2=perch, Kvädöfjärden; M3=blue mussel, Väderöarna; L6=dab, Fladen; P3=flounder, Väderöarna. Below the header of each plot the results from several statistical calculations are reported:

\[ T_v = \ldots \text{lp\% or dp\%} = \ldots \] \( T_v \) is the target level (see Chapter 10) calculated on a lipid weight base (\( \text{lp\%} \)) or on dry weight base (\( \text{dp\%} \)), original target value was given on a wet weight basis.

\[ n(\text{tot})= \] The first line reports the total number of analyses included together with the number of years (\( n(\text{yrs})= \)).

\[ m= \] The overall geometric mean value together with its 95\% confidence interval is reported on the second line of the plot (N.B. d.f. = n of years - 1).

\[ \text{slope}= \] reports the slope, expressed as the yearly percentage change together with its 95\% confidence interval.

\[ \text{sd(lr)}= \] reports the coefficient of variation around the regression line as a measure of between-year variation, together with the lowest detectable change in the current time series with a power of 80\%, one-sided test, \( \alpha=0.05 \). The last figure on this line is the estimated number of years required to detect an annual change of 10\% with a power of 80\%, one-sided test, \( \alpha=0.05 \).

\[ \text{power}= \] reports the power to detect a log-linear trend in the time series (Nicholson & Fryer, 1991). The first figure represents the power to detect an annual change of 10\% with the number of years in the current time series. The second figure is the power estimated as if the slope were 10\% a year and the number of years were ten. The third figure is the lowest detectable change (given in percent per year) for a ten year period, with the current
between-year variation at a power of 80%. The results of the power analyses from the various time series are summarised in chapter nine.

\( r^2 \) reports the coefficient of determination \((r^2)\) together with a p-value for a two-sided test \( (H_0: \text{slope} = 0) \) i.e. a significant value is interpreted as a true change, provided that the assumptions of the regression analysis are fulfilled.

\( y(09) = \) reports the concentration estimated from the regression line for the last year together with a 95% confidence interval, e.g. \( y(09) = 2.55(2.17, 3.01) \) is the estimated concentration for the year 1996, where the residual variance around the regression line is used to calculate the confidence interval. Provided that the regression line is relevant to describe the trend, the residual variance might be more appropriate than the within-year variance in this respect.

\( \tau_{ao} = \) reports Kendall’s ‘\( \tau \)’, and the corresponding p-value.

\( \text{sd(sm)} = \) reports the coefficient of variation around the smoothed line. The significance of this line could be tested by means of an Analysis of Variance (see section 7.1.3). The p-value is reported for this test. A significant result will indicate a non-linear trend component. After the p-value, the minimum trend (%/year), likely to be detected, at a power of 80%, during a period of 10 years, should a log-linear trend occur, is shown. This estimate is compensated for by the loss of degrees of freedom, considering the smoother.

Below these nine lines are additional lines with information concerning the regression of the last 10 years.

In some cases where an extreme outlying observation may decrease confidence in the regression line, the ordinary regression line is replaced by the ‘Kendall-Theil Robust line’, (see Helsel & Hirsch 1995, p. 266). In these cases only the ‘Theil’-slope and Kendall’s ‘\( \tau \)’ are reported.

**7.5 Legend for the three dimensional maps**

The height of the bars represents the arithmetic mean for the last three years or less if results are not available.
8 The power of the programme

Before starting to interpret the results from the statistical analyses of the time series, it is essential to know with what power temporal changes can be detected (i.e. the chance to reveal true trends with the investigated matrices). It is crucial to know whether a negative result from a trend test indicates a stable situation or if the monitoring programme is too poor to detect even serious changes in the contaminant load in the environment. One approach to this problem is to estimate the power of the time series based on the ‘random’ between-year variation. Alternatively, the lowest detectable trend could be estimated at a fixed power to represent the sensitivity of the time series.

The first task would thus be to estimate the ‘random’ between-year variation. In the results presented below, this variation is calculated using the residual distance from a log-linear regression line. In many cases the log-linear line, fitted to the current observations, seems to be an acceptable ‘neutral’ representation of the true development of the time series. In cases where a significant ‘non-linear’ trend has been detected (see above), the regression line may not serve this purpose; hence the sensitivity- or power-results based on such time series are marked with an asterix in the tables below. These results are also excluded from estimations of median performances.

Another problem is that a single outlier could ruin the estimation of the between-year variation. As an example, the time series of lead concentrations in fish liver seem to suffer from occasional outliers, especially in the beginning of the investigated period, 1981 - 1984. The estimated median sensitivity of these series is 12.5% a year. If a few outliers, identified by means of objective statistical criterias are deleted, the calculated median sensitivity improves to 5.8%. In the presented results, suspected outliers are included, which means that the power and sensitivity might be underestimated.
The number of years that various contaminants have been analysed and detected from the monitored sites is reported in table 8.1. Generally the monitoring of trace metals has continued for about 25 - 30 years; PCB and DDT for about 25 - 30 years (spring-caught herring and guillemot egg however, for more than 35 years; HCH, HCB and PCDD/PCDF for about 20 years; PBDE/HBCDD for about 10 years; and PFCs only for about 5 years).

**Table 8.1. Number of years that various contaminants have been analysed.**

|                 | C1   | C2   | V2   | C3   | C4   | V4   | C6   | C7   | G5   | G6   | P1   | P2   | Z1   | Z2   | Z3   | M2   | M6   | M3   | L6   | P3   | U8   |
|-----------------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| **Hg**          | 29   | 29   | 18   | 30   | 30   | 18   | 30   | 18   | 31   | 31   | 15   | 15   | 13   | 27   | 29   | 14   | 15   | 13   | 27   | 29   | 14   | 15   | 37   |
| **Pb**          | 27   | 27   | 14   | 29   | 29   | 13   | 29   | 14   | 28   | 28   | 14   | 13   | 11   | 15   | 14   | 14   | 26   | 14   | 14   | 14   | 14   | 14   | 14   |
| **Cd**          | 28   | 28   | 14   | 29   | 29   | 13   | 29   | 14   | 25   | 29   | 14   | 14   | 11   | 15   | 14   | 14   | 14   | 28   | 14   | 14   | 14   | 14   | 14   |
| **Ni**          | 15   | 15   | 14   | 15   | 15   | 13   | 15   | 14   | 14   | 15   | 14   | 11   | 15   | 14   | 14   | 14   | 15   | 14   | 15   | 14   | 14   | 14   | 14   |
| **Cr**          | 15   | 15   | 14   | 15   | 15   | 13   | 15   | 14   | 15   | 15   | 14   | 11   | 14   | 14   | 14   | 15   | 15   | 14   | 14   | 14   | 14   | 14   | 14   |
| **Cu**          | 28   | 28   | 14   | 29   | 29   | 13   | 29   | 14   | 29   | 29   | 14   | 11   | 15   | 14   | 14   | 14   | 27   | 14   | 14   | 14   | 14   | 14   | 14   |
| **Zn**          | 27   | 27   | 14   | 28   | 28   | 13   | 28   | 12   | 28   | 28   | 14   | 14   | 11   | 15   | 14   | 14   | 14   | 27   | 14   | 14   | 14   | 14   | 14   |
| sPCB            | 30   | 29   | 36   | 31   | 30   | 35   | 30   | -    | -    | -    | -    | -    | -    | -    | -    | -    | 24   | 25   | 13   | 15   | 39   |      |
| CB-153          | 21   | 21   | 21   | 23   | 22   | 21   | 22   | 14   | 21   | 20   | 15   | 22   | 11   | 15   | 15   | 15   | 22   | 21   | 5    | 6    | 22   |      |
| DDE             | 30   | 29   | 36   | 31   | 30   | 35   | 30   | 14   | 29   | 29   | 23   | 27   | 11   | 15   | 15   | 15   | 25   | 25   | 14   | 15   | 39   |      |
| α-HCH           | 21   | 20   | 21   | 23   | 22   | 20   | 22   | 13   | 21   | 22   | 15   | 21   | 11   | 15   | 15   | 15   | 22   | 21   | 6    | 6    | 22   |      |
| HCB             | 21   | 19   | 21   | 22   | 22   | 21   | 22   | 13   | 21   | 20   | 15   | 22   | 11   | 15   | 15   | 15   | 19   | 19   | 6    | 6    | 23   |      |
| TCDD-eqv        | 19   | 26   | -    | -    | 19   | -    | 20   | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | 35   |      |
| BDE-47          | 11   | 10   | 8    | 11   | 11   | 7    | 11   | 10   | 11   | 11   | -    | -    | -    | 9    | 10   | 10   | -    | -    | 35   |      |
| HBCDD           | 11   | 10   | 8    | 11   | 11   | 7    | 11   | 10   | 11   | 11   | -    | -    | -    | 9    | 10   | 10   | -    | -    | 36   |      |

PFCs
The number of years required to detect an annual change of 10% with a power of 80% is shown in table 8.2. Power is to a great extent dependent on the length of the time series. The possibility to statistically verify an annual change of 10% at a power of 80% generally requires 8-12 years for the organic substances.

Table 8.2. Number of years required to detect an annual change of 10% with a power of 80%. C1=herring, Harufjärden; C2=herring, Ängskärsklubb; C3=herring, Landsort; C4=herring, Utlängan; C6=herring, Fladen; V2=spring caught herring, Ängskärsklubb; V4=spring-caught herring, Karlshkrona archipelago; U8=guillemot egg, St Karlsö; G5=cod, south east of Gotland; G6=cod, Fladen; P1=Holmöarna, P2=perch, Kvädöfjärden; M2=blue mussel, Kvädöfjärden; M6=blue mussel, Fladen; M3=blue mussel, Väderöarna; L6=dab, Fladen; P3=flounder, Väderöarna.

### Mercury
Based on geometric means on a fresh weight basis

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### Organochlorines, bromines, flourines
Based on geometric means on a lipid weight basis, fresh weight for PFOS

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* indicates a significant non-linear trend component
The smallest trend able to be detected within a 10 year period with a power of 80% is presented both for the entire time series and for the latest 10 year period (Table 8.3). The table shows that the sensitivity for lead and cadmium is approximately the same (10% - 20%), whereas for zinc and copper it is somewhat better (5 - 10%). For PCB, DDE, HCH and HCB the estimated sensitivity is about 6 - 12%. For the TCDD-eqv the estimated median sensitivity is 12%, and for BDE-47 and HBCDD 10 - 16%. Biological variables such as the condition index for herring, cod and perch show a sensitivity of about 1 - 2%.

Table 8.3. Lowest detectable trend within a 10 year period with a power of 80% for different variables in various matrices at different sites. The top row for every substance gives the figure based on the residual variance for the whole period, whereas the bottom row gives the figure for the last 10 years of the time series. If no figure is given for the last 10 year period, this indicates that the time series show a significant non-linear trend component. The figure for the whole time period is calculated considering the non-linear trend component.

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Based on geometric means on a fresh weight basis

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### Organochlorines, bromines, flourines
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The power to detect an annual change of 10% covering the monitoring period, i.e. the length of the time series, varies depending on site and investigated contaminant (table 8.4). For the long time series, the estimated power is in most cases close to 100%. For the shorter time series of BDE-47 and HBCDD, estimated power is about 30-100%.

Table 8.4. Power to detect an annual change of 10% covering the entire monitoring period. The length of the time series varies depending on site and investigated contaminant. Where a considerable increase in power has been achieved during the most recent 10 year period, this value has been used instead.

**Mercury**
Based on annual geometric mean concentrations on a fresh weight basis

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**Other trace metals**
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**Organochlorines, bromines, flourines**
Based on geometric means on a lipid weight basis, fresh weight for PFOS

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<th>M6</th>
<th>M3</th>
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<td>.16</td>
<td>.90</td>
<td>*.94</td>
</tr>
<tr>
<td>HBCDD</td>
<td>.40</td>
<td>.32</td>
<td>.27</td>
<td>.92</td>
<td>.60</td>
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</tbody>
</table>

* indicates a significant non-linear trend component
9 Pollutant regulation: conventions and legislation

9.1 The Stockholm Convention on Persistent Organic Pollutants

The Stockholm Convention on Persistent Organic Pollutants (POPs) is an international agreement requiring measures for reducing or preventing release of dangerous substances into the environment. The Stockholm Convention was adopted in 2001 and entered into force in 2004. The convention deals with organic compounds that are persistent and remain in the environment for a long time, have a potential for long-range transport, bioaccumulate in fatty tissue in organisms and have adverse effects on human health or the environment. Initially, 12 chemicals were included in the treaty in 2001 (aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, mirex, toxaphene, PCB, hexachlorobenzene, polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans). In May 2009, an amendment was adopted into the convention, and nine additional chemicals were listed as POPs (hexa-/heptabromdiphenylether, tetra-/pentabromodiphenylether, chlordecone, hexabromobiphenyl, lindane, α- and β–hexachlorocyklohexane, pentachlorobenzen and PFOS). Three more substances have been nominated to be included on the list, and are currently under review by the Persistent Organic Pollutants Review Committee (short-chained chlorinated paraffins, endosulfan and hexabromocyclododecane) (www.pops.int).

9.2 The Helsinki Convention

The Helsinki Convention is the Convention on the Protection of the Marine Environment of the Baltic Sea. It was signed in 1992 by all the states bordering the Baltic Sea, entered into force in 2000, and is governed by the Helsinki Comission (HELCOM). The main focus of the convention is to protect the marine environment of the Baltic Sea from all sources of pollution, with the future vision being a healthy Baltic Sea. The Baltic Sea Action Plan (BSAP) is a program within HELCOM that aims to restore a good ecological status of the marine environment by 2012. Joint monitoring of pollutants in the Baltic Sea is important to evaluate the status of the Baltic Sea. Data from the Swedish national monitoring program is reported to HELCOM every year via the International Council for the Exploration of the Sea (ICES) (www.helcom.fi).

9.3 The Oslo Paris Convention

The convention for the protection of the marine environment of the North-East Atlantic (The Oslo Paris Convention, OSPAR) was adopted in 1992 after a meeting of The Oslo and The Paris Commissions, and entered into force in 1998. Within OSPAR, six different working areas have been identified that address the main areas of concern (the Biodiversity and Ecosystem Strategy, the Eutrophication Strategy, the Hazardous Substances Strategy, the Offshore Industry Strategy, the Radioactive Substances Strategy and a Strategy for the Joint Assessment and Monitoring Programme). The OSPAR Hazardous Substances Strategy works to prevent pollution of the marine environment. The aim is to achieve levels
near background concentrations for naturally occurring substances, and close to zero for man-made synthetic substances. The hazardous substances work is implemented by OSPAR’s Hazardous Substances Committee, which is working to achieve this goal by 2020. Within OSPAR, hazardous substances are defined as substances that are persistent, bio accumulative and toxic (PBT). OSPAR has a list of chemicals of priority concern, and a list of chemicals of possible concern. These lists are continuously being updated as knowledge on the substances is improved. Data from the Swedish national monitoring program is reported to OSPAR every year through ICES (www.ospar.org).

9.4 The Convention on Long-Range Trans boundary Air Pollution

The Convention on Long Range Trans boundary Air Pollution (CLRTAP) was initiated in 1972 at a United Nations Conference on the Human Environment in Stockholm. After the scientific findings that acidification in Swedish lakes was caused by sulphur emission from continental Europe, the necessity for international measures to reduce emissions to air that had environmental effects far from the source, was addressed. In 1979 the convention was signed in Geneva, and entered into force in 1983. Initially, the convention focused on sulphuric compounds causing acidification, but later eight protocols were added for other groups of substances e.g. nitrogen oxides, volatile organic compounds (VOCs) and persistent organic pollutants (POPs) (http://www.unece.org/env/lrtap/lrtap_h1.htm).

9.5 EU chemical legislation

9.5.1 REACH

REACH is the EU chemicals policy that entered into force on the 1st of June 2007 (EC 1907/2006). REACH stands for Registration, Evaluation, Authorization and Restriction of Chemical Substances. The policy places more responsibility on industry, and importers and users have to gather information about their chemicals which they then report to the European Chemicals Agency (ECHA) based in Helsinki. ECHA manages REACH by gathering information and keeps databases of chemicals used in the EU. (http://ec.europa.eu/environment/chemicals/reach/reach_intro.htm).

9.5.2 RoHS directive

The Directive on the Restriction of Hazardous Substances (RoHS) was adopted in February 2003. The RoHs directive reduces the use of six chemical substances in electrical or electronic products that were released on the market after July 2006. These substances are mercury, cadmium, lead, chromium VI, polybrominated biphenyls and polybrominated diphenyl ethers. The maximum allowed amount of these substances (based on weight) is 0.01% for cadmium, and 0.1% for the other substances. (http://www.kemi.se/templates/Page____3794.aspx).
9.5.3 Water Framework Directive

The Water Framework Directive (WFD) aims to achieve good ecological and chemical status of all surface waters and ground water bodies in the EU by 2015. WFD was adopted in October 2000, and deals with fresh water as well as coastal-zone and estuary waters. Within the WFD, a list of 33 prioritized substances has been established, and later eight additional substances were added. To evaluate if “good chemical status” has been achieved, threshold values or Environmental Quality Standards (EQS) have been established for the listed substances (see chapter 10). It is the responsibility of each member state to assess and report if the goal has been fulfilled. (http://ec.europa.eu/environment/water/water-framework/index_en.html).

9.5.4 Marine Strategy Framework Directive

The Marine Strategy Framework Directive (MSFD) was adopted in 2008 with the aim of achieving good environmental status in all European marine waters by 2020. Two of eleven descriptors that have been identified for good environmental status deal with contaminants. These are “contaminants and pollution effects” and “contaminants in fish and other sea food”. The implementation for Swedish waters will be based on the regional international conventions of OSPAR and HELCOM. (http://ec.europa.eu/environment/water/index_en.htm).

9.6 Swedish chemical legislation

One of the 16 Swedish environmental quality objectives is “A non-toxic environment”, which means that concentrations of non-naturally occurring substances should be close to zero, and naturally occurring substances should be close to background concentrations. Their impact on human health and ecosystems should be negligible (http://www.miljomal.se/4-Giftfri-miljo/Definition/). The agency responsible for coordinating this work is the Swedish Chemicals Agency (KEMI). The Swedish chemical legislation is following the EU legislations. Much of the national legislations that existed before June 2007 were replaced by REACH. (http://www.kemi.se/templates/Page____3064.aspx).
10 Target levels for chemical status assessment

Good Environmental Status (GES), in accordance with the Marine Strategy Framework Directive 2008/56/EC (MSFD), is defined as “concentrations of contaminants at levels not giving rise to pollution effects”. GES is determined from quality assessments based on target levels representing a threshold that should not be exceeded. Established to protect sensitive organisms from the harmful effects of hazardous substances, target levels have been developed within several groups or conventions (the EU, OSPAR etc.). Target levels used to assess chemical status in this report have been selected based on the following requirements (for more detailed information see Boalt et al. 2011):

1) Primarily, internationally agreed target levels such as Environmental Quality Standards (EQS) or recommendations for foodstuffs developed within the EU and Environmental Assessment Criteria (EAC) developed by OSPAR, are used;
2) EQSs are prioritized before EACs;
3) If recommendations for foodstuffs are lower than EQSs, these are preferred;
4) If no agreed EQS or EAC are available, QS are used if these are lower than foodstuff recommendations;
5) If reliable target levels have been produced with specific regard to Swedish environmental conditions, these are selected prior to internationally agreed target levels (e.g. HCH);
6) Only one type of target level is applied within each substance group (e.g. we do not mix EQS and EACs depending on availability for different PCB or PAH congeners).

For substances where internationally agreed target levels for chemical status classification in biota are not available, concentrations are presented without evaluation against target levels (e.g Cu, Zn, As, Ag).
Table 10.1. Target levels for various environmental pollutants.

<table>
<thead>
<tr>
<th>Group of substance</th>
<th>Target levels</th>
<th>Fish (µg/kg ww)</th>
<th>Mussels (µg/kg dw)</th>
<th>Background reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metals</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cadmium</td>
<td></td>
<td>160</td>
<td>EC, QS sec. poisn.³</td>
<td></td>
</tr>
<tr>
<td>Lead</td>
<td></td>
<td>300</td>
<td>EC, food stuffs¹</td>
<td></td>
</tr>
<tr>
<td>Mercury</td>
<td></td>
<td>20</td>
<td>EC, QS sec. poisn.³</td>
<td></td>
</tr>
<tr>
<td>Nickel</td>
<td></td>
<td>670</td>
<td>EC, QS human health²</td>
<td></td>
</tr>
<tr>
<td>PAHs</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fluoranthene</td>
<td></td>
<td>110</td>
<td>OSPAR EAC</td>
<td></td>
</tr>
<tr>
<td>Anthracene</td>
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<td>OSPAR EAC</td>
<td></td>
</tr>
<tr>
<td>Naphthalene</td>
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<td>OSPAR EAC</td>
<td></td>
</tr>
<tr>
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<td>1700</td>
<td>OSPAR EAC</td>
<td></td>
</tr>
<tr>
<td>Pyrene</td>
<td></td>
<td>100</td>
<td>OSPAR EAC</td>
<td></td>
</tr>
<tr>
<td>Benzo(a)anthracene</td>
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<td>Benzo(aj)pyrene</td>
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<td>600</td>
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<tr>
<td>Benzo(ghi)perylene</td>
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</tr>
<tr>
<td>Pesticides</td>
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<td></td>
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<td>HCH (incl. lindane)</td>
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<td>IVL ⁴</td>
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<td>PCBs</td>
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<td>CB-138</td>
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<td>0.09</td>
<td>OSPAR BAC</td>
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</tr>
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<td>CB-153</td>
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<td>OSPAR BAC</td>
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</tr>
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<td>CB-156</td>
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<td>OSPAR BAC</td>
<td></td>
</tr>
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<td>0.11</td>
<td>OSPAR BAC</td>
<td></td>
</tr>
<tr>
<td>Other</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>ΣPCDDs+PCDFs</td>
<td></td>
<td>0.23 ng/kg</td>
<td>EC, QS sec. poisn.³</td>
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<tr>
<td>General ng WHO98-TEQ</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BDEs (congeners 28, 47, 99, 100, 153, and 154)</td>
<td>274 ng/g</td>
<td>EC, QS human health²</td>
<td></td>
<td></td>
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<tr>
<td>HCB</td>
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<td>9.7</td>
<td>EC, QS human health²</td>
<td></td>
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<td>EC, QS human health²</td>
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<td>167</td>
<td>EC, QS sec. poisn.³</td>
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</table>

1) Maximum levels in foodstuffs
2) Food uptake by man via fishery products
3) Protection against secondary poisoning, amount / prey tissue
4) EQS set for water translated to concentrations in biota
### 11 Condition

The stoutness of fish, i.e. weight versus length, is a common measure of the ‘degree of well-being’ of an individual or a population.

In this report the commonly used ‘condition factor’, $K$, (Vibert & Lagler, 1961) is used:

$$K = 100 \frac{W}{L^3}$$

where weight ($W$) is given in grams and length ($L$) in centimetres.

#### 11.1 Spatial variation

Average condition factor, estimated over a period of more than 30 years, was slightly lower in herring sampled at Harufjärden (Fig. 11.1) in the northern parts of the Bothnian Bay compared to samples from Fladen and Väderöarna on the Swedish west coast (Fig. 11.2).

The average condition factor, estimated for the whole time period, was slightly higher for eelpout from Väderöarna than for eelpout from the two Baltic sampling sites.

#### 11.2 Temporal variation

Significant decreasing trends in herring condition factor were observed from Harufjärden, Landsort and Utlängan (autumn and spring) (Fig. 11.1, 11.2). At Ängskärsklubb, it increased in spring-caught herring for the whole time series. The increase at Ängskärsklubb may be explained by an unintentional increase in average age over time in the collected samples (Fig. 11.2).

The condition factor estimated for cod showed a significant increasing trend at both Gotland and Fladen over the whole period examined (Fig. 11.3). The observed increase might be explained by the simultaneous decrease in population density during the period examined.

Significantly decreasing trends in both perch and eelpout condition factor were observed at Holmöarna (0.24% and 1.1% respectively) (Fig. 11.3, 11.4).
## Condition factor, herring

### Harufjärden (3-5)

| $n(\text{tot}) = 639$, $n(\text{yrs}) = 30 | m = 0.628 (0.614, 0.643) | \text{slope} = -0.45 \% (0.62, -0.27) | \text{power} = 1.01, 1.01, 1.01 | y(09) = 0.617 (0.569, 0.660) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Angskärsklubb (2-4), spring

| $n(\text{tot}) = 590$, $n(\text{yrs}) = 32 | m = 0.631 (0.612, 0.651) | \text{slope} = 0.40 \% (-0.15, 0.65) | \text{power} = 1.27, 0.039 | y(08) = 0.679 (0.644, 0.717) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Landsort (3-5)

| $n(\text{tot}) = 512$, $n(\text{yrs}) = 31 | m = 0.647 (0.630, 0.664) | \text{slope} = -0.74 \% (-0.95, -0.53) | \text{power} = 1.01, 1.01, 1.01 | y(09) = 0.609 (0.583, 0.638) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Utlangan (2-4)

| $n(\text{tot}) = 624$, $n(\text{yrs}) = 30 | m = 0.652 (0.633, 0.672) | \text{slope} = -0.67 \% (-1.0, 0.67) | \text{power} = 1.01, 1.01, 1.01 | y(09) = 0.586 (0.566, 0.607) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Figure 11.1. Condition factor for herring from Harufjärden, Angskärsklubb, Landsort and Utlangan (time series starting in 1978).

## Condition factor, herring

### Angskärsklubb (2-4), spring

| $n(\text{tot}) = 590$, $n(\text{yrs}) = 32 | m = 0.631 (0.612, 0.651) | \text{slope} = 0.40 \% (-0.15, 0.65) | \text{power} = 1.27, 0.039 | y(08) = 0.679 (0.644, 0.717) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Karlskrona (2-4), spring

| $n(\text{tot}) = 651$, $n(\text{yrs}) = 35 | m = 0.655 (0.644, 0.666) | \text{slope} = -0.15 \% (-0.30, -0.01) | \text{power} = 1.01, 0.01, 0.01 | y(09) = 0.679 (0.644, 0.667) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Fladen (2-3)

| $n(\text{tot}) = 675$, $n(\text{yrs}) = 30 | m = 0.675 (0.644, 0.717) | \text{slope} = -0.70 \% (-2.1, 0.75) | \text{power} = 1.28, 0.035 | y(09) = 0.694 (0.667, 0.723) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Väderöarna

| $n(\text{tot}) = 288$, $n(\text{yrs}) = 14 | m = 0.739 (0.719, 0.760) | \text{slope} = -0.70 \% (-2.1, 0.75) | \text{power} = 1.28, 0.035 | y(09) = 0.715 (0.710, 0.739) |
| 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 0.9 | 1.0 | 1.1 | 1.2 | 1.3 |

### Figure 11.2. Condition factor for herring from Angskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1972, 1972, 1980 and 1994 respectively).
Figure 11.3. Condition factor for cod and perch from south east Gotland and Fladen (cod); and Holmöarna and Kvädöfjärden (perch) (time series starting in 1978 and 1980 respectively).

Figure 11.4. Condition factor for eelpout from Holmöarna, Kvädöfjärden and Väderöarna (time series starting in 1995, 1995 and 1988 respectively).
Fat content is determined in samples that are analysed for organochlorines i.e. herring, eelpout (dab and flounder) muscle, cod liver, blue mussel soft body and guillemot egg. A strong negative correlation between organochlorine concentration (expressed on a fat weight basis) and fat content in spring-caught herring has been shown (Bignert et al. 1993), but also between the concentration of various metals and fat content in cod liver (Grimås et al. 1985). The analysed concentrations of these contaminants were therefore adjusted for varying fat content.

In general, an extremely low fat content due to, for example starvation, may cause elevated concentrations of organochlorines expressed on a fat weight basis.

The sample fat content is determined after extraction with acetone and hexane with 10% ether without heating (Jensen et al. 1983) in the present investigation. Results of the fat determination may vary considerably depending on the extraction method used.

In herring muscle tissue, the subcutaneous fat layer was removed before samples were prepared. Analyses of fat content, including skin and subcutaneous fat, showed a fat content at least 1.5 times higher than samples without skin.

### 12.1 Spatial variation

Today, the fat content in autumn-caught herring from the Baltic is similar in muscle tissue from almost all sites examined (Utlängan is the exception during the past three years) (table 12.1). At the beginning of the 1980s however, samples from Ängskärsklubb in the Bothnian Sea and Harufjärden in the Bothnian Bay were lower compared to samples from the Baltic Proper.

The fat content in herring from the Skagerrak varied and was sometimes twice as high compared to herring muscle from the Baltic and the Kattegatt. This is not surprising since Atlantic herring muscle tissue may contain more than 15% fat (table 12.1).

The fat content in cod liver was highly variable even between specimens caught at the same time at the same site. Geometric mean fat content over time in samples from south east of Gotland was more than 2.5 times higher compared to cod livers from the Kattegatt. This difference was significant (table 12.1).

### 12.2 Temporal variation

In the Baltic, significant decreasing trends in fat content were observed in herring muscle tissue from Harufjärden, Landsort and Utlängan (autumn and spring) (Fig. 12.1, 12.2). Fat decrease at Harufjärden, Landsort and Utlängan (spring) seemed to have ceased over the past decade. The fat content in herring from Utlängan (autumn) was exceptionally low during the last three years, less than 2%. An increasing trend was observed for fat in herring muscle from Fladen for the past 10 years (Fig. 12.2).
Overall, *increasing* trends in fat content were found in cod liver from the south east of Gotland and Fladen (Fig. 12.3). Fluctuating fat content in cod has to be considered when evaluating the time series of trace metals in cod liver (see above).

Significant *decreasing* trends of fat content in perch muscle were observed at both Holmöarna and Kvädöfjärden in the Baltic (Fig. 12.3). The time series of blue mussels from Väderöarna showed a significant decrease in fat content during the past 10 years (Fig. 12.4). Eelpout from Holmöarna in the Baltic proper and Väderöarna on the west coast are also showing a significant *decline* in fat content over the whole time period (Fig. 12.5). No linear trend is seen for fat content in guillemot egg (Fig. 12.6).
### 12.3 Seasonal variation

Fat content in spring-caught herring from Ängskärsklubb showed approximately the same mean value as herring from the same site caught in the autumn, whereas herring from Karlskrona archipelago caught in the autumn had about 30% higher mean fat content compared to spring-caught herring from the same area (except for the past three years) (table 12.1).

#### Table 12.1. Trend (in %) for concentration of fat (%) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s fat concentration values are estimated from the trend (%) if p<0.05, or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n years</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>Fat concentration last year (%) (95% CI)</th>
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<tr>
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<td></td>
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<td>3-5</td>
<td>502</td>
<td>30</td>
<td>78-09</td>
<td>-1.4 (-2.3, -.46)*</td>
<td>2.2 (1.9-2.6)</td>
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<td>Ängskärskl. aut.</td>
<td>3-5</td>
<td>467</td>
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<td>2.3 (1.9-2.8)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>- spring</td>
<td>2-5</td>
<td>652</td>
<td>36</td>
<td>2.9 (2.4-3.4)</td>
<td></td>
</tr>
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<td>Landsort</td>
<td>3-5</td>
<td>473</td>
<td>31</td>
<td>78-09</td>
<td>-1.5 (-2.8, -2.3)*</td>
<td>2.8 (2.2-3.5)</td>
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<td>2-4</td>
<td>481</td>
<td>30</td>
<td>80-09</td>
<td>-3.9 (-5.1, -2.7)*</td>
<td>1.6 (1.3-2.0)</td>
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<td>- spring</td>
<td>2-4</td>
<td>638</td>
<td>35</td>
<td>-2.2 (-3.1, -1.4)*</td>
<td>1.4 (1.1-1.6)</td>
</tr>
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<td>80-09</td>
<td>3.9 (3.2-4.9)</td>
<td>5.6 (3.7-8.5)</td>
</tr>
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<td>278</td>
<td>14</td>
<td>95-09</td>
<td></td>
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<td><strong>Cod liver</strong></td>
<td></td>
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<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>336</td>
<td>30</td>
<td>80-09</td>
<td>1.7 (.77, 2.5)*</td>
<td>67 (58-78)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>362</td>
<td>29</td>
<td>80-09</td>
<td>2.5 (.58, 4.5)*</td>
<td>27 (19-38)</td>
</tr>
<tr>
<td><strong>Perch muscle</strong></td>
<td></td>
<td></td>
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<tr>
<td>Holmöarna</td>
<td>292</td>
<td>23</td>
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<td></td>
<td>-0.81 (-1.1, -0.49)*</td>
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<tr>
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<td>28</td>
<td>80-09</td>
<td></td>
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<tr>
<td>Holmöarna</td>
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<td>95-07</td>
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<td>.55 (.44-.70)</td>
</tr>
<tr>
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<td>125</td>
<td>15</td>
<td>95-09</td>
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<td>.60 (.51-.72)</td>
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<td>18</td>
<td>88-09</td>
<td></td>
<td>-2.9 (-5.4, -1.4)*</td>
<td>.53 (.40-.69)</td>
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<tr>
<td>Fladen</td>
<td>3-6</td>
<td>158</td>
<td>13</td>
<td>81-94</td>
<td>-3.7 (-5.2, -2.2)*</td>
<td>.61 (.54-.68)</td>
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<tr>
<td><strong>Flounder muscle</strong></td>
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<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>4-6</td>
<td>190</td>
<td>15</td>
<td>80-94</td>
<td>-3.4 (-5.7, -1.0)*</td>
<td>.60 (.50-.73)</td>
</tr>
<tr>
<td><strong>Blue mussel</strong></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>104</td>
<td>27</td>
<td>81-09</td>
<td></td>
<td></td>
<td>.89 (.61-1.3)</td>
</tr>
<tr>
<td>Väderöarna</td>
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<td>29</td>
<td>80-09</td>
<td></td>
<td>1.5 (1.1-1.9)</td>
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<tr>
<td>Kvädöfjärden</td>
<td>80</td>
<td>15</td>
<td>95-09</td>
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<td>1.4 (1.1-1.7)</td>
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<td><strong>Guillemot egg</strong></td>
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<tr>
<td>St. Karlsö</td>
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<td>69-09</td>
<td></td>
<td>11 (11-13)</td>
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</tbody>
</table>

* significant trend, p < 0.05
Figure 12.1. Fat percentage in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1978 and 1980 respectively).

Figure 12.2. Fat percentage in herring muscle from Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1972, 1972, 1980 and 1995 respectively).
Fat %, cod liver and perch muscle

Fat %, blue mussel soft body

**Figure 12.3.** Fat percentage for cod liver from south east Gotland and Fladen (time series starting 1980), and perch muscle from Holmöarna and Kvädöfjärden (time series starting in 1980).

**Fat %, blue mussel soft body**

**Fladen**

- n(tot)=104, n(yrs)=27
- m=1.07 (0.879, 1.31)
- slope=1.4% (3.8, 1.0)
- SD(lr)=53%, 3.6%, 15 yr
- power=1.0, 0.3319%
- y(09)=.93 (61, 1.30)
- tao=.17, NS
- SD(sm)=0.01, 0.98%
- SD(lr)=3.0% (7.2, 1.3)
- SD(lr)=17%, 6.1%, 8 yr
- power=1.0, 0.3319%
- r²=2.25, NS

**Väderöarna**

- n(tot)=111, n(yrs)=29
- m=1.32 (1.14, 1.53)
- slope=2.4% (3.8, 2.5)
- SD(lr)=23%, 2.5%, 13 yr
- power=1.0, 0.514%
- y(09)=1.46 (1.11, 1.93)
- r²=0.2, NS
- tao=0.089, NS
- SD(sm)=0.01, 0.98%
- SD(lr)=3.0% (7.2, 1.3)
- SD(lr)=20%, 7.3%, 9 yr
- power=0.97, 0.97%
- r²=4.3, pc.038

**Kvadofjärden**

- n(tot)=80, n(yrs)=15
- m=1.28 (1.13, 1.44)
- slope=2.4% (3.8, 2.5)
- SD(lr)=23%, 2.5%, 13 yr
- power=1.0, 0.514%
- y(09)=1.37 (1.08, 1.74)
- r²=0.2, NS
- tao=0.089, NS
- SD(sm)=0.01, 0.98%
- SD(lr)=3.0% (7.2, 1.3)
- SD(lr)=20%, 7.3%, 9 yr
- power=0.97, 0.97%
- r²=4.3, pc.038

**Figure 12.4.** Fat percentage for blue mussel soft body from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1981, 1980, and 1995 respectively).
Figure 12.5. Fat percentage in eelpout muscle from Holmöarna, Kvädöfjärden and Väderöarna (time series starting in 1995, 1995 and 1988 respectively).

Figure 12.6. Fat percentage in guillemot eggs from Stora Karlsö (time series starting in 1969).
13 Mercury - Hg

13.1 Introduction

13.1.1 Usage, Production and Sources

Mercury exists naturally in the environment in a number of chemical and physical forms. The main inorganic forms include Hg^0 (metallic), Hg^{++} (mercurous), Hg^{2+} (mercuric). Organic forms include CH₃HgCH₃ (dimethylmercury) and CH₃Hg+ (monomethylmercury) (Suzuki et al. 1991).

Some of the more well-known uses of mercury include thermometers, barometers, sphygmomanometers (blood pressure cuffs), float valves (e.g. ball cock in flushing system of toilets), some electrical switches, amalgam for dental restoration, batteries, fluorescent lamps, anti-lock braking systems (ABS) in some 4WD vehicles and airbag sensors in some vehicle models. It can also be found in beauty products, such as mascara, as thiomersal. For a comprehensive list of mercury usage in everyday life, see Huber (1998). Highly toxic and bioaccumulatory methylmercury compounds were previously used as fungicides or were unwanted byproducts of the chemical industry (Clarkson 1992).

Natural sources of mercury include volcanoes, forest fires, fossil fuels, petroleum and cinnabar ore, which is mined primarily in Spain and Italy, although shortages of this rare metal have encouraged mining in other countries (Calvert 2007). There are numerous atmospheric anthropogenic sources of mercury such as fossil fuel combustion, mining, smelting and solid waste combustion; and soil and water anthropogenic sources such as agricultural application of fertilisers, industrial wastewater disposal, landfills, the manufacture of cement and metals, and through other industrial processes. In Sweden, a south to north gradient exists in atmospheric mercury concentration, due to the south being closer to source points in Europe (Wängberg & Munthe 2001). However, mercury use has almost ceased in Sweden (AMAP/UNEP 2008).

13.1.2 Environmental Fate

Mercury concentration in fish is highly correlated with water pH, with acidic conditions favouring mercury methylation; increased water temperature is known to increase methylation rates (Doetzel 2007). Sulfate reducing bacteria has been shown to be a controlling factor of mercury methylation in estuarine sediments (Choi & Bartha 1994). Fish biology also influences mercury levels, with age, size and diet affecting bioaccumulation rates (Doetzel 2007).
13.1.3 Toxic Effects

Mercury is a bioaccumulator (Clarkson 1992). Methylmercury is the form of mercury of most concern to human health and ecosystem processes. Methylmercury combines with the amino acid cysteine to form a structure similar to another amino acid, methionine, which penetrates all mammalian cells and easily crosses the blood-brain barrier, from whence the central nervous system can be affected (Suzuki et al. 1991, Huber 1998). High exposure can affect brain development, with young children and infants the most at risk (Doetzel 2007), as methylmercury disturbs cell division and therefore development (Huber 1998).

The severity of symptoms after mercury exposure depends upon exposure level. Symptoms related to severe exposure are well documented after two major disasters of methylmercury contamination in Iraq in 1972, and Japan in 1957 (for a brief overview see Huber 1998; Amin-Zaki et al. 1974; Rustam & Hadmi 1974; Clarkson 1992). Symptoms are related to type of exposure, for example, inhalation of elemental mercury vapours results in respiratory problems, followed by neurological disturbance and general systemic effects. However, one of the most common routes of mercury exposure is via ingestion of methylmercury (Ratcliffe et al. 1996), often through consumption of contaminated fish (Huber 1998), the risk of which can be greater for in utero children in pregnant women (Koren & Bend 2010). Exposure becomes problematic if contaminated fish (or other contaminated substances) are eaten often, and neurological effects in both adults and children in utero can be seen (Ratcliffe et al. 1996).

Wildlife in all environments are affected by mercury accumulation; however animals in aquatic systems appear to show more intense bioaccumulation/biomagnification effects than terrestrial species (Huber 1998). Bioaccumulation usually occurs through diet (Huber 1998). A biomagnification effect is seen in fish at higher trophic levels (i.e. piscivorous fish) compared to those at lower trophic levels (da Silva et al. 2005). In the 1960s, the use of methylmercury compounds as fungicides on seed grain that was eaten by small birds, which were in turn preyed upon by large bird species that then suffered from severe population declines, led to the realisation that methylmercury compounds were an ecological poison (Clarkson 1992). While methylmercury accumulates in fish muscle, highest concentrations are generally seen in the blood, spleen, kidney and liver; in mammals and birds, highest concentrations are typically seen in the feathers and fur (Huber 1998). Embryos and very young animals tend to be the most affected by mercury damage due to its ability to interfere with cell division processes (Huber 1998).

13.1.4 Conventions, aims and restrictions

The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that mercury discharges were to be reduced by 70% between 1985 and 1995, using 1985 as the base year.

The Minister Declaration from 1988, within HELCOM, calls for a reduction of the discharge of mercury to air and water by 50% by 1995, with 1987 as the base year.

The use of mercury in paper pulp industries has been banned in Sweden since 1966.

According to a governmental proposition (1993/94:163), the aim was that all mercury usage in Sweden should have ceased by 2000.
13.1.5 Target Levels

The target level (TL) used for Hg in the time series for fish is 20 ug/kg wet weight. For further information on TL and selection of target level see chapter 10.

In Swedish top layer soils (mor), the highest mercury concentrations are seen in the south, decreasing towards the north, with considerable local variation. Mercury concentrations vary regionally, with means from 0.5 mg/kg to 0.2 mg/kg seen. Natural background levels in mor/top layer soils are estimated to be 0.07 mg/kg, based on concentrations seen from the least affected northern areas. Natural background mercury concentrations in pike are estimated to be 0.2 mg/kg (European Communities 2002). Mercury concentrations in the ocean range from 0.7 – 1.1 pmol/L, with no difference between surface and deeper waters (Berlin et al. 2007). A decrease in mercury concentrations in surface waters of the Baltic Proper have been observed since 2000 (Pohl & Hennings 2006).

13.2 Methods

13.2.1 Analytical Information

Mercury is one of the mandatory contaminants that should be analysed and reported within both the OSPARCOM and HELCOM conventions.

The Department of Applied Environmental Science (ITM) at Stockholm University has determined the concentration of mercury in fish muscle and blue mussel soft body since 2007. Prior to 2007, the concentration of mercury in fish muscle and blue mussel soft body was determined using a ‘Mercury Monitor LCD 3200’ detector at the Department of Environmental Assessment at the Swedish University of Agricultural Sciences (SLU) up until 2006. The quantification limit is estimated to approximately 10 ng/g dry weight.

In 1992, new analytical equipment was introduced and great efforts have been made to intercalibrate the new method by reanalysing old samples, both dried extracts and samples from the Environmental Specimen Bank.

Please note that since 2007, the analytical laboratory for metals changed from SLU to ITM at SU. See chapter 6 section 6.1 for further details.
13.3 Results

13.3.1 Spatial Variation

Herring muscle from Lagnö shows the highest mercury concentrations of all herring samples 2007-2009 (Fig. 13.1).

In the time series, the herring from Ängskärsklubb show the highest mercury concentration (except for the last few years) (Fig 13.2). This might be due to local discharge levels. Samples collected during the 1980s from Ängskärsklubb are therefore most probably not representative of mercury concentration in the Bothnian Sea. At the beginning of the 1980s, mercury concentrations in herring from Ängskärsklubb ranged from 60 - 180 ng/g.

Among the other herring sites, Harufjärden showed the highest mercury concentration over time, being significantly higher than Landsort, Utlängan and Fladen. The time series from Utlängan in the southern Baltic Proper showed the lowest mercury concentrations in the Baltic with a geometric mean concentration of about 20 ng/g (Fig 13.2).

Cod muscle tissue from Fladen in the Kattegatt (52 ng/g) showed significantly higher concentrations than samples from south east of Gotland (40 ng/g) (Fig 13.5). Perch muscle samples from Holmåarna in the Quark showed significantly higher concentrations (58 ng/g)
compared to perch samples from Kvädöfjärden (28 ng/g) on the coast of the Baltic Proper (Fig 13.4). The estimated geometric mean concentration for Holmöarna 2009 was about two times higher than for Kvädöfjärden in 2009.

Mercury concentration in flounder from the Skagerrak showed values in the same range as Danish flounder samples from the Belt Sea, but significantly lower compared to Danish flounder samples from the Sound (ICES, 1995).

Mercury in blue mussels from Fladen in the Kattegatt and Väderöarna in Skagerrak showed no spatial variation (Fig. 13.7). The overall mean concentration in blue mussel samples from the two sites exceeded the upper limit range of ‘present background concentrations in pristine areas within the OSPAR Convention Area’, proposed to be between 5 - 10 ng/g wet weight (ICES, 1997).

The estimated mean concentrations for 2009 in herring and cod muscle (except for cod from Fladen (52 ng/g), perch and eelpout from Holmöarna (58 and 64 ng/g respectively) and eelpout from Kvädöfjärden (64 ng/g), all fall inside the proposed range of ‘present background concentrations in pristine areas within the OSPAR Convention Area’ (10 - 50 ng/g fresh weight in round fish, ICES, 1997).

13.3.2 Temporal variation

There is no common general trend for mercury in herring muscle for the investigated time series (Fig. 13.2, 13.3). Mercury was monitored in spring-caught herring from Ångskärsklubb and Karlskrona for four years at the beginning of the 1970s (Fig. 13.2). These series were continued in 1996. Both series show a significant decrease. The time series from Landsort show a significant decrease during the last ten years. From the Swedish west coast, a significant increase of about 1% is seen for herring from Fladen.
Figure 13.2. Mercury concentrations (ng/g fresh weight) in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting 1980). The green line denotes the suggested target value for mercury in fish.

The time series from Ängskärsklubb in the Bothnian Sea shows a very large between-year variation. Although the sampling site at Ängskärsklubb is located relatively far off the coast, mercury concentration in herring samples could be influenced by local discharges. Ängskärsklubb may thus not be representative of the Bothnian Sea.

During 1995 – 1996, the estimated mean concentration in herring muscle from Ängskärsklubb was on par with that measured in comparable samples from Landsort. However, in 1997 and 1999, the geometric mean concentrations increased to the same level as that recorded at the beginning of the 1980s.
Figure 13.3. Mercury concentrations (ng/g fresh weight) in herring muscle from Ängskärsklubb (spring), Karlskonna (spring), Fladen and Väderöarna (time series starting in 1972, 1970, 1980 and 1995 respectively). The green area denotes the levels below the suggested target value for mercury in fish.

The number of years required to detect an annual change of 10% varied between 9-13 (16 for Ängskärsklubb) years for the herring time series. The power to detect a 10% annual change was close to 100% for most of the time series.
Perch muscle samples from Kvädöfjärden in the Baltic Proper show a significant decreasing trend (except for the last ten years where a significant increasing trend is observed), whereas no significant trend is seen at Holmöarna (Fig. 13.4).

Figure 13.4. Mercury concentrations (ng/g fresh weight) in perch muscle from Holmöarna and Kvädöfjärden (time series starting in 1991 and 1981 respectively). The green area denotes the levels below the suggested target value for mercury in fish.
Cod from both Fladen and south east of Gotland show a significant increasing linear trend of 1 and 3% respectively (Fig. 13.5).

Figure 13.5. Mercury concentrations (ng/g fresh weight) in cod muscle from south east Gotland and Fladen (time series starting in 1979). The green area denotes the levels below the suggested target value for mercury in fish.
Mercury concentration in eelpout shows a significant increasing trend at Väderöarna. At Kvädöfjärden and Holmöarna on the other hand decreasing trends are indicated for the whole time period and significant decreasing trends are observed during the last ten years for both sites (Fig. 13.6).

Figure 13.6. Mercury concentrations (ng/g fresh weight) in eelpout muscle from Holmöarna, Kvädöfjärden and Väderöarna (time series starting in 1995). The green area denotes the levels below the suggested target value for mercury in fish.
Mercury concentration in blue mussels show no linear trend for any of the sites (Fig. 13.7).

**Figure 13.7.** Mercury concentrations (ng/g fresh weight) in blue mussel soft body tissue from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1981, 1980 and 1995 respectively).
Guillemot eggs from Stora Karlsö in the Baltic proper show a significant decrease in mercury concentration of about 1.7% a year (Fig 13.8). It should be noted that the mercury analysis in this time series has been carried out in a retrospective study i.e. all analyses were performed at one occasion at the same laboratory.

**Figure 13.8.** Mercury concentrations (ng/g fresh weight) in guillemot eggs (early laid) from St. Karlsö (time series starting in 1969).

### 13.3.3 Species Differences

Significant differences in mean mercury concentration (ng/g wet weight) were found in fish muscle and blue mussel soft body between species on the Swedish west coast (table 13.1).

Holmöarna: Eelpout (64) > Perch (58)
Kvädöfjärden: Eelpout (63) > Perch (28) – Blue mussel (18)
Fladen: Cod (52) > Herring (31) > Blue mussel (14)
Väderöarna: Eelpout (40) – Herring (29) - > Blue mussel (15)

The mercury concentration in blue mussel was, for all sites, lower than in fish muscle. The levels found in guillemot eggs were 3 - 20 times higher compared to levels in fish muscle.

No significant differences in mercury concentrations were found between spring- and autumn-caught herring from Ångskärsklubb and Karlskrona.
13.4 Conclusion

Within the current research data, there is considerable spatial variation in mercury concentration between sites within species. Finnish mercury analyses of herring muscle samples between 1980 - 83 from the eastern part of the Bothnian Sea showed concentrations around 20 ng/g (ICES, 1995), i.e. the same level as results from Ängskärsklubb in 1994 - 1996.

Finnish data of mercury levels in cod from the Bothnian Sea and the mouth of the Gulf of Finland showed concentrations in the same range as the Swedish data from Gotland (ICES, 1995). However, the mercury concentration in cod muscle from Fladen was within the same range as in cod muscle from the same age class from reference stations along the Norwegian coast (Green & Rönningen, 1994) analysed at the Norwegian Institute for Water Research (NIVA).

The results concerning changes in mercury concentration in the investigated matrices are inconsistent. Mercury concentration in guillemot eggs decreased, whereas the concentration in herring from the northern Baltic Proper fluctuated. In most cases, the observed trends do not meet the North Sea Conference or HELCOM aims for mercury reduction. Future changes in mercury concentration have to be studied carefully, and possible analytical problems thoroughly investigated.

Generally, mercury concentration is above the suggested target level for concentrations in fish for the protection of predators against secondary poisoning of 20 ng/g wet weight, but below 100 ng/g wet weight.

The concentration in fish muscle from the various sites all fall below the Swedish National Food Administration (SNFA) suggested limits for human consumption (500 ng/g fresh weight) by a factor of 6 - 25. However, the suggested limit for children’s food is 50 ng/g, which is close to the overall mean concentration in fish muscle from most of the investigated sites (SLVFS, 1993).

Perttilä et al. (1982) examined heavy metal concentrations in herring muscle (from specimens aged 1 – 4 years), caught from the Gulf of Finland in 1981. Mercury concentrations were highest in older specimens (0.044 mg/kg). Mercury levels in perch muscle from specimens caught in the Pomeranian Bay and Szczecin Lagoon were examined seasonally between 1996 – 1997. Concentrations ranged from 0.028 – 0.120 µg/g wet weight (Szefer et al. 2003). Mercury content in guillemot feathers from the Baltic, the Kattegatt, the Faroe Islands and Greenland was measured, and found to be higher in the Baltic and Kattegatt (Appelquist et al. 1985). Mercury concentration was examined in the eggs of a number of Norwegian seabirds, including guillemot, in 1983. In guillemot eggs, levels ranged from 0.08 – 0.13µg/g (Barrett et al. 1985).
Table 13.1. Trend (in %) for mercury (ng/g fresh weight) assessed from the annual geometric mean in various matrices. The age interval for fish and length interval for blue mussels, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s mercury concentration values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>Mercury concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Herring muscle</strong></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>467</td>
<td>29</td>
<td>80-09</td>
<td>38 (31-46)</td>
<td></td>
</tr>
<tr>
<td>Ångskärskl. aut.</td>
<td>3-5</td>
<td>449</td>
<td>29</td>
<td>80-09</td>
<td>39 (26-60)</td>
<td></td>
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<td>” spring</td>
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<td>72-75,96-09</td>
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<td>23 (19-27)</td>
<td></td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>439</td>
<td>30</td>
<td>80-09</td>
<td>27 (21-37)</td>
<td></td>
</tr>
<tr>
<td>Utlängan, aut.</td>
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<td>454</td>
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<td>80-09</td>
<td>20 (15-27)</td>
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<tr>
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<td>232</td>
<td>18</td>
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<td>-.92 (-1.7,-.16)*</td>
<td>18 (16-20)</td>
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<td>2-3</td>
<td>525</td>
<td>30</td>
<td>80-09</td>
<td>1.2 (.13, 2.4)*</td>
<td>31 (25-37)</td>
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<td>256</td>
<td>14</td>
<td>95-09</td>
<td></td>
<td>29 (22-38)</td>
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<tr>
<td><strong>Cod muscle</strong></td>
<td></td>
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<td>SE Gotland</td>
<td>3-4</td>
<td>424</td>
<td>31</td>
<td>79-09</td>
<td>3.3 (2.4, 4.2)*</td>
<td>44 (38-52)</td>
</tr>
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<td>2-4</td>
<td>502</td>
<td>31</td>
<td>79-09</td>
<td>.93 (.02, 1.8)*</td>
<td>53 (45-63)</td>
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<td><strong>Perch muscle</strong></td>
<td></td>
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<td>174</td>
<td>15</td>
<td>91,95-09</td>
<td></td>
<td>58 (45-77)</td>
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<td>Kvädöfjärden</td>
<td>269</td>
<td>27</td>
<td>81-09</td>
<td>-2.1 (-3.8, -.40)*</td>
<td>28 (21-36)</td>
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<td>Eelpout muscle</td>
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<td></td>
</tr>
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<td>Holmörarna</td>
<td>109</td>
<td>12</td>
<td>95-09</td>
<td>-3.7 (-8.1, .62)</td>
<td>64 (48-85)</td>
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<td>152</td>
<td>95-09</td>
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<td>95-09</td>
<td>6.2 (1.4, 11)*</td>
<td>40 (27-60)</td>
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<td>278</td>
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<td>Väderöarna</td>
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<td>248</td>
<td>14</td>
<td>81-94</td>
<td>46 (25-83)</td>
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<td>shell 1  Fladen</td>
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<td>Väderöarna</td>
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<td>29</td>
<td>80-09</td>
<td></td>
<td>15 (11-19)</td>
<td></td>
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<tr>
<td>Kvädöfjärden</td>
<td>127</td>
<td>13</td>
<td>95-09</td>
<td></td>
<td>18 (11-30)</td>
<td></td>
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<tr>
<td><strong>Guillemot egg</strong></td>
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<td></td>
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<td>St. Karlsö</td>
<td>285</td>
<td>37</td>
<td>69-09</td>
<td>-1.7 (-2.3, -1.1)*</td>
<td>190 (170-220)</td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
14 Lead - Pb

14.1 Introduction

14.1.1 Usage, Production and Sources

Lead is produced in many isotopes, but only three are stable. There are four natural isotopes, $^{204}$Pb, $^{206}$Pb, $^{207}$Pb and $^{208}$Pb. $^{204}$Pb is slightly radioactive, and has a half life of 22.2 years. In nature, lead is usually found in ore with zinc, silver or copper. Atmospheric sources of lead in Sweden show a south to north gradient, due to northward atmospheric transport from sources located in other parts of Europe (Renberg et al. 2000). The main sources of lead pollution in Sweden come from ammunition, lead petrol emissions and associated contamination in roadside soils (although leaded gasoline was eliminated in 1994 in Sweden (Faiz et al. 1996)), lead pigments, cables and batteries. There are also point sources (e.g. metal works) that have resulted in high local pollution (Bergbäck et al. 1992), e.g. a secondary lead smelter in Landskrona where lead from car batteries is recycled (Farago et al. 1999).

There are numerous other uses for lead, including, but not limited to, lead in car batteries, in the ballast keel of sailboats, scuba diving weight belts, fishing sinkers, firearms (bullets and shot), colouring elements in paints and ceramic glazes, PVC plastics, lead sheeting used for sound proofing, lining chemical treatment baths, storage vessels, weathering, roofing, cladding, organ pipes, soldering, electrodes, high voltage power cables, tennis racquets, statues, sculptures, anti-knocking additive in aviation fuel, leaded gasoline, solar energy cells and infrared detectors and coffins. Houses built prior to 1980 are at a higher risk of having been painted with lead-based paints. Many cities did (and some still do) use lead water and sewage pipes. Lead can leach out of the water pipes into drinking water. Lead arsenate was the most commonly used insecticide in deciduous fruit tree orchards prior to the introduction of DDT in 1947. High lead levels are still found in some top soils in the USA (Peryea & Creger 1993; Peryea & Kammereck 1995).

14.1.2 Environmental Fate

Increased acidity levels appear to contribute to increased lead bioavailability in soils (Jin et al. 2005). In lakes, the level of lead in fish body tissues is often greater in low-alkalinity waters compared to lakes with a higher pH (Spry & Wiener 1991). These results indicate that pH may influence lead bioavailability in water systems and sediments.

14.1.3 Toxic Effects

Lead is a non-essential element (Tewari et al. 1987) and a known neurotoxin, damaging the nervous system and causing brain and blood disorders. The toxic effects of lead involves several organ systems and biochemical activities. The risk is highest for children and those in utero, partly because of high permeability of the blood-brain barrier and placenta (Klaassen & Rozman, 1991). Some neurophysiological development effects can be seen in children even at low levels of lead exposure (Gidlow 2004).

Lead is known to bioaccumulate in soft tissue, but to a much greater extent in the bone matrix. Approximately 90% of the total amount of lead in humans is found in the skeleton.
Between 90% to 95% of lead that is found in blood is isolated in the red blood cells where haemoglobin synthesis can be inhibited, and subsequently symptoms such as anaemia are seen (Gidlow 2004). In females, lead is a known abortifacient, but problems in male reproduction are equivocal (Gidlow 2004).

In animals, absorbed lead enters the blood and soft tissues, but is eventually redistributed to the bones. In birds, lead shot is a common cause for lead poisoning (Cook & Trainer 1966; Pattee et al. 1981), and there have been reports of fishing sinkers causing bird deaths (Locke et al. 1981). In Sweden, bird death from lead poisoning is more common in swans, geese and ducks, but has also been reported in woodpeckers (Mörner & Petersson 1999). Lead levels were found to be highest in woodpecker liver and kidney (Mörner & Petersson 1999).

14.1.4 Conventions, Aims and Restrictions
The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that the lead discharges were to be reduced by 70% between 1985 and 1995, using 1985 as the base year.

The Minister Declaration from 1988, within HELCOM, calls for a reduction of the discharges of lead to air and water by 50% by 1995, with 1987 as the base year.

14.1.5 Target Levels
The target level (TL) used for Pb in the time series for herring and perch is 300 ug/kg wet weight. For further information on TL and selection of target level see chapter 10. The original TL has been recalculated to dry weight in liver for each time series to fit the presented data. The recalulation of the TL for liver is based on a study that compared concentration of Pb in muscle and liver, herring and perch, with the ratio 8 and 5.3 respectively (Strandmark et al, 2008) and the recalculation to dry weight is based on the dry weight in each time series. The recalculated target level (Tv) together with the dry percentage (dp) is shown above the statistical information in each time series.

The recommended limit for children’s food is set by the Swedish National Food Administration (SNFA) at 50 ng/g fresh weight (SLVFS, 1993)

14.2 Methods

14.2.1 Analytical Information
Lead is one of the mandatory contaminants that should be analysed and reported within both the OSPARCOM and HELCOM conventions.

The concentration of heavy metals, except mercury, in fish liver and blue mussel soft body was determined using an atomic absorption spectrophotometer with a graphite furnace at the Department of Environmental Assessment at the Swedish University of Agricultural Sciences (SLU) up until 2003. The quantification limit is estimated to approximately 100 ng/g dry weight for zinc, approximately 10 ng/g dry weight, which implies that the concentrations in herring, flounder and dab are approximately 10 - 20 times above the quantification limit.

Please note that since 2007, the analytical laboratory for metals changed from SLU to the Department of Applied Environmental Science (ITM) at Stockholm University. See chapter 6 section 6.1 for further details.
14.3 Results

14.3.1 Spatial variation

Figure 14.1. Spatial variation in lead concentration (ug/g dry weight) in herring liver.

Ångskärsklubb has the highest concentration of lead in herring liver 2007-2009. High levels are also observed in the Sout Baltic Proper and at Kullen on the Swedish west coast (Fig 14.1).

The lead concentration in blue mussels from the Swedish west coast were not significantly higher compared to blue mussel samples of similar length from a reference site at Kobbefjord, Greenland (Riget et al. 1993). Mussel samples from all three sites (Kvädöfjärden, Fladen, Väderöarna) showed mean levels below the ‘background concentration at diffuse loading’ in blue mussels for lead of <5 μg/g dry weight, proposed by Knutzen and Skie (1992).

14.3.2 Temporal variation

At Harufjärden, Ångskärsklubb (autumn), Landsort, Utlängan (autumn) and Fladen, the investigated time series in herring liver show significant decreasing trends (Fig. 14.2, 14.3)
Figure 14.2 (above) and 14.3 (below). Lead concentrations (ug/g dry weight) in herring liver from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1981)(above); and Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1996, 1997, 1981 and 1995 respectively). The green area denotes the levels below the suggested target value for lead in fish.

Pb, ug/g dry w., herring liver

Figure 14.2 (above) and 14.3 (below). Lead concentrations (ug/g dry weight) in herring liver from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1981)(above); and Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1996, 1997, 1981 and 1995 respectively). The green area denotes the levels below the suggested target value for lead in fish.

Pb, ug/g dry w., herring liver
The number of years required to detect an annual change of 10% varied between 10 - 18 years for the herring time series, with a power to detect a 10% annual change ranging from 0.78 (shorter series) to 1.0 (longer series). An annual change greater than 10% would likely be detected.

Lead concentrations in cod liver (after adjusting for varying fat content) show decreasing trends from south east of Gotland and Fladen (Fig. 14.4).

Lead concentrations in the shorter time series of perch liver show decreasing trends from both Kvädöfjärden and Holmöarna (Fig. 14. 5).
Figure 14.4. Lead concentrations (ug/g dry weight) in cod liver from south east Gotland and Fladen (time series starting in 1981).

Figure 14.5. Lead concentrations (ug/g dry weight) in perch liver from Holmöarna and Kvädöfjären (time series starting in 1995). The green area denotes the levels below the suggested target value for lead in fish.
The lead concentration in blue mussel soft body from Fladen show a significant decreasing trend (Fig. 14.6).

**Pb, ug/g wet w., blue mussel softbody**

**Fladen**
- n(tot)=508, n(yrs)=26
- m=.185 (.138, .247)
- SD(yr)=65%,4.6%,17 yr
- tao= -.40, p=.004
- SD(sm)=30, bc=.087,19%
- SD(yr)=48%,21%,14 yr
- power=.28,38%,17%
- y(09)=.091 (.057,.145)
- r2=.35, p<.001 *
- tao=.40, p=.004
- SD(sm)=30, bc=.087,19%
- SD(yr)=48%,21%,14 yr
- power=.28,38%,17%
- y(09)=.091 (.057,.145)
- r2=.35, p<.001 *

**Väderöarna**
- n(tot)=515, n(yrs)=27
- m=.237 (.198,.284)
- SD(yr)=65%,3.4%,14 yr
- tao= -.17, NS
- SD(sm)=31, NS,17%
- SD(yr)=60%,28%,16 yr
- power=.27,22%, NS
- y(09)=.219 (.152,.314)
- r2=.01, NS
- tao=.17, NS
- SD(sm)=31, NS,17%
- SD(yr)=60%,28%,16 yr
- power=.27,22%, NS
- y(09)=.219 (.152,.314)
- r2=.01, NS

**Kvädöfjärden**
- n(tot)=138, n(yrs)=14
- m=.250 (.178,.352)
- SD(yr)=8.0%(-21,5.3)
- tao=.033, NS
- SD(sm)=38, NS,20%
- SD(yr)=52%,23%,15 yr
- power=.25,34%,19%
- y(09)=.315 (.158,.630)
- r2=.22, NS
- tao=.033, NS
- SD(sm)=38, NS,20%
- SD(yr)=52%,23%,15 yr
- power=.25,34%,19%
- y(09)=.315 (.158,.630)
- r2=.22, NS

**Figure 14.6.** Lead concentrations (ug/g dry weight) in blue mussel soft body tissue from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1981, 1981 and 1995 respectively).

The time series of lead in guillemot eggs shows a significant decreasing trend of 15% per year (Fig. 14.7).
Pb, ug/g dry w., guillemot egg, early laid

Figure 14.7. Lead concentrations (ug/g dry weight) in guillemot eggs from Stora Karlsö (time series starting in 1996).

14.3.3 Species differences

Significant differences in mean lead concentration (µg/g dry weight) were found in fish liver and blue mussel soft body between species marked with a ‘>’:

Holmöarna: Eelpout (0.03) > Perch (0.009)
Kvädöfjärden: Blue mussel (4.3) > Eelpout (0.05) > Perch (0.01)
Fladen: Blue mussel (0.86) > Herring (0.04) > Cod (0.01)
Väderöarna: Blue mussel (1.1) > Eelpout (0.08) > Herring (0.04)

The lead concentration in blue mussel soft body tissue was generally much higher than concentrations in fish liver. The concentration in eelpout liver was about two to four times higher than perch liver in the analysed samples.
14.4 Conclusion

On a spatial scale, lead levels in herring muscle from individuals aged from 1 – 6 years old sampled in the Gulf of Finland in 1981, were found to vary little, from 0.04 – 0.06 mg/kg (Perttilä et al. 1982). No difference was seen in lead concentrations of blue mussels examined between three sites; these lead concentrations were not significantly different to results seen from similar sized blue mussels sampled from a reference site in Kobbefjord, Greenland (Riget et al. 1993).

Over time, lead concentrations have decreased in most species at most sites. This probably reflects a general decrease of lead in the environment; however more still needs to be done to meet and exceed target levels set at the North Sea Convention and HELCOM. Jorhem and Sundström (1993) found lead levels to be about 75% lower in fish samples (Baltic herring, cod and pike) from 1983 – 1990, compared with a previous study from 1973 - 1982 (Jorhem et al. 1984).

The lead concentrations are all below the suggested target level based on maximum levels in foodstuffs, 300 ng/g wet weight. The recommended limit for children’s food, as set by the Swedish National Food Administration, is 50ng/g fresh weight. Current concentrations of lead in herring liver from the examined sites are substantially lower than this.
Table 14.1. Trend (in %) for lead (ug/g dry weight, wet weight for blue mussels) assessed from the annual geometric mean in various matrices. The age interval for fish and length interval for blue mussels, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s lead concentration values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
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<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>Lead concentration last year (95% CI)</th>
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<td>427</td>
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<td>81-09</td>
<td>-5.3 (-8.5, -2.1)*</td>
<td>0.034 (.020-.057)</td>
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<td>-4.1 (-5.5, -2.8)*</td>
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<td>0.18 (.10-.30)</td>
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<td>0.51 (.43-.62)</td>
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<td>392</td>
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<td>95-09</td>
<td>.083 (.054-.13)</td>
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<td>.77 (-3.0, 4.6)*</td>
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<td>14</td>
<td>81-94</td>
<td>-0.06 (-5.4,5.3)*</td>
<td>173 (115-260)</td>
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<td>508</td>
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<td>-5.2 (-8.1, -2.2)*</td>
<td>.091 (.057-.15)</td>
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<td>St. Karlsö</td>
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<td>96-09</td>
<td>-15 (-24,-6.7)*</td>
<td>0.012 (.006-.024)</td>
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</tr>
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</table>

* significant trend, p < 0.05
15 Cadmium – Cd

The time series of cadmium concentrations in fish liver and blue mussel soft body started in 1981.

15.1 Introduction

15.1.1 Usage, Production and Sources
Cadmium is a chemical element widely used in many industrial processes and products. Within the EU, the main use of cadmium is for the production of rechargeable nickel-cadmium batteries and for metal plating and alloys. It is also used as a colour pigment in paints and a stabiliser in plastics. Cadmium is an impurity in phosphate rock used to manufacture fertilisers.

Natural processes, such as volcanic emissions and weathering of cadmium-bearing rocks release cadmium to both air and water. Anthropogenic sources include metal production, burning of fossil fuels, incorrect waste disposal (mainly Ni-Cd batteries) and transportation. Phosphate fertilisers used to be the main source of cadmium to agricultural land in Sweden, however, the applied amount has successively decreased since 1993 due to regulatory restrictions of the cadmium content in fertilisers. The main sources of cadmium to the Baltic Sea are point sources and riverine runoff (HELCOM 2010). Atmospheric deposition accounts for ca. 15%. According to HELCOM (2010), the waterborne input of cadmium to the Baltic Sea has decreased 91% and the atmospheric deposition 46% between 1990-2007. Despite this significant overall reduction, no decrease in the cadmium load from Swedish rivers to the Baltic Sea has been observed in the last 15 years (Naturvårdsverket 2007).

15.1.2 Environmental Fate
The environmental fate of cadmium depends largely on the surrounding conditions, e.g. pH, redox condition, salinity and presence of organic matter, which influence its chemical form. In water, cadmium exists as dissolved ions and soluble or insoluble complexes. Soluble cadmium is relatively mobile in water and in soil. Under oxic conditions, cadmium primarily adsorb to organic matter and form oxide/hydroxide complexes, while the formation of less soluble Cd-sulfides is dominating under reducing conditions. Cadmium tend to partition to sediments and the levels in sediment are often at least an order of magnitude higher than in the overlying water column. In soils it may be very mobile, particularly under acidic conditions, and the amount of cadmium transported from land to sea via rivers is often strongly correlated with the annual run-off.

Increasing salinity generally increase the soluble fraction of Cd. This is due to a combination of an increased formation of soluble chloride-complexes and by competition with Ca\(^{2+}\) for adsorption sites on suspended particles. However, the bioavailable fraction (i.e. the free Cd\(^{2+}\) ion) decrease with increasing salinity, since the Cd-chloride complexes are not available for uptake.

15.1.3 Toxic Effects
Cadmium is highly toxic to aquatic organisms. It can bioaccumulates and be transferred through the food chain. Cadmium does not undergo any direct metabolisation but can bind
to specific metal-binding proteins, e.g. metallothionein, preventing it from exerting its toxicity. Relatively large amounts of cadmium can be retained in the body bound to metallothionein. Chronic exposure results in the accumulation of cadmium in the kidney and liver. Kidney damage is the main toxic effect of chronic exposure to cadmium.

The most common source of cadmium for humans is via cigarette smoke (Godt et al. 2006). There is also a low risk of being exposed to cadmium via oral and dermal pathways (Godt et al. 2006). Cadmium is generally found in the liver or kidneys, 30% of the cadmium body burden is found in the kidneys. The kidneys are the main organ for long term cadmium accumulation in humans, leading to renal tube dysfunction (Godt et al. 2006). Bone tissues are secondarily affected. At very high exposure rates, effects on the respiratory system (e.g. emphysema) are known, while the nervous system in developing animals appears to be sensitive (Godt et al. 2006). There have been some effects on reproduction, and some proof of carcinogenic effects. Cadmium transported in blood plasma becomes bound to albumin and is then preferentially taken up by the liver, where metallothionein is synthesised. The placenta is only a partial barrier to foetal exposure. Cadmium is excreted in faeces and urine (Godt et al. 2006).

15.1.4 Conventions, Aims and Restrictions
The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that cadmium discharges were to be reduced by 70% between 1985 and 1995, using 1985 as the base year.

The Minister Declaration from 1988, within HELCOM, calls for a reduction in discharges of cadmium to air and water by 50% by 1995, with 1987 as the base year.

The Swedish Parliament has agreed on a general reduction of cadmium discharge, aiming at a reduction of 70% between 1985 and 1995, and further, that all use of cadmium that implies a risk of discharge to the environment, in a longer term perspective, will cease (prop 1990/91:90, JoU 30, rskr.343).

In 1982, the use of cadmium in electroplating and as a thermal stabiliser was banned in Sweden.

In 1987, a fee on batteries containing cadmium was introduced in Sweden. This fee was raised considerably in 1991.

In 1993, the content of cadmium in fertilisers was restricted to 100g/ton of phosphorus in Sweden.

15.1.5 Target Levels
The target level (TL) used for Cd in the time series for herring and perch is 160 ug/kg wet weight muscle. For further information on TL and selection of target level see chapter 10. Since most data presented here are ug Cd per dry weight liver, the original TL has been recalculated for comparison. The recalculation of the TL for liver is based on a study that compared concentrations of Cd in the muscle and liver of herring and perch (Strandmark et al., 2008). The liver:muscle ratio was 570 and 566 for herring and perch respectively. (Strandmark et al, 2008) and the recalculation to dry weight is based on the dry weight in each time series. The recalculated target level (Tv) together with the dry weight percentage (dp) is shown above the statistical information in each time series.

15.2 Methods

15.2.1 Analytical Information
Cadmium is one of the mandatory contaminants that should be analysed and reported within both the OSPARCOM and the HELCOM conventions.
15.3 Results

15.3.1 Spatial variation

Figure 15.1. Spatial variation in cadmium concentration (ug/g dry weight) in herring liver.

The Bothnian Sea showed higher levels of cadmium in herring liver compared to the Bothnian Bay, the Baltic Proper and the Swedish west coast. The highest level was observed at Gaviksfläden (Fig 15.1).

Overall, mean cadmium concentrations in herring liver from the Baltic showed significantly higher concentrations when compared to Fladen in the Kattegatt and Väderöarna in the Skagerrack on the Swedish west coast (table 15.1). The geometric mean concentration in herring liver for 1981 - 2009 from Landsort and Utlängan (the Baltic Proper) show 4 and 4.4 times higher values respectively, compared to samples from the Kattegatt and Skagerrak (table 15.1).

Eelpout livers from Holmöarna in the southern Bothnian Bay and Kvädöfjärden in the Baltic Proper, showed about four to six times higher geometric mean cadmium concentrations (1995-2009) compared to samples from Väderöarna in the Skagerrak (table 15.1).
Blue mussels from Kvädöfjärden, analysed between 1995 - 2009, showed about three times higher concentrations compared to blue mussel samples from the Swedish west coast (table 15.1). The samples from the Swedish west coast showed mean levels similar to that found in blue mussels from the Belgian coast (Vyneke et al. 1999) and did not exceed the ‘high background concentration at diffuse loading’ for cadmium in blue mussels (<2 µg/g dry weight) proposed by Knutzen and Skie (1992), whereas the samples from Kvädöfjärden did. All blue mussel samples exceeded the range of ‘present background concentrations in pristine areas within the OSPAR Convention Area’ proposed at 0.070-0.11 µg/g wet weight (ICES 1997). The estimated geometric mean concentration from Kvädöfjärden exceeded this concentration by about four times.

Cadmium concentrations in cod livers from Fladen in the Kattegatt were significantly higher (about five times higher on a dry weight basis, and two times higher on a fresh weight basis) compared to samples from south east of Gotland. This may be explained by the average fat content in cod liver from Gotland being about 2.5 times higher compared to samples from the Kattegatt. The Swedish data from south east of Gotland were in the same range as Finnish data of cod liver from the Gulf of Finland and the Bothnian Sea.

15.3.2 Temporal variation

Between 1981-2000, cadmium concentrations in herring liver from Utlängan (autumn) in the Baltic Proper show a significant increasing log-linear trend (Fig 15.2). However, during the last decade this trend has shifted to a decreasing trend (Fig 15.2). At the same location, for herring collected in the spring a decreasing trend is observed for the whole time period (1995-2009) (Karlskrona, Fig 15.3). Total cadmium concentrations increased about 2 to 5 times during 1981 - 1995 at Angskärsklubb, Landsort and Utlängan. In recent years, these increases have levelled out (Fig. 15.2, 15.3).

The herring liver cadmium concentrations at all sites are more than 50 times lower than the recalculated target level (Tv).

The number of years required to detect an annual change of 10% varied between 8 - 14 years for the herring time series, with a power to detect a 10% annual change ranging from 0.92 to 1.0.
Figure 15.2. Cadmium concentrations (ug/g dry weight) in herring liver from Harufjärden, Ängskärsklubb, Landsort and Utllängan (time series starting in 1981). The green area denotes the levels below the suggested target value for cadmium in fish.

Figure 15.3. Cadmium concentrations (ug/g dry weight) in herring liver from Ängskärsklubb (spring), Karlskrona, Fladen and Väderöarna (time series starting in 1995, 1995, 1980 and 1995, respectively). The green area denotes the levels below the suggested target value for cadmium in fish.
Cadmium concentrations in cod liver samples (adjusted for varying fat content) from south east of Gotland showed a significant decreasing trend. A decreasing trend is also observed for the cod from Fladen (Fig. 15.4).

Cadmium concentrations in perch liver samples showed no trend at any of the sites (Fig. 15.5). The perch liver cadmium concentrations at both sites are more than 800 times lower than the recalculated target level (Tv).
Cd, ug/g dry w., perch liver

Holmoarna
- n=83, mean=140.9, median=14
- slope=5.9% (2.0, 7.9)
- y=97.59, 95%-confidence interval (CI)=2%
- y=40.5 (26.7, 61.5)
- n=44, median=81, 75%-CI=30%
- slope=1.4% (0.97, 21%
- power=.97/.59/13%
- y=0.405 (0.267, 0.615)
- n=44, median=81, 75%-CI=30%
- slope=1.4% (0.97, 21%
- power=.97/.59/13%
- y=0.405 (0.267, 0.615)

Kvadofjarden
- n=51, mean=135.8, median=14
- slope=-4.1% (4.3, 7.2)
- y=95.79, 95%-CI=1%
- y=36.2 (27.2, 46.6)
- n=64, median=36, 75%-CI=18%
- slope=0.6% (5.6, 5.6)
- power=1.0/0.79/10%
- y=0.405 (0.267, 0.615)
- n=64, median=36, 75%-CI=18%
- slope=0.6% (5.6, 5.6)
- power=1.0/0.79/10%
- y=0.405 (0.267, 0.615)

Figure 15.5. Cadmium concentrations (ug/g dry weight) in perch liver from Holmoarna and Kvadofjarden (time series starting in 1995). The green area denotes the levels below the suggested target value for cadmium in fish.

Cd, ng/g fresh w., eelpout liver

Holmoarna
- n=83, mean=140.9, median=14
- slope=5.9% (2.0, 7.9)
- y=97.59, 95%-CI=2%
- y=40.5 (26.7, 61.5)
- n=44, median=81, 75%-CI=30%
- slope=1.4% (0.97, 21%
- power=.97/.59/13%
- y=0.405 (0.267, 0.615)
- n=44, median=81, 75%-CI=30%
- slope=1.4% (0.97, 21%
- power=.97/.59/13%
- y=0.405 (0.267, 0.615)

Kvadofjarden
- n=51, mean=135.8, median=14
- slope=-4.1% (4.3, 7.2)
- y=95.79, 95%-CI=1%
- y=36.2 (27.2, 46.6)
- n=64, median=36, 75%-CI=18%
- slope=0.6% (5.6, 5.6)
- power=1.0/0.79/10%
- y=0.405 (0.267, 0.615)
- n=64, median=36, 75%-CI=18%
- slope=0.6% (5.6, 5.6)
- power=1.0/0.79/10%
- y=0.405 (0.267, 0.615)

Vaderomarna
- n=83, mean=140.9, median=14
- slope=5.9% (2.0, 7.9)
- y=97.59, 95%-CI=2%
- y=40.5 (26.7, 61.5)
- n=44, median=81, 75%-CI=30%
- slope=1.4% (0.97, 21%
- power=.97/.59/13%
- y=0.405 (0.267, 0.615)
- n=44, median=81, 75%-CI=30%
- slope=1.4% (0.97, 21%
- power=.97/.59/13%
- y=0.405 (0.267, 0.615)

Figure 15.6. Cadmium concentrations (ug/g dry weight) in eelpout muscle from Holmoarna, Kvadofjarden and Vaderomarna (time series starting in 1995).
Cadmium concentrations in eelpout samples from Holmöarna and Väderöarna showed significant increasing trends; however, the between-year variation at Holmöarna is large (Fig. 15.6).

Cadmium concentrations in blue mussel soft body tissue have shown a significant decreasing trend at Fladen and Kvädöfjärden. Inconsistent concentrations are seen at Väderöarna (Fig. 15.7).

**Figure 15.7.** Cadmium concentrations (µg/g wet weight) in blue mussel soft body tissue from Kvädöfjärden, Fladen and Väderöarna (time series starting in 1981, 1981 and 1995 respectively).

### 15.3.3 Species differences

Significant differences in mean cadmium concentration (µg/g dry weight) were found in fish liver and blue mussel soft body between species marked with a ‘>’:

- **Holmöarna:** Eelpout (2.64) > Perch (0.41)
- **Kvädöfjärden:** Blue mussel (4.3) > Eelpout (1.7) > Perch (0.37)
- **Fladen:** Blue mussel (0.96) > Herring (0.39) > Cod (0.05)
- **Väderöarna:** Blue mussel (1.1) > Herring (0.44) – Eelpout (0.41)

Cadmium concentration in blue mussel soft body tissue is thus about two to nine times higher than the concentration found in fish liver. The concentration in eelpout liver is about twice as high as in perch liver in the analysed samples. The concentration found in guillemot eggs was at least 500 times lower (dry weight) when compared to herring liver.
15.4 Conclusion
Generally, cadmium concentration was higher in samples taken on the Baltic coast compared to samples from the Swedish west coast, with the exception of cod. In regards to temporal variation, the rapid increase in cadmium concentrations observed at Ängskärsklubb and Landsort appears to have stopped, and this trend has now reversed.

Cadmium is concentrated in internal organs, i.e. the liver, whereas the concentration in muscle tissues is very low. Analysed values for perch and herring muscle are 0.8 and 4 ng/g dry weight respectively (Strandmark et al. 2008). Cadmium concentrations of 0.8 - 4 ng/g indicates that there is no immediate risk for human consumption, since the suggested EU limit for human consumption of fish is 160 ng/g fresh weight.

A general remark for extra caution is appropriate when interpreting analyses of low concentrations near the quantification level, as in water or muscle samples. An improved analysis technique may lead to decreasing concentrations due to a decreased risk of sample contamination.
Table 15.1. Trend (in %) for cadmium (ug/g dry weight) assessed from the annual geometric mean in various matrices. The age interval for fish and length interval for blue mussels, the total number of samples and the number of years for the various time-series are shown in the three four columns. Last year’s cadmium concentration values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>Cadmium concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>447</td>
<td>28</td>
<td>81-09</td>
<td>1.2 (.90-1.7)</td>
<td></td>
</tr>
<tr>
<td>Angskärskl. aut.</td>
<td>3-5</td>
<td>450</td>
<td>28</td>
<td>81-09</td>
<td>1.6 (1.1-2.3)</td>
<td></td>
</tr>
<tr>
<td>” spring</td>
<td></td>
<td>132</td>
<td>14</td>
<td>96-09</td>
<td>3.2 (2.1-5.1)</td>
<td></td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>433</td>
<td>26</td>
<td>81-09</td>
<td>1.6 (1.3-2.1)</td>
<td></td>
</tr>
<tr>
<td>Utlangsän, aut.</td>
<td>2-4</td>
<td>432</td>
<td>293</td>
<td>81-09</td>
<td>1.4 (.32, 2.8)*</td>
<td>1.8 (1.4-2.2)</td>
</tr>
<tr>
<td>” spring</td>
<td></td>
<td>122</td>
<td>13</td>
<td>96-09</td>
<td>-3.3 (-6.8,-.13)</td>
<td>1.4 (1.1-1.9)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>515</td>
<td>29</td>
<td>82-09</td>
<td>.39 (.33-.47)</td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>256</td>
<td>14</td>
<td>95-09</td>
<td>4.0 (1.6, 6.3)*</td>
<td>.44 (.36-.54)</td>
<td></td>
</tr>
<tr>
<td>Cod liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>353</td>
<td>25</td>
<td>81-06</td>
<td>-6.0 (-7.8,-4.2)*</td>
<td>.013 (.010-.017)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-4</td>
<td>465</td>
<td>29</td>
<td>81-09</td>
<td>-2.2 (-4.6,-26)</td>
<td>.066 (.044-.098)</td>
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<td>Perch liver</td>
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<tr>
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<td>14</td>
<td>95-09</td>
<td></td>
<td>.41 (.27-.62)</td>
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<tr>
<td>Kvädojfjärden</td>
<td>135</td>
<td>14</td>
<td>95-09</td>
<td></td>
<td>.37 (.27-.50)</td>
<td></td>
</tr>
<tr>
<td>Eelpout liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Holmöarna</td>
<td>98</td>
<td>11</td>
<td>95-07</td>
<td></td>
<td>12 (.45, 19)*</td>
<td>2.0 (1.3-2.1)</td>
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<tr>
<td>Kvädojfjärden</td>
<td>125</td>
<td>15</td>
<td>95-09</td>
<td></td>
<td>1.5 (.89-2.6)</td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>116</td>
<td>14</td>
<td>95-09</td>
<td></td>
<td>5.7 (.42, 11)*</td>
<td>.35 (.23-.54)</td>
</tr>
<tr>
<td>Dab liver</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>3-6</td>
<td>257</td>
<td>14</td>
<td>81-94</td>
<td>3.7 (-4.8,12)*</td>
<td>.81 (.42-1.5)</td>
</tr>
<tr>
<td>Flounder liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>4-6</td>
<td>239</td>
<td>14</td>
<td>81-94</td>
<td>1.7 (-3.7,7.0)*</td>
<td>.53 (.35-.80)</td>
</tr>
<tr>
<td>Blue mussel</td>
<td>shell 1</td>
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<tr>
<td>Fladen</td>
<td>523</td>
<td>27</td>
<td>81-09</td>
<td></td>
<td>-2.2 (-3.3, 1.0)*</td>
<td>.16 (.13-19)</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>531</td>
<td>28</td>
<td>81-09</td>
<td></td>
<td>-2.0 (-3.7,-.12)</td>
<td>.20 (.17-.24)</td>
</tr>
<tr>
<td>Kvädojfjärden</td>
<td>138</td>
<td>14</td>
<td>95-09</td>
<td></td>
<td>-3.2 (-4.6,-.13)</td>
<td>.52 (.40-.68)</td>
</tr>
</tbody>
</table>

Matrix          | age     | n   | yrs  | year | trend % (95% CI) | Cadmium concentration last year (95% CI) |
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
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<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
The analysis of nickel concentration in fish liver started on samples collected in 1995.

16.1 Introduction

16.1.1 Target Levels

The target level (TL) used for nickel in the time series for herring and perch is 670 ug/kg wet weight. For further information on TL and selection of target level see chapter 10. The original TL has been recalculated to dry weight in liver for each time series to fit the presented data. The recalculation of the TL for liver is based on a study that compared the concentration of nickel in the muscle and liver of herring and perch, with the ratio 9 and 5.3 respectively (Strandmark et al, 2008). The recalculation to dry weight is based on the dry weight in each time series. The recalculated target level (Tv) together with the dry percentage (dp) is shown above the statistical information in each time series.

16.2 Methods

16.2.1 Analytical Information

Please note that since 2007, the analytical laboratory for metals changed from the Swedish University of Agricultural Sciences (SLU) to the Department of Applied Environmental Science (ITM) at Stockholm University. See chapter 6 section 6.1 for further details.

16.3 Results

16.3.1 Spatial variation

Significantly lower nickel concentrations were observed in herring liver from Fladen and Väderöarna compared to samples from the Baltic sites (accept for the off shore site in the Bothnian Sea). The overall highest concentrations was observed it the south Bothnian sea and the north Baltic proper (Fig. 16.1).

Mussels from all three sites showed mean levels below the upper limit of the ‘high background concentration at diffuse loading’ in blue mussels for nickel of <5 µg/g dry weight, proposed by Knutzen and Skie (1992) (table 16.1).
16.3.2 Temporal variation
The herring time series from Fladen (Fig. 16.2, 16.3), cod liver from south east of Gotland and Fladen (Fig. 16.4), perch liver from Kväddöfärden (Fig. 16.5), and eelpout from Kväddöfärden and Väderöarna (Fig. 16.6), all show significant decreasing trends. Significant increasing trends for the last ten years were observed for herring at Ångskärsklubb (spring) and Utlängan (spring).
### Figure 16.2 (above) and 16.3 (below). Nickel concentrations (ug/g dry weight) in herring liver from Harufjärden, Ångskärsklubb, Landsort and Utlängan (time series starting in 1995)(above); and Ångskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1996, 1996, 1995 and 1995 respectively). The green area denotes the levels below the suggested target value for nickel in fish.

### Ni, ug/g dry w., herring liver

<table>
<thead>
<tr>
<th>Location</th>
<th>Time (yr)</th>
<th>m (ug/g)</th>
<th>SD(lr)</th>
<th>Power</th>
<th>r²</th>
<th>tao</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Harufjärden</strong></td>
<td>1995-2015</td>
<td>0.083</td>
<td>63%</td>
<td>0.72</td>
<td>NS</td>
<td></td>
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<tr>
<td><strong>Ångskärsklubb</strong></td>
<td>1995-2015</td>
<td>0.083</td>
<td>60%</td>
<td>0.76</td>
<td>NS</td>
<td></td>
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<td><strong>Landsort</strong></td>
<td>1995-2015</td>
<td>0.083</td>
<td>56%</td>
<td>1.00</td>
<td>NS</td>
<td></td>
</tr>
<tr>
<td><strong>Utlängan</strong></td>
<td>1995-2015</td>
<td>0.083</td>
<td>51%</td>
<td>0.34</td>
<td>NS</td>
<td></td>
</tr>
</tbody>
</table>

Ni, ug/g dry w., herring liver

<table>
<thead>
<tr>
<th>Location</th>
<th>Time (yr)</th>
<th>m (ug/g)</th>
<th>SD(lr)</th>
<th>Power</th>
<th>r²</th>
<th>tao</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Angskärsklubb</strong>, spring</td>
<td>1995-2015</td>
<td>0.166</td>
<td>37%</td>
<td>0.97</td>
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<tr>
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<td>1995-2015</td>
<td>0.152</td>
<td>23%</td>
<td>1.00</td>
<td>NS</td>
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<tr>
<td><strong>Fladen (2-3)</strong></td>
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<td>0.063</td>
<td>53%</td>
<td>0.85</td>
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<tr>
<td><strong>Väderöarna</strong></td>
<td>1995-2015</td>
<td>0.039</td>
<td>66%</td>
<td>0.59</td>
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</table>
Figure 16.4. Nickel concentrations (ug/g dry weight) in cod liver from south east Gotland and Fladen (time series starting in 1995).

Figure 16.5. Nickel concentrations (ug/g dry weight) in perch liver from Holmöarna and Kvädöfjärden (time series starting in 1995). The green area denotes the levels below the suggested target value for nickel in fish.
Ni, ng/g fresh w., eelpout liver

Holmoarna

Figure 16.6. Nickel concentrations (ug/g dry weight) in eelpout liver from Holmöarna, Kvädöfjärden and Väderöarna (time series starting in 1995).

Ni, ug/g wet w., blue mussel soft body

Figure 16.7. Nickel concentrations (ug/g dry weight) in blue mussel soft body tissue from Kvädöfjärden, Fladen and Väderöarna (time series starting in 1995).
Figure 16.8. Nickel concentrations (ug/g dry weight) in guillemot eggs Stora Karlsö (time series starting in 1996).

Nickel has only been analysed since 1995 (twelve years) by SLU and for three years by ITM, and therefore the possibilities to detect time trends are limited. For herring the number of years required to detect an annual change of 10% varies between 9 - 17 years. The power to detect an annual change of 10% ranges from 0.59 - 1.0.

16.3.3 Conclusion

Nickel concentration in herring liver is lower on the west coast compared to the east coast of Sweden. Blue mussels examined from all three sites were below the upper limit of the ‘high background concentration at diffuse loading’ in blue mussels for nickel of <5 μg/g dry weight, proposed by Knutzen and Skie (1992).

Inconsistent trends in nickel concentration were seen for all the matrices accept guillemot and cod which showed decreasing trends; however, as analyses of these time series only began in 1995, the ability to detect time trends is still limited.

The measured cadmium concentrations are all below the suggested target level based on food uptake by humans via fish products, of 670 ng/g wet weight.
Table 16.1. Trend (in %) for nickel (ug/g dry weight, wet weight for blue mussels) assessed from the annual geometric mean in various matrices. The age interval for fish and length interval for blue mussels, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s nickel concentration values are estimated from the trend if $p<0.05$ or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
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<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>Nickel concentration last year (95% CI)</th>
</tr>
</thead>
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<td></td>
<td></td>
<td></td>
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<td></td>
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<td>Harufj. autumn</td>
<td>3-5</td>
<td>194</td>
<td>15</td>
<td>95-09</td>
<td>0.083 (0.045-0.15)</td>
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<td>15</td>
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<td>0.11 (0.074-0.15)</td>
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<td>95-09</td>
<td>0.11 (0.079-0.15)</td>
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<td>2-3</td>
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<tr>
<td>Holmöarna</td>
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<td>0.059 (0.026-0.13)</td>
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<tr>
<td>Kvädöfjärden</td>
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<td>0.058 (0.033-0.10)</td>
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<tr>
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<tr>
<td>Holmöarna</td>
<td>98</td>
<td>11</td>
<td>95-07</td>
<td>0.14 (0.095-0.21)</td>
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<tr>
<td><strong>Blue mussel</strong></td>
<td>shell</td>
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<td>Kvädöfjärden</td>
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<td>14</td>
<td>95-09</td>
<td>0.29 (0.22-0.39)</td>
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<tr>
<td>Fladen</td>
<td>234</td>
<td>15</td>
<td>95-09</td>
<td>0.33 (0.26-0.43)</td>
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<td>15</td>
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<td>3.8 (0.00, 7.5)</td>
<td>0.31 (0.22-0.42)</td>
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<tr>
<td>St. Karlsö</td>
<td>140</td>
<td>14</td>
<td>96-09</td>
<td>-6.5 (-13,-0.04)*</td>
<td>0.040 (0.025-0.067)</td>
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</tr>
</tbody>
</table>

* significant trend, $p < 0.05$
The analysis of chromium concentration in fish liver started on samples collected in 1995.

17.1 Introduction

17.1.1 Target Levels

The ‘background concentration at diffuse loading’ in blue mussels for chromium is <3 µg/g dry weight, proposed by Knutzen and Skie (1992).

17.2 Methods

17.2.1 Analytical Information

Please note that since 2007, the analytical laboratory for metals changed from the Swedish University of Agricultural Sciences (SLU) to the Department of Applied Environmental Science (ITM) at Stockholm University. See chapter 6 section 6.1 for further details.
### 17.3 Results

#### 17.3.1 Spatial variation

![Spatial variation in chromium concentration (ug/g dry weight) in herring liver.](Image)

The concentration of chromium in herring liver is quite even along the Swedish coast. Somewhat higher concentrations were observed in herring liver from Harufjärden in the Bothnian Bay and Utlängan in the south Baltic Proper (Fig 17.1).

Chromium concentration in blue mussel samples from the Kattegatt show a geometric mean concentration in the same range as mussels from the Baltic Proper (Fig. 17.4). These concentrations were about two to three times higher compared to samples from the Skagerrak (table 17.1), and close to or above the ‘high background concentration at diffuse loading’ in blue mussels for chromium of $<3 \mu g/g$ dry weight, proposed by Knutzen and Skie (1992). Samples from the Skagerrak were well below this value.

#### 17.3.2 Temporal variation

Chromium decreased significantly in all herring time series(Fig. 17.2, 17.3, in cod from Fladen and south east of Gotland,, in perch from Holmöarna and Kvädojärden, and in guillemot eggs. This decrease is probably explained by a change of method in the analysis for chromium in 2004.
Cr, ug/g dry w., herring liver

Figure 17.2. Chromium concentrations (ug/g dry weight) in herring liver from Harufjärden, Ängskärsklubb, Landsort and Utängan (time series starting in 1995).

The required minimum years to detect an annual change of 10 % varies between 12 - 20 years for herring. The power to detect an annual change of 10 % ranges between 0.45 - 0.99.
**Figure 17.3.** Chromium concentrations (ug/g dry weight) in herring liver from Ångskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1996, 1996, 1995 and 1995 respectively).

Cr, ug/g wet w., blue mussel softbody

**Figure 17.4.** Chromium concentrations (ug/g dry weight) in blue mussel soft body from Kvädöfjärden, Fladen and Väderöarna (time series starting in 1995).
Figure 17.4. Chromium concentrations (ug/g dry weight) in guillemot eggs from St. Karlsö (time series starting in 1996)

17.4 Conclusion

The concentration of chromium in blue mussel soft body tissue at the three sites varied across years, although the geometric mean concentration was in the same range as blue mussels from the Baltic Proper.

Decreases were seen across all time series for all species sampled (accept for blue mussels from Väderöarna); however these decreases might be explained by a change in method for chromium analysis in 2004.
Table 17.1. Trend (in %) for chromium (ug/g dry weight, wet weight for blue mussels) assessed from the annual geometric mean in various matrices. The age interval for fish and length interval for blue mussels, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).  

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<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>Chromium concentration last year (95% CI)</th>
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<td>95-09</td>
<td>-17 (-22, -13)*</td>
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<td>.011 (.006-.020)</td>
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<td>95-09</td>
<td>-21 (-30, -13)*</td>
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</tr>
<tr>
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<td>-22 (-29, -15)*</td>
<td>.012 (.007-.022)</td>
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<td><strong>Cod liver</strong></td>
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<td>95-09</td>
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<tr>
<td>St. Karlsö</td>
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<td>95-09</td>
<td>( )</td>
<td>.18 (.11-.28)</td>
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</table>

* significant trend, p < 0.05
18 Copper - Cu

18.1 Introduction

18.1.1 Usage, Production and Sources

Copper is a nutritionally essential metal, and concentration is regulated by homeostatic mechanisms. Free copper is effectively controlled by metallothionein synthesis (da Silva & Williams 1994) induced by copper itself or by other substances. Although copper is not believed to accumulate with continued exposure, changes found in biological tissues may still reflect changes in concentration of the ambient water.

Copper occurs naturally in rocks, soil, water, sediment and at low levels in air. In its natural (metallic) form, copper can be found in electrical wiring and some water pipes, for example, plumbing, building wire, telecommunications, automotive electrical wiring and air conditioning systems (Dorsey et al. 2004). Copper compounds can be found in alloys such as brass and bronze. Other anthropogenic sources include road run off (Rice et al. 2002) and mining of copper ore. Copper compounds can be commonly found in use in agriculture as fungicides, as wood, leather and fabric preservative, or for water treatment (Dorsey et al. 2004).

18.1.2 Conventions, Aims and Restrictions

The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that copper discharge was to be reduced by 50% between 1985 - 1995, using 1985 as the base year.

The Minister Declaration from 1988, within HELCOM, calls for a reduction in the discharge of copper to air and water by 50% by 1995, with 1987 as the base year.

18.1.3 Target Levels

Average copper concentration in the earth’s crust is 50 ppm (Dorsey et al. 2004). The ‘background concentration at diffuse loading’ in blue mussels for copper is <10 µg/g dry weight, proposed by Knutzen and Skie (1992).

18.2 Methods

18.2.1 Analytical Information

Please note that since 2007, the analytical laboratory for metals changed from the Swedish University of Agricultural Sciences (SLU) to the Department of Applied Environmental Science (ITM) at Stockholm University. See chapter 6 section 6.1 for further details.
18.3 Results

18.3.1 Spatial variation

In herring, no significant differences in mean copper concentration were found between the sampling sites. Somewhat higher concentrations were observed in herring liver from Gaviks fjärden and Angskärsklubb in the Bothnian Sea (Fig 18.1).

![Spatial variation in copper concentration (µg/g dry weight) in herring liver.](image)

**Figure 18.1.** Spatial variation in copper concentration (µg/g dry weight) in herring liver.

The copper concentration in blue mussels from the Swedish west coast is not significantly different compared to blue mussel samples of similar length from a reference site at Kobbefjord, Greenland (Riget et al. 1993). Mussel samples from all three sites showed mean levels below the ‘high background concentration at diffuse loading’ in blue mussels for copper of <10 µg/g dry weight, proposed by Knutzen and Skie (1992).

18.3.2 Temporal variation

It was found that copper significantly increased in herring liver from Utlängan (Fig. 18.1). By contrast, significantly decreasing trends were found at Fladen for herring (Fig. 18.2) and blue mussels (Fladen and Väderöarna) (Fig. 18.4).
Figure 18.1. Copper concentrations (ug/g dry weight) in herring liver from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1981).

Cu, ng/g dry w., herring liver

Figure 18.2. Copper concentrations (ug/g dry weight) in herring liver from Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1996, 1996, 1981 and 1995 respectively).
Figure 18.3. Copper concentrations (ug/g dry weight) in perch liver from Holmöarna and Kvädöfjärden (time series starting in 1995).

Concentrations of copper have increased significantly over the last ten years in perch liver from Holmöarna and Kvädöfjärden (Fig. 18.3). A significantly decreasing trend is seen at Fladen for blue mussels (Fig. 18.4).

Figure 18.4. Copper concentrations (ug/g dry weight) in blue mussel soft body from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1981, 1981 and 1995 respectively).
Cu, ug/g dry w., guillemot egg, early laid

Figure 18.5. Copper concentrations (ug/g dry weight) in guillemot eggs from St. karlsö (time series starting in 1996).

The number of years required to detect an annual change of 10% varied between 7 - 11 years for the herring time series at a power of 80%.

18.3.3 Species differences
Significant differences in mean copper concentration were found in fish liver and blue mussel soft body between species marked with a ‘>’:

Kvädöfjärden: Eelpout (21) > Perch (9) > Blue mussel (7.9)
Fladen: Herring (10) > Cod (8) > Blue mussel (5.4)
Väderöarna: Eelpout (12) > Herring (10) > Blue mussel (5.2)

18.4 Conclusion
There was no significant spatial variation in copper concentration in herring liver. Copper concentration in blue mussels showed no significant difference to blue mussels examined in Kobbefjord, Greenland (Riget et al. 1993).

In general, copper concentration has been inconsistent across all species and sites. Concentrations seen in herring liver over time show a significant increase at Utlängan (autumn), a decrease at Fladen, with no trend detected at the other sites. Concentrations detected in perch liver have increased significantly over the last ten years, whereas a significant decreasing trend has been observed for blue mussels at Fladen only.

Copper concentration in liver from Baltic herring is about 4.5 times higher than the concentration reported from the edible parts of herring. For cod, the concentration in liver is
about 40 - 60 times higher, and for perch about 12 - 14 times. Concentrations in edible parts are reported by Jorhem and Sundström (1993).

Table 18.1. Trend (in %) for copper (ug/g dry weight, wet weight for blue mussels) assessed from the annual geometric mean in various matrices. The age interval for fish and length interval for blue mussels, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s copper concentration values are estimated from trends if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
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<tr>
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<th>Age</th>
<th>n</th>
<th>n yrs</th>
<th>Year</th>
<th>Trend % (95% CI)</th>
<th>Copper concentration last year (95% CI)</th>
</tr>
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<td>Herring liver</td>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>446</td>
<td>28</td>
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<td>451</td>
<td>28</td>
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<td>14 (11-16)</td>
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<tr>
<td></td>
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<td>433</td>
<td>29</td>
<td>81-09</td>
<td>11 (9.3-14)</td>
</tr>
<tr>
<td></td>
<td>Utlängan, aut.</td>
<td>2-4</td>
<td>433</td>
<td>29</td>
<td>81-09 .78 (.08, 1.6)</td>
<td>15 (13-18)</td>
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<tr>
<td></td>
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<td></td>
<td></td>
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</tr>
<tr>
<td></td>
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<td>2-3</td>
<td>515</td>
<td>29</td>
<td>81-09 -1.8 (-2.8, -.76)*</td>
<td>10 (8.5-12)</td>
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<tr>
<td></td>
<td>Väderöarna</td>
<td>256</td>
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<td></td>
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</tr>
<tr>
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<td>397</td>
<td>29</td>
<td>81-09</td>
<td>11 (9.0-14)</td>
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<tr>
<td></td>
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<td>484</td>
<td>29</td>
<td>81-09 -2.5 (-4.5, -.44)</td>
<td>8.6 (6.2-12)</td>
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<tr>
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<td>Holmöarna</td>
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<td>14</td>
<td>95-09</td>
<td></td>
<td>9.0 (6.7-12)</td>
</tr>
<tr>
<td></td>
<td>Kvädojärden</td>
<td>135</td>
<td>14</td>
<td>95-09</td>
<td></td>
<td>9.2 (7.2-12)</td>
</tr>
<tr>
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<td>Holmöarna</td>
<td>98</td>
<td>11</td>
<td>95-07</td>
<td></td>
<td>13 (8.5-19)</td>
</tr>
<tr>
<td></td>
<td>Kvädojärden</td>
<td>125</td>
<td>15</td>
<td>95-09</td>
<td></td>
<td>21 (17-25)</td>
</tr>
<tr>
<td></td>
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<td>14</td>
<td>95-09</td>
<td>4.1 (.37, 8.6)</td>
<td>401 (28-58)</td>
</tr>
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<td>Fladen</td>
<td>3-5</td>
<td>257</td>
<td>14</td>
<td>81-94</td>
<td>18 (14-23)</td>
</tr>
<tr>
<td>Flounder liver</td>
<td>Väderöarna</td>
<td>4-6</td>
<td>239</td>
<td>14</td>
<td>81-94</td>
<td>51 (35-74)</td>
</tr>
<tr>
<td>Blue mussel</td>
<td>Fladen</td>
<td>523</td>
<td>27</td>
<td>81-09</td>
<td>-2.9 (-3.9, -1.9)*</td>
<td>.75 (.64-.88)</td>
</tr>
<tr>
<td></td>
<td>Väderöarna</td>
<td>531</td>
<td>28</td>
<td>81-09</td>
<td>-2.8 (-4.4, -1.2)*</td>
<td>.73 (.56-.95)</td>
</tr>
<tr>
<td></td>
<td>Kvädojärden</td>
<td>138</td>
<td>14</td>
<td>95-09</td>
<td></td>
<td>1.2 (1.0-1.4)</td>
</tr>
<tr>
<td>Guillemot egg</td>
<td>St. Karlsö</td>
<td>140</td>
<td>14</td>
<td>96-09</td>
<td></td>
<td>3.0 (2.7-3.3)</td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
19 Zinc - Zn

The zinc concentration time series in fish liver and blue mussel soft body, presented below, started in 1981.

19.1 Introduction

19.1.1 Usage, Production and Sources

Zinc is a nutritionally essential metal naturally present in some foods. It is a biological requirement for many animals and plants (Zinc factsheet 2011). Zinc concentration is regulated by homeostatic mechanisms. Hence, it is not believed to accumulate with continued exposure, but changes found in biological tissues may still reflect changes in concentration of the ambient water. Zinc occurs naturally in the environment, but most zinc comes from human activities such as mining, steel production and coal burning. In its pure form, anthropogenic sources of zinc can include its use in galvanising steel and iron to prevent rust and corrosion; it is mixed with other metals to form alloys such as brass and bronze; and it is used to make dry cell batteries (Draggan 2008). Zinc compounds are used in industry for things such as making white paints and ceramics, producing rubber, preserving wood and dyeing fabrics (Draggan 2008). Tyre tread material contains approximately 1% weight of zinc. Wear of tyres on road surfaces can contribute a small amount of zinc to the environment (Councell et al. 2004). Some sunscreens use zinc oxide nanoparticles (Osmond & McCall 2010); other zinc compounds can be found in, for example, deodorants, nappy rash cream and anti-dandruff shampoo (Draggan 2008).

19.1.2 Environmental Fate

Zinc is present in water, air and soil. In air, zinc is present mostly as small particles that fall to the earth and drain into waterways with precipitation. Most of this zinc ends up settling in sediment at the bottom of water bodies; however some zinc can remain bound to the soil. Dissolved zinc in water can increase acidity levels (Draggan 2008).

19.1.3 Conventions, Aims and Restrictions

The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that zinc discharges were to be reduced by 50% between 1985 and 1995, using 1985 as the base year.

The Minister Declaration from 1988, within HELCOM, calls for a reduction in the discharge of zinc to air and water by 50% by 1995, with 1987 as the base year.
19.2 Methods

19.2.1 Analytical Information
Please note that since 2007, the analytical laboratory for metals changed from the Swedish University of Agricultural Sciences (SLU) to the Department of Applied Environmental Science (ITM) at Stockholm University. See chapter 6 section 6.1 for further details.

19.3 Results

19.3.1 Spatial variation
In herring, no significant differences in mean copper concentration were found between the sampling sites. Somewhat higher concentrations were observed in herring liver from most of the sampling sites in the Baltic compared to the Swedish west coast (Fig 19.1).

![Figure 19.1. Spatial variation in zinc concentration (ug/g dry weight) in herring liver.](image)

Zinc concentration in cod liver from Fladen was significantly higher than in cod liver from south east of Gotland (table 19.1). This may be explained by the significantly lower fat content in cod liver from Fladen, since zinc concentration is negatively correlated with fat content.

Zinc concentrations in blue mussels from all three investigated sites were below the proposed background concentrations for the North Sea (ICES, 1997) (table 19.1).
19.3.2 Temporal Variation

Significant decreasing trends were seen in herring liver from Ängskärsklubb (spring) and at Väderöarna for the whole period and from Fladen during the last ten years. A significant increasing trend was seen for herring from Utlangan (Fig. 19.2, 19.3). Guillemot eggs show a decreasing trend over the entire period analysed (table 19.1).

Zn, ug/g dry w., herring liver

**Harufjärden (3-5)**
- n(tot)=457, n(yrs)=27
- m=85.6 (79.3, 92.5)
- slope=-.22% (-1.2, .72)
- SD(yr)=20%,1.1%,8 yr
- power=1.0/1.0/5.5%
- y(09)=75.4 (67.4, 84.5)
- r²=.07, NS
- tao=-.23, p<.089
- SD(sm)=3.0, p<.091, 4.7%
- SD(lr)=11%,4.0%,7 yr
- power=1.0/1.0/4.0%

**Angskärsklubb (3-5)**
- n(tot)=437, n(yrs)=27
- m=85.5 (79.3, 92.5)
- slope=.60% (-.35, 1.5)
- SD(yr)=20%,1.5%,9 yr
- power=1.0/377.2%
- y(09)=91 (78, 105)
- r²=.06, NS
- tao=.16, NS
- SD(sm)=3.6, p<.032,5.7%
- SD(lr)=21%,7.6%,9 yr
- power=.96/.96/7.2%

**Landsort (3-5)**
- n(tot)=415, n(yrs)=28
- m=80.7 (76.0, 85.7)
- slope=-.50% (-1.2,.21)
- SD(yr)=15%,1.1%,8 yr
- power=1.0/377.2%
- y(09)=83.1 (71.7, 96.4)
- r²=.00, NS
- tao=-.01, NS
- SD(sm)=3.0, p<.091, 4.7%
- SD(lr)=11%,4.0%,7 yr
- power=1.0/1.0/4.0%

**Utlangan (2-4)**
- n(tot)=437, n(yrs)=28
- m=86.4 (82.1, 90.8)
- slope=-3.0% (-9.6,3.6)
- SD(yr)=20%,1.5%,9 yr
- power=1.0/377.2%
- y(09)=93 (85, 102)
- r²=.12, NS
- tao=.29, p<.033 *
- SD(sm)=2.0, p<.007,3.2%
- SD(lr)=9.4%,3.4%,6 yr
- power=1.0/1.0/3.4%

**Figure 19.2.** Zinc concentrations (ug/g dry weight) in herring liver from Harufjärden, Ängskärsklubb, Landsort and Utlangan (time series starting in 1981).
Zn, ug/g dry w., herring liver

Figure 19.3. Zinc concentrations (ug/g dry weight) in herring liver from Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1996, 1996, 1981 and 1995 respectively).

A significant decreasing trend was seen for cod from Fladen (Fig. 19.4). Zinc concentrations in blue mussel soft body tissue decreased significantly at Fladen and Kvädöfjärden (Fig. 19.5).
**Figure 19.4.** Zinc concentrations (ug/g dry weight) in cod liver from south east Gotland and Fladen (time series starting in 1981).

**Zn, ug/g dry w., cod liver**

**Fat adjusted geometric means**

### SE Gotland (3-4)
- n(tot)=373, n(yrs)=28
- m=33.6 (30.4,37.1)
- slope=0.03% (1.3,1.2)
- SD(y)=27%, 1% 10 yr
- power=1.0/0.9/6.9%
- y(09)=33.4 (27.6,40.7)
- r²=0.0, NS
- tao=.09, NS
- SD(sm)=7.1, NS, 2.9%
- slope=3.4 (4.5,4.4)
- SD(y)=21%, 7.4%, 9 yr
- power=97/97/7.4%
- r²=0.0, NS

### Fladen (2-4)
- n(tot)=442, n(yrs)=28
- m=67.0 (65.7,73.8)
- slope=1.1% (2.2,0.07)
- SD(y)=24%, 1.7%, 10 yr
- power=1.0/0.8/8.7%
- y(09)=57.6 (48.2,68.8)
- r²=0.1, p<.049
- tao=.19, NS
- SD(sm)=4.3, NS, 9.7%
- slope=4.1% (10,66)
- SD(y)=25%, 7.8%, 9 yr
- power=95/95/7.8%
- r²=0.0, NS

**Figure 19.5** Zinc concentrations (ug/g dry weight) in blue mussel soft body from Fladen, Vädreöarna and Kvädöfjärden (time series starting in 1981, 1981 and 1995 respectively).
The number of years required to detect an annual change of 10% varied between 7 - 9 years for the herring time series, with a power of 1.0 to detect a 10% annual change for all of the herring time series.

Zn, ug/g dry w., guillemot egg, early laid

Figure 19.6 Zinc concentrations (ug/g dry weight) in guillemot eggs from St. Karlsö (time series starting in 1996).

19.3.3 Species Differences
Significant differences in mean zinc concentration were found in fish liver and blue mussel soft body, between species marked with a ‘>’:

Holmöarna: Eelpout (138) > Perch (108)
Kvädfjärden: Eelpout (137) > Perch (100)
Fladen: Blue mussel (112) – Herring (75) > Cod (43)
Väderöarna: Blue mussel (108) – Herring (72)

The concentrations in spring-caught herring from Ängskärklubb and Utlängan was considerably higher compared to samples from the same areas in the autumn (table 19.1).

19.4 Conclusion
No significant differences in zinc concentration were observed in herring between sampling sites in the Baltic Sea and the Swedish west coast. Zinc concentration in liver from Baltic herring is about 1.5 times higher than that reported from the edible parts of herring. For cod, the concentration in the liver is about 6 - 8 times higher, and for perch about 3.5 times. Concentrations in edible parts are reported by Jorhem and Sundström (1993).
Zinc concentration in blue mussels from the Swedish west coast was not significantly different compared to blue mussel samples of similar length from a reference site at Kobbefjord, Greenland (Riget et al. 1993).

Over time, zinc concentration has been inconsistent between fish species, with some significant increases and decreases seen between sites.

**Table 19.1.** Trend (in %) for zinc (ug/g dry weight, wet weight for blue mussels) assessed from the annual geometric mean in various matrices. The age interval for fish and length interval for blue mussels, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s zinc concentration values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>Zinc concentration last year (95% CI)</th>
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<tbody>
<tr>
<td>Herring liver</td>
<td></td>
<td></td>
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<td></td>
<td></td>
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<td>Harufj. autumn</td>
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<td>81-09</td>
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<td>27</td>
<td>81-09</td>
<td></td>
<td>83 (72-96)</td>
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<td>14</td>
<td>96-09</td>
<td>-1.9 (-3.7, -.04)*</td>
<td>110 (93-120)</td>
<td></td>
</tr>
<tr>
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<td>28</td>
<td>81-09</td>
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<td>13</td>
<td>96-09</td>
<td>.56 (-.01, 1.1)</td>
<td>93 (85-100)</td>
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<td>490</td>
<td>28</td>
<td>81-09</td>
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<td>75 (66-85)</td>
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<td>81-09</td>
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<td>81-94</td>
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<td></td>
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<td>523</td>
<td>27</td>
<td>81-09</td>
<td>-1.3 (-2.3,-.24)*</td>
<td>13 (12-16)</td>
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</tr>
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<td>81-09</td>
<td></td>
<td>15 (12-19)</td>
<td></td>
</tr>
<tr>
<td>Kvädöfjärden</td>
<td>138</td>
<td>14</td>
<td>95-09</td>
<td>-3.7 (-6.2,-1.3)*</td>
<td>11 (9.3-14)</td>
<td></td>
</tr>
<tr>
<td>Guillemot egg</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>140</td>
<td>14</td>
<td>96-09</td>
<td></td>
<td>42 (36-50)</td>
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</tbody>
</table>

* significant trend, p < 0.05
20.1 Introduction

20.1.1 Uses, Production and Sources
Arsenic is a natural component of the earth’s crust, and found in all environmental media (WHO 2001). Major anthropogenic sources of environmental arsenic contamination are via industrial smelters, coal-fired power plants and production and use of arsenic pesticides and herbicides (Eisler 1994). An estimation of world arsenic production showed that copper chrome arsenate (CCA) used in timber treatment accounts for most arsenic use; however this source has recently decreased due to new arsenic compound regulations, which has seen the industry sector turn to arsenic-free preparations (KEMI).

Elemental arsenic is produced by reduction of arsenic trioxide (As$_2$O$_3$) with charcoal, which in turn is produced as a by-product of metal smelting operations, especially in copper smelting (WHO 2001) (Eisler 2007). Sweden was the world’s leading producer of arsenic trioxide, with ore from the Boliden area containing the highest levels of arsenic (Eisler 2007) (SGU). Dumped chemical munitions from the end of World War II possibly contribute to increased arsenic levels in the Baltic Sea, Skagerak and Kattegatt environment (HELCOM 2010) (OSPAR 2005) (Garnaga et al. 2006).

Marine organisms tend to contain much higher levels of arsenic compared to terrestrial and freshwater organisms. This is due to a low phosphate concentration resulting in a high arsenate:phosphate ratio. The main type of arsenic accumulated in marine organisms is a water-soluble form called arsenobetaine (WHO 2001). This form has a low toxicity and is quickly excreted via urine (SGU 2005) (Eisler 2007).

20.1.2 Toxic Effects
Acute, subacute and chronic arsenic effects can involve a number of organ systems including the respiratory, gastrointestinal, cardiovascular, nervous, and haematopoietic systems and disturbance of liver function, which has been observed in both humans and animals after chronic exposure. Evidence of affects on the heart have been found in humans (WHO 1981).

In general, inorganic arsenic is more toxic than organic arsenic to aquatic biota, with trivalent species being more toxic than pentavalent. The toxic effects are modified by numerous biological factors such as water temperature, pH, organic content, phosphate concentration, suspended solids and the presence of other substances and toxicants (Eisler 1994). Arsenic from water bioaccumulates in aquatic organisms, but there has been no evidence of biomagnification in the food web (Eisler 1994) (SGU 2005).

20.1.3 Conventions, Aims and Restrictions
Restrictions on the use of arsenic as a wood preservative are described in Annex XVII of the EU Regulation (EC) 1907/2006 on the Registration, Evaluation and
Authorisation of Chemicals (REACH).

COMMISSION DIRECTIVE 2006/139/EC (20th December 2006) amending Council Directive 76/769/EEC in regards to restrictions on the marketing and use of arsenic compounds for the purpose of adapting Annex I to technical progress, states that arsenic compounds may not be used in the EU as substances and constituents of preparations intended for, amongst other things, the preservation of wood. Wood treated with arsenic compounds may not be sold on the EU market.

20.1.4 Target Levels
Concentrations in water are usually < 10 μg/l (WHO 2001). Average levels of arsenic in seawater at a salinity of 35 ppm is 2.6 - 3 μg. (SGU 2005). Dissolved arsenic in seawater collected in 1983 from the Baltic Sea was on average 0.76 μg/l, with a range from 0.45 - 1.11 μg/l (Stoeppler et al. 1986).

Within Sweden the natural mean levels of arsenic in sediment is 10 mg/kg dry weight, with variations of 5 - 20 mg/kg dry weight (SGU 2005).

In a study of marine species from the coast of Bohus, the mean concentration of arsenic measured in blue mussels was 10 mg/kg, with a range from 0.39 - 19 dry weight; in eelpout, the mean concentration was 11 mg/kg, with a range from 9 – 13 mg/kg; and in cod liver, the mean concentration was 19 mg/kg with a range from 5 – 37 mg/kg.

20.2 Methods

20.2.1 Analytical Information
Arsenic has only been analysed for a few years within the national Swedish monitoring programme (2007 onwards). See chapter 6 section 6.1 for further details.
20.3 Results

20.3.1 Spatial Variation

Figure 20.1. Spatial variation in arsenic concentration (ug/g dry weight) in herring liver.

The concentration of arsenic in herring liver from the Swedish west coast is higher than the concentration in herring liver from the Baltic. The highest concentration was observed at Fladen (Fig. 20.1).

20.4 Conclusion

A study with several fish species showed that arsenic concentrations are positively correlated with salinity for fish taken from the Baltic Sea and North Sea (Larsen & Francesconi 2003).

The concentration of arsenic in herring liver seems to be higher at the Swedish west coast than in the Baltic, but this result has to be treated with caution since arsenic only has been analysed for three years. This could possibly be explained by the difference in salinity between the Baltic and the North Sea.
21 Silver - Ag

21.1 Introduction

21.1.1 Uses, Production and Sources
Silver is a noble metal (resistant to corrosion and oxidation) that occurs naturally, especially in sulfide-rich ores and in combination with other noble metals and copper, lead and zinc (Eisler 1996) (IVL 2007). The main source of silver today is as a by-product in copper and lead smelting. In Sweden, silver is extracted in a copper mine near Gällivare, a lead mine at Arjeplog, and mines close to Skelefteå (IVL 2007).

Anthropogenic sources of silver come mainly from smelting operations, the manufacture and disposal of certain photographic and electrical supplies, coal combustion and cloud seeding (WHO 2002). Silver is used for jewellery, ornaments, tableware, utensils and currency (Eisler 1996) (IVL 2007) (WHO 2004). Electronics, batteries and solders containing silver may appear as solid waste either deposited in landfills or burnt in waste incinerators. Dispersal of residues in the environment may occur via leakage or emissions to the air (IVL 2007).

In medicine, silver is used for its bactericidal properties. Soluble silver compounds are used as antiseptic and bacteriostatic agents, as disinfectants (WHO 2004); and as antiseptic and antiodour agents in products such as washing machines, refrigerators, socks and shoes (IVL 2007). Metallic silver is used in amalgam dental fillings alloyed with mercury and small amounts of other metals (IVL 2007).

Silver concentration in biota has been found to be higher near sewage outfalls, electroplating plants, mine waste and silver-iodide-seeded areas, than from more distant sites (Eisler 1997).

21.1.2 Toxic Effects
Silver has no known biological function in living organisms (IVL 2007). It occurs naturally in several oxidation states. The most common states are elemental silver Ag\(^0\) and the monovalent ion Ag\(^+\). Soluble silver salts are generally more toxic than insoluble salts. Silver as ionic Ag\(^+\) is one of the most toxic metals known to aquatic organisms in laboratory studies (Eisler 1996) (IVL). The availability of free silver in the marine environment is, however, strongly controlled by salinity due to the affinity of silver to chloride ions (Eisler 1996) (WHO 2002). Silver also has an affinity for suspended particles (Gill et al. 1994). Free silver ion concentrations can range from 47 % when there is a low content of chloride ions and suspended solids, to 0.01 % in marine systems (Gill et al. 1994). In fish, silver has been found to induce the metal-binding protein metallothionein
In seawater the key mechanism of acute toxicity appears to involve osmoregulatory failure (Hogstrand & Wood 1998).

21.1.3 Conventions, Aims and Restrictions

Silver and all of the chemical compounds that emit silver or silver ions, should be regarded as a biocide product if its purpose is to prevent growth of bacteria. Silver used as a biocide product is restricted by the European directive 98/8/EC concerning the placing of biocidal products on the market.

21.1.4 Target Levels

The tolerable daily intake of silver for humans has been set at 5 µg/kg body weight (IRIS 1991). WHO recommendations for protection of groundwater reports a critical concentration of 50 µg/l (WHO 2004).

Silver is comparably rare in the earth’s crust. The crustal abundance is estimated at 0.07 mg/kg, predominantly concentrated in basalt (Eisler 1996). Average concentration of silver in natural waters are 0.2 – 0.3 µg/l (WHO 2004).

In Sweden, the analyses of background concentrations of silver have shown concentrations of 0.07 mg/kg in the fine particulate fraction of moraine, and 0.2 mg/kg in the fine fraction of sediment soils (SGU 2006). In analysed lake sediments, measured concentrations were 0.16 – 0.66 mg/kg dry weight (Grahn et al. 2006), and 5 – 22 mg/kg dry weight (IVL 2007). Background concentrations of silver in fish muscle from lakes have been measured as <0.21 µg/kg fresh weight (IVL 2007).

21.2 Methods

21.2.1 Analytical Information

Arsenic has only been analysed for a few years within the national Swedish monitoring programme (2007 onwards). See chapter 6 section 6.1 for further details.
21.3 Results

21.3.1 Spatial Variation

Figure 21.1. Spatial variation in silver concentration (ug/g dry weight) in herring liver.

The concentration of silver in herring liver from the Bothnian Sea and Bothnian Bay is for the majority of the sampling sites higher than in herring liver from the Baltic Proper and the Swedish west coast (Fig. 21.1).

21.4 Conclusion

The concentration of silver in herring liver seems to be higher in the Bothnian Sea and Bothnian Bay, but this result has to be treated with caution since silver only has been analysed for three years.
22 PCBs, Polychlorinated biphenylenes

Updated 11.03.31

22.1 Introduction

22.1.1 Usage

PCBs have been used in a wide variety of manufacturing processes, especially as plasticizers, insulators and fire retardants. They are widely distributed in the environment due to inappropriate handling of waste material, or for example, leakage from large condensers and hydraulic systems.

The number of possible congeners is 209, as it has one to ten chlorine atoms. Twenty of these congeners have non-ortho chlorine substitutions, and so can attain a planar structure similar to the highly toxic polychlorinated dibenzo-\(p\)-dioxins and dibenzofurans (McKinney et al. 1985; Serico et al. 1991).

22.1.2 Toxicological Effects

The toxicological effects of PCBs on, for example, reproduction in mink, is well documented (Aulerich et al. 1977; Jensen et al. 1977; Bleavins et al. 1980).

22.1.3 Conventions, Aims and Restrictions

In 1992, HELCOM revised the PCBs for which special bans and restrictions on transport, trade, handling, use and disposal were imposed. The Minister Declaration from 1988, within HELCOM, calls for a reduction of stable organic substances by 50\% by 1995, with 1987 as the base year.

The Minister Declaration from 1996, within HELCOM, and the declaration in Esbjerg 1995, calls for measures for toxic, persistent, bioaccumulating substances to have ceased completely by the year 2020.

PCB is one of the initial 12 Persistent Organic Pollutants (POPs) included in The Stockholm Convention on POPs, an international agreement requiring measures for reducing or preventing release of dangerous substances into the environment. The Stockholm Convention was adopted in 2001 and entered into force in 2004. In 1973, the use of PCBs were banned in Sweden, except for sealed systems. In 1978, all new use of PCBs were forbidden.
22.1.4 Target Levels

The target level used for CB-153 in the time series for fish is 0.1 ug/kg wet weight. For further information on target levels and selection of target level see chapter 10. The original target level has been recalculated for each time series based on the lipid percentage. The recalculated target level (Tv) together with the lipid percentage (lp) is shown above the statistical information in each time series.

22.2 Methods

22.2.1 Analytical Information

Seven CB-congeners (CB-28, CB-52, CB-101, CB-118, CB-138, CB-153 and CB-180) are listed as mandatory contaminants that should be analysed and reported within both the OSPARCOM and the HELCOM conventions. In the proposed revised guidelines for OSPARCOM (1996) the congeners CB-105 and CB-156 are added to that list.

See chapter 6, sections 6.2 and 6.3 for further information on analysis methods for PCBs.

The concentration of PCBs in fish muscle, cod liver, blue mussel soft body and guillemot eggs was determined using a gas chromatograph (GC) equipped with an electron capture detector.

Before 1988, PCBs were analysed by a packed column GC. The total sum of PCBs was estimated from 14 peaks after calibration with Aroclor 1254 (Jensen et al. 1983). During 1988, analysis on a capillary column was introduced, allowing analysis of individual congeners (Eriksson et al. 1994). The approximate quantification limit for the capillary column for the analysed congeners is shown in table 22.1.

Although the relative abundance of various CB-congeners is considered fairly constant, both geographical differences and temporal changes in the ratios between the investigated congeners can be shown (see below).

It has been discovered that congener CB-163, and possibly also CB-164, interferes with CB-138 (see also Roos et al. 1990). This implies that the reported concentration of CB-138 also includes a minor contribution from CB-163 and possibly also from CB-164.

The sum of PCBs (sPCB) presented in this report were estimated from the concentration of peak 10 (PCB10) in the chromatogram from packed column chromatography, using the ratio R1=PCB10/sPCB. PCB10 constitutes approximately 11 - 14% of the total amount of PCB in herring; 13 - 15% in cod; 16 - 17% in perch; 12 % in blue mussels; and 18% in guillemot eggs. Thus, the ratio varies between matrices but is very stable within the same matrix at the same sampling site - the coefficient of variation is found, with few exceptions, to be between 3.5 - 6% (see CV1 in table 22.1). From 1989 onwards, PCB10 concentrations were estimated using the ratio R2=(CB-138 + CB-163)/PCB10. CB-138 + CB-163 constitute about 60 - 80% of PCB10, and 7 - 12% of the total sum of PCBs in herring. Mean ratios are given in table 22.2.

The sum of PCBs until 1988 was estimated according to:

\[ sPCB = PCB10 / R_1 \]

and after 1988:

\[ sPCB = (CB-138+CB-163) /(R_1 \cdot R_2) \]
Table 22.1. Mean ratios between peak 10 and the total sum of PCBs from packed column gas chromatography (GC) \( R_1 \), and mean ratios between CB-138+CB-163 (capillary GC) and PCB10 \( R_2 \). The number of analyses \( n \) and the Coefficient of Variation (CV) for the two ratios are given.

<table>
<thead>
<tr>
<th>Fish</th>
<th>Location</th>
<th>( n )</th>
<th>( R_1 )</th>
<th>CV</th>
<th>( n_2 )</th>
<th>( R_2 )</th>
<th>C.I.</th>
<th>CV</th>
<th>( R_1 \cdot R_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring</td>
<td>Harufjärden</td>
<td>169</td>
<td>.14</td>
<td>4.0</td>
<td>19</td>
<td>.73</td>
<td>.67-.76</td>
<td>9.1</td>
<td>.098</td>
</tr>
<tr>
<td></td>
<td>Ångskärsklubb</td>
<td>188</td>
<td>.14</td>
<td>5.1</td>
<td>20</td>
<td>.83</td>
<td>.79-.88</td>
<td>11</td>
<td>.12</td>
</tr>
<tr>
<td></td>
<td>“ spring”</td>
<td>397</td>
<td>.13</td>
<td>5.1</td>
<td>25</td>
<td>.79</td>
<td>.75-.82</td>
<td>11</td>
<td>.10</td>
</tr>
<tr>
<td></td>
<td>Landsort</td>
<td>159</td>
<td>.12</td>
<td>5.2</td>
<td>29</td>
<td>.61</td>
<td>.59-.63</td>
<td>7.4</td>
<td>.070</td>
</tr>
<tr>
<td></td>
<td>Utlängan</td>
<td>94</td>
<td>.12</td>
<td>5.4</td>
<td>20</td>
<td>.65</td>
<td>.62-.68</td>
<td>9.8</td>
<td>.075</td>
</tr>
<tr>
<td></td>
<td>“ spring”</td>
<td>371</td>
<td>.12</td>
<td>5.3</td>
<td>10</td>
<td>.67</td>
<td>.64-.69</td>
<td>5.4</td>
<td>.080</td>
</tr>
<tr>
<td></td>
<td>Fladen</td>
<td>191</td>
<td>.13</td>
<td>5.3</td>
<td>25</td>
<td>.82</td>
<td>.79-.86</td>
<td>10</td>
<td>.11</td>
</tr>
<tr>
<td>Cod</td>
<td>Gotland</td>
<td>152</td>
<td>.14</td>
<td>4.0</td>
<td>11</td>
<td>.69</td>
<td>.65-.72</td>
<td>7.3</td>
<td>.093</td>
</tr>
<tr>
<td></td>
<td>Fladen</td>
<td>176</td>
<td>.15</td>
<td>5.9</td>
<td>10</td>
<td>.85</td>
<td>.81-.89</td>
<td>6.9</td>
<td>.13</td>
</tr>
<tr>
<td>Perch</td>
<td>Holmöarna</td>
<td>140</td>
<td>.17</td>
<td>5.3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Kvädöfjärden</td>
<td>108</td>
<td>.16</td>
<td>6.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dab</td>
<td>Fladen</td>
<td>153</td>
<td>.18</td>
<td>5.9</td>
<td>10</td>
<td>.71</td>
<td>18</td>
<td>.13</td>
<td></td>
</tr>
<tr>
<td>Flounder</td>
<td>Väderöarna</td>
<td>137</td>
<td>.13</td>
<td>9.8</td>
<td>5</td>
<td>.74</td>
<td>11</td>
<td>.096</td>
<td></td>
</tr>
<tr>
<td>Blue mussel</td>
<td>Fladen</td>
<td>5</td>
<td>.12</td>
<td>11.</td>
<td>1</td>
<td>.74</td>
<td>-</td>
<td>.087</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Väderöarna</td>
<td>9</td>
<td>.12</td>
<td>5.6</td>
<td>1</td>
<td>.95</td>
<td>-</td>
<td>.11</td>
<td></td>
</tr>
<tr>
<td>Guillemot</td>
<td>St. Karlsö</td>
<td>211</td>
<td>.18</td>
<td>3.5</td>
<td>30</td>
<td>.77</td>
<td>.74-.80</td>
<td>9.8</td>
<td>.14</td>
</tr>
</tbody>
</table>

Table 22.2. Approximate quantification limit (capillary column, GC) for the analysed CB-congeners.

<table>
<thead>
<tr>
<th>Congener</th>
<th>ng/g, fat weight</th>
</tr>
</thead>
<tbody>
<tr>
<td>CB-28 (2,4,4’-tri CB)</td>
<td>4</td>
</tr>
<tr>
<td>CB-52 (2,2’,5,5’-tetra CB)</td>
<td>4</td>
</tr>
<tr>
<td>CB-101 (2,2’,4,5,5’-penta CB)</td>
<td>4</td>
</tr>
<tr>
<td>CB-118 (2,3’,4,4’,5-penta CB)</td>
<td>5</td>
</tr>
<tr>
<td>CB-138 (2,2’,3,4,4’,5-hexa CB)</td>
<td>6</td>
</tr>
<tr>
<td>CB-153 (2,2’,4,4’,5,5’-hexa CB)</td>
<td>5</td>
</tr>
<tr>
<td>CB-180 (2,2’,3,4,4’,5,5’-hepta CB)</td>
<td>4</td>
</tr>
</tbody>
</table>
22.3 Results

22.3.1 Spatial variation

Figure 22.1. Spatial variation in concentration (ug/g lipid weight) of CB-153 in herring muscle. (N.B. the bar in the most northern part of the Bothnian Bay represents the site Rånefjärden; Harufjärden shows lower concentrations but is hidden behind).

Herring muscle from Ängskärsklubb, Långvindsfjärden and Gaviksfjärden in the Bothnian Sea, Lagnö, Utlängan and Hanö Bay in the Baltic Proper show elevated concentrations of CB-153 compared to Harufjärden in the Bothnian Bay and Fladen; Kullen and Väderöarna (lipid weight) at the west coast (Figure 22.1). However only two years are presented for the offshore sampling sites.

The estimated concentration of CB-153 (wet weight) for 2007-2009 from Harufjärden in the Bothnian Bay showed similar concentration, when compared to Kullen close to Öresund, Fladen in the Kattegatt and Väderöarna in the Skagerrak, and significantly lower concentrations than herring samples from the Bothnian Sea and the Baltic Proper.

The ratio of CB-118:CB-153 is significantly lower at Ängskärsklubb compared to all of the other sites. Herring from Landsort has the highest ratio.

A significant difference was found between CB-153 (lipid weight) concentrations analysed in cod liver from south east of Gotland (higher) and the Kattegatt (lower).
22.3.2 Temporal variation

sPCB concentration (lipid weight) decreased over time across all species examined, with most of these trends being significant (table 22.3). The same decreasing trend is seen in all species examined for CB-153 over time; however in this instance, all decreases seen are significant (table 22.4). The concentration of sPCB (the sum of PCBs estimated from CB-138 or peak 10 from packed column chromatography) in herring muscle from all herring sites in the Baltic and on the west coast, show significant decreases between 1978/80 – 2009 (Fig. 22.2, 22.3). The average decrease varies between -5 and -10% per year. A similar significant decrease within the same range (5 and 10% a year) is also seen in the two time series of spring-caught herring between 1972 – 2009 (Fig. 22.3). This implies a total decrease of PCB concentration in herring muscle of about 70% at Ängskärsklubb and 90% at Karlskrona since the beginning of the 1970s.

Figure 22.2. sPCB concentrations (ug/g lipid weight) in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1978, 1978, 1978 and 1980 respectively).
Figure 22.3. sPCB concentrations (ug/g lipid weight) in herring muscle from Ängskärsklubb (spring), Karlskrona (spring), and Fladen (time series starting in 1972, 1972 and 1980 respectively).

Significant decreasing trends for sPCB are observed in blue mussels from the west coast (Fig. 22.4) and guillemot eggs (1969 - 2009) (Fig. 22.5). The latter trend corresponds to a total decrease of almost 90% since the beginning of the 1970s.
Figure 22.4. sPCB concentrations (ug/g lipid weight) in blue mussels from Fladen and Väderöarna (time series starting in 1984).

Figure 22.5. sPCB concentrations (ug/g lipid weight) in guillemot eggs from St. Karlsö (time series starting in 1969).

CB-153 shows a similar decreasing trend in herring muscle as sPCB at all sites, except for Ängskärsklubb (spring) where no trend is observed (Fig. 22.6, 22.7). Extremely high PCB concentrations are recorded from Landsort in 1996 (Fig. 22.6). This can probably be explained by the very low fat content in herring that year.
CB-153, ug/g lipid w., herring muscle

Harufjärden (3-5)

T = 0.03, l/p = 3.0
\( n = 0.49 (0.046, 0.61) \)
slope = 4.3% (2.7, 6.4)
SD(y) = 1.4% (0.48, 1.5)
y(09) = 0.32 (0.23, 0.45)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

T = 0.02, l/p = 3.0
\( n = 0.49 (0.046, 0.61) \)
slope = 4.0% (0.48, 1.1)
y(09) = 0.12 (0.95, 0.34)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

Tv = 0.01, l/p = 2.9
\( n = 21 (15, 27) \)
slope = 6.5% (5.5, 7.5)
SD(y) = 1.4% (0.48, 1.5)
y(09) = 0.12 (0.95, 0.34)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

T = 0.02, l/p = 2.9
\( n = 0.24 (0.23, 0.52) \)
slope = 4.3% (2.7, 6.4)
SD(y) = 1.4% (0.48, 1.5)
y(09) = 0.32 (0.23, 0.45)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

Tv = 0.02, l/p = 2.9
\( n = 0.24 (0.23, 0.52) \)
slope = 4.0% (0.48, 1.1)
y(09) = 0.12 (0.95, 0.34)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

Figure 22.6. CB-153 concentrations (ug/g lipid weight) in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utllängan (time series starting in 1987, 1989, 1987 and 1988 respectively). The green area denotes the levels below the suggested target value for CB-153 in fish.

CB-153, ug/g lipid w., herring muscle

Angskärsklubb (3-5)

T = 0.03, l/p = 2.9
\( n = 0.24 (0.23, 0.52) \)
slope = 4.3% (2.7, 6.4)
SD(y) = 1.4% (0.48, 1.5)
y(09) = 0.32 (0.23, 0.45)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

T = 0.03, l/p = 2.9
\( n = 0.24 (0.23, 0.52) \)
slope = 4.0% (0.48, 1.1)
y(09) = 0.12 (0.95, 0.34)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

Angskärsklubb, spring

T = 0.03, l/p = 2.9
\( n = 0.24 (0.23, 0.52) \)
slope = 4.3% (2.7, 6.4)
SD(y) = 1.4% (0.48, 1.5)
y(09) = 0.32 (0.23, 0.45)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

T = 0.03, l/p = 2.9
\( n = 0.24 (0.23, 0.52) \)
slope = 4.0% (0.48, 1.1)
y(09) = 0.12 (0.95, 0.34)
\( \tau = 37, p < 0.01 \)
power = 48.48, 15%

Figure 22.7. CB-153 concentrations (ug/g lipid weight) in herring muscle from Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1989, 1987, 1988 and 1995 respectively). The green area denotes the levels below the suggested target value for CB-153 in fish.
The cod time series from south east Gotland in the Baltic Proper and Fladen on the west coast, show significant decreasing trends for sPCB, but no trends are indicated for CB-153 at the two cod sampling sites (Fig. 22.8). In the perch CB-153 time series, concentrations have decreased at both Holmöarna and Kvädöfjärden (Fig. 22.9). CB-153 shows a significant decreasing trend at Kvädöfjärden for eelpout muscle (Fig. 22.10).

### CB-153, µg/g lipid w., cod liver

#### SE Gotland (3-4)

- **n(tot)=173, n(yrs)=21**
- **m=.04, NS**
- **slope=-.07, NS**
- **power=1.0/.50/14%**
- **y(09)=.060 (.019,.217)**
- **r²=.04, NS**
- **tao=-.07, NS**
- **SD(sm)=.53, NS,14%**
- **slope=-4.6%(-16,6.8)**
- **SD(lr)=47%,19%,14 yr**
- **power=.39/.39/17%**
- **r²=.09, NS**

#### Fladen (2-3)

- **n(tot)=166, n(yrs)=20**
- **m=.436 (.367,.517)**
- **slope=-.63%(-3.6,2.3)**
- **power=1.0/.53/14%**
- **y(09)=.410 (.294,.572)**
- **r²=.01, NS**
- **tao=-.03, NS**
- **SD(sm)=.46, NS,12%**
- **slope=-4.8%(-16.6,6.8)**
- **SD(lr)=47%,19%,14 yr**
- **power=.39/.39/17%**
- **r²=.09, NS**

---

**Figure 22.8.** CB-153 concentrations (µg/g lipid weight) in cod liver from south east Gotland and Fladen (time series starting in 1989).
CB-153, ug/g lipid w., perch muscle

Holmoarna

Tv=0.013, lp%=.78
n(tot)=149, n(yrs)=15
slope=-.65%, 14.4 yr
SD(%)=37.7, 13 yr
power=.97, 0.013

Tv=.014, lp%=.71
n(tot)=221, n(yrs)=22
slope=-.52%, 1.7 yr
SD(%)=49.5, 14 yr
power=1.0, .013

Figure 22.9. CB-153 concentrations (ug/g lipid weight) in perch muscle from Holmöarna and Kvädöfjärden (time series starting in 1989 and 1984 respectively). The green area denotes the levels below the suggested target value for CB-153 in fish.

CB-153, ug/g lipid w. Eelpout muscle

Holmoarna

Tv=0.013, lp%=.81
n(tot)=96, n(yrs)=11
slope=-.70%, 14.7 yr
SD(%)=64, 16 yr
power=.252, .027
y(07)=.122 (.059, .253)

Tv=.016, lp%=.61
n(tot)=125, n(yrs)=15
slope=-.70%, 14.7 yr
SD(%)=62, 16 yr
power=.599, NS
y(09)=.082 (.056, .118)

Figure 22.10. CB-153 concentrations (ug/g lipid weight) in eelpout muscle from Holmöarna, Kvädöfjärden and Väderöarna (time series starting in 1995). The green area denotes the levels below the suggested target value for CB-153 in fish.
Decreasing trends are seen for CB-153 in blue mussels and guillemot eggs (Fig. 22.11, 22.12).

### CB-153, ug/g lipid w., blue mussel

**Fladen**
- n(tot)=88, n(yrs)=22
- slope=-7.1% (8.5, -5.6)
- SD(lm)=4.0, 5.6%
- power=0.9 (0.5, 1.4)
- y(09)=0.031 (0.021, 0.042)
- tao=-.75, p<.001
- SD(sm)=1.0, 7.8%

**Väderöarna**
- n(tot)=86, n(yrs)=21
- slope=-5.2% (7.8, -2.5)
- SD(lm)=3.9, 4.0%
- power=1.0 (0.6, 1.4)
- y(09)=0.034 (0.022, 0.042)
- tao=.47, p<.001
- SD(sm)=1.0, 7.8%

**Kvädöfjärden**
- n(tot)=75, n(yrs)=15
- slope=-3.3% (4.4, -2.2)
- SD(lm)=2.0, 2.1%
- power=1.0 (0.6, 0.9)
- y(09)=0.058 (0.047, 0.070)
- tao=.47, p<.001
- SD(sm)=2.0, 2.1%

The number of years required to detect an annual change of 10% for CB-153 varies between 9 - 16 years for the herring, perch, mussel and cod time series.

### CB-153, ug/g lipid w., guillemot egg

**St Karlsö**
- n(tot)=218, n(yrs)=22
- slope=-4.1% (5.2, -3.0)
- SD(lm)=5.0, 5.0%
- power=1.0 (0.7, 1.4)
- y(09)=0.054 (0.046, 0.062)
- tao=-.69, p<.001
- SD(sm)=2.0, 2.0%

**Figure 22.11.** CB-153 concentrations (ug/g lipid weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1988, 1988 and 1995 respectively).

**Figure 22.12.** CB-153 concentrations (ug/g lipid weight) in guillemot eggs from Stora Karlsö (time series starting in 1988).
22.4 Conclusion

PCB concentrations varied between species and sites; however temporally, the concentration of PCBs has decreased by approximately 5 - 10% per year in herring and cod from the Baltic Sea and the Kattegatt, as well as from guillemot eggs and perch from the Baltic Sea since the end of the 1970s.

In all areas, CB-153 concentration is above the suggested target level based on OSPAR BAC (Background Assessment Criteria) of 0.1 wet weight.

Table 22.3 Trend (in %) for sPCB (µg/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s sPCB concentration values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>sPCB concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Herring msc.</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>420</td>
<td>30</td>
<td>78-09</td>
<td>-8.6 (-9.9,-7.2)*</td>
<td>0.16 (.13-.21)</td>
</tr>
<tr>
<td>Ångskärskl. aut.</td>
<td>3-5</td>
<td>359</td>
<td>29</td>
<td>78-09</td>
<td>-7.6 (-8.7,-6.4)*</td>
<td>0.32 (.26-.38)</td>
</tr>
<tr>
<td>” spring</td>
<td>2-5</td>
<td>634</td>
<td>36</td>
<td>72-09</td>
<td>-4.6 (-5.7,-3.5)*</td>
<td>1.1 (0.90-1.5)</td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>402</td>
<td>31</td>
<td>78-09</td>
<td>-5.7 (-6.8,-4.6)*</td>
<td>0.55 (.45-.66)</td>
</tr>
<tr>
<td>Utlångan, aut.</td>
<td>3-4</td>
<td>299</td>
<td>30</td>
<td>80-09</td>
<td>-5.5 (-6.5,-4.4)*</td>
<td>0.48 (.40-.57)</td>
</tr>
<tr>
<td>” spring</td>
<td>2-4</td>
<td>626</td>
<td>35</td>
<td>72-09</td>
<td>-9.4 (-10,-8.5)*</td>
<td>0.59 (.49-.71)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>492</td>
<td>30</td>
<td>80-09</td>
<td>-7.8 (-8.8,-6.8)*</td>
<td>0.13 (.11-.15)</td>
</tr>
<tr>
<td><strong>Cod liver</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>315</td>
<td>29</td>
<td>80-08</td>
<td>-5.8 (-7.3,-4.3)*</td>
<td>0.94 (.74-.12)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>332</td>
<td>28</td>
<td>80-08</td>
<td>-6.0 (-8.1,-3.9)*</td>
<td>1.2 (.85-1.7)</td>
</tr>
<tr>
<td><strong>Perch muscle</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Holmöarna</td>
<td>4-7</td>
<td>269</td>
<td>21</td>
<td>80-07</td>
<td>-8.9 (-11,-7.1)*</td>
<td>0.23 (.18-.31)</td>
</tr>
<tr>
<td>Kvädöfjärden</td>
<td>3-4</td>
<td>232</td>
<td>25</td>
<td>80-08</td>
<td>-8.9 (-11,-6.6)*</td>
<td>0.11 (.076-.16)</td>
</tr>
<tr>
<td><strong>Dab muscle</strong></td>
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<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>3-6</td>
<td>158</td>
<td>13</td>
<td>81-94</td>
<td>-4.6 (-12.2,8)</td>
<td>0.72 (.40-.13)</td>
</tr>
<tr>
<td><strong>Flounder msc</strong></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>4-6</td>
<td>143</td>
<td>15</td>
<td>80-94</td>
<td>-2.8 (-7.4,1.8)</td>
<td>1.7 (1.2-2.6)</td>
</tr>
<tr>
<td><strong>Blue mussel</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>93</td>
<td>24</td>
<td>84-09</td>
<td>-7.3 (-9.0,-5.7)*</td>
<td>0.18 (.14-.23)</td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>94</td>
<td>25</td>
<td>84-09</td>
<td>-6.6 (-8.8,-4.4)*</td>
<td>0.23 (.17-.32)</td>
<td></td>
</tr>
<tr>
<td><strong>Guillemot egg</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>410</td>
<td>39</td>
<td>69-09</td>
<td>-9.0 (-9.5,-8.5)*</td>
<td>10 (9.3-12)</td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
** Pooled samples
Table 22.4. Trend (in %) for CB-153 (µg/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s CB-153 concentration values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>trend % (95% CI)</th>
<th>CB-153 concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring msc.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>297</td>
<td>21</td>
<td>-4.3 (-7.2,-1.4)*</td>
<td>.032 (.023-.045)</td>
</tr>
<tr>
<td>Ångskärskl. aut.</td>
<td>3-5</td>
<td>295</td>
<td>20</td>
<td>-5.7 (-8.1,-3.2)*</td>
<td>.069 (.051-.092)</td>
</tr>
<tr>
<td>” spring</td>
<td></td>
<td>271</td>
<td>21</td>
<td></td>
<td>.18 (.13-.27)</td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>324</td>
<td>23</td>
<td>-5.1 (7.9,-2.3)*</td>
<td>.057 (.040-.071)</td>
</tr>
<tr>
<td>Utlångan, aut.</td>
<td>3-4</td>
<td>286</td>
<td>22</td>
<td>-2.0 (-4.5,0.40)</td>
<td>.075 (.056-.095)</td>
</tr>
<tr>
<td>” spring</td>
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<td>266</td>
<td>21</td>
<td>-4.4 (-6.9,-2.0)*</td>
<td>.11 (.083-.135)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>348</td>
<td>22</td>
<td>-6.7 (-8.5,-5.0)*</td>
<td>.020 (.016-.025)</td>
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<tr>
<td>Väderöarna</td>
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<td>255</td>
<td>14</td>
<td>-5.2 (-9.4,-1.0)*</td>
<td>.014 (.010-.020)</td>
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<tr>
<td>Cod liver</td>
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<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>173</td>
<td>21</td>
<td>.17 (.14,.22)</td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>166</td>
<td>20</td>
<td>.41 (.29,.57)</td>
<td></td>
</tr>
<tr>
<td>Perch muscle</td>
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<td></td>
<td></td>
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<tr>
<td>Holmöarna</td>
<td>149</td>
<td>15</td>
<td>89,95-09</td>
<td>-8.5 (-14,-3.4)*</td>
<td>.039 (.026-.061)</td>
</tr>
<tr>
<td>Kvådfjärden</td>
<td>221</td>
<td>22</td>
<td>84,89-09</td>
<td>-5.2 (-8.8,-1.7)*</td>
<td>.029 (.019-.043)</td>
</tr>
<tr>
<td>Eelpout muscle</td>
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</tr>
<tr>
<td>Holmöarna</td>
<td>96</td>
<td>11</td>
<td>95,97-07</td>
<td></td>
<td>.12 (.059-.25)</td>
</tr>
<tr>
<td>Kvådfjärden</td>
<td>125</td>
<td>15</td>
<td>95-09</td>
<td>-10 (-14,-5.5)*</td>
<td>.082 (0.56-.12)</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>125</td>
<td>15</td>
<td>95-09</td>
<td></td>
<td>.28 (0.16-.48)</td>
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<tr>
<td>Dab muscle</td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>3-6</td>
<td>5</td>
<td>5</td>
<td>89-94</td>
<td>-</td>
</tr>
<tr>
<td>Flounder msc</td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>4-6</td>
<td>6</td>
<td>6</td>
<td>89-94</td>
<td>-</td>
</tr>
<tr>
<td>Blue mussel **</td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>89</td>
<td>22</td>
<td>88-09</td>
<td>-7.1 (-8.5,-5.6)*</td>
<td>.023 (.020-.028)</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>86</td>
<td>21</td>
<td>88-09</td>
<td>-5.2 (-7.8,-2.5)*</td>
<td>.030 (.022-.042)</td>
</tr>
<tr>
<td>Kvådfjärden</td>
<td>75</td>
<td>15</td>
<td>95-09</td>
<td>-2.0 (-3.4,-52)*</td>
<td>.052 (0.046-.058)</td>
</tr>
<tr>
<td>Guillemot egg</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>218</td>
<td>22</td>
<td>88-09</td>
<td>-7.5 (-8.8,-6.2)*</td>
<td>1.8 (1.6-.2)</td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
** Pooled samples
23 DDTs, Dichlorodiphenylethanes

Updated 11.03.31

23.1 Introduction

23.1.1 Conventions, Aims and Restrictions

The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that DDT discharges are to be reduced by 50% between 1985 and 1995, using 1985 as the base year.

In 1992, the Helsinki Convention (HELCOM) revised the DDTs for which special bans and restrictions on transport, trade, handling, use and disposal were imposed. The Minister Declaration from 1988, within HELCOM, calls for a reduction of stable organic substances by 50% by 1995, with 1987 as the base year.

DDT is one of the initial 12 Persistent Organic Pollutants (POPs) included in The Stockholm Convention on POPs, an international agreement requiring measures for reducing or preventing release of dangerous substances into the environment.

The Stockholm Convention was adopted in 2001 and entered into force in 2004.

In Sweden, DDT was partially banned as a pesticide in 1970, and completely banned in 1975 due to its persistence and environmental impact.

23.1.2 Target Levels

The target level used for DDE in the time series for fish is 5 ug/kg wet weight. For further information on target levels and selection of target level see chapter 10. The original target level has been recalculated for each time series based on the lipid percentage. The recalculated target level (Tv) together with the lipid percentage (lp) is shown above the statistical information in each time series.

23.2 Methods

23.2.1 Analytical Information

The concentration of DDTs in fish muscle and blue mussel soft body was determined using a gas chromatograph (GC) equipped with an electron capture detector.

See chapter 6, section 6.2 for further information on analysis methods for DDTs.

Before 1988, DDTs (DDT, DDE, DDD) were analysed on a packed column GC. During 1988, analyses on a capillary column was introduced. The two methods give slightly different results for the various DDT-compounds. In table 23.1, the mean ratio ‘capillary column results’/‘packed column results’ from various sites and matrices are presented.
When the concentrations are close to the quantification limit (DL) for the packed column GC, the results seem to be under-estimated. This is particularly true for the estimated sum of DDTs (sDDT), since DDT and DDD may fall below DL, hence only DDE will constitute the sum. To avoid this bias at low levels, only samples with DDE concentrations above 0.2 µg/g were selected to calculate the ratios below. Only analyses where DDE, DDD and DDT were all present in levels above DL were included in the sDDT ratio. When it was possible to estimate these ratios, they were in general close to one. There were a few exceptions - at Landsort both the DDE and DDT ratios were lower than one, indicating over-estimated concentrations from the packed column possibly due to interference with other compounds in the DDE and DDT peaks in the packed column chromatogram. At Fladen, the DDE ratio was significantly above one, indicating under-estimated DDE concentrations from the packed column GC.

In the time series presented below, DDE is shown for herring, cod, perch, eelpout and blue mussels, and sDDT for flounder, dab and guillemot, where the ratio of one has been used.

Table 23.1. Ratios of DDE, DDT, DDD and sDDT analysed on a capillary column, versus the same samples analysed on a packed column gas chromatography (GC), and the corresponding 95% confidence intervals.

<table>
<thead>
<tr>
<th>Sample Type</th>
<th>n</th>
<th>DDE 95% C.I.</th>
<th>n</th>
<th>DDT 95% C.I.</th>
<th>n</th>
<th>DDD 95% C.I.</th>
<th>n</th>
<th>sDDT 95% C.I.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring muscle</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufjärden</td>
<td>6</td>
<td>1.1 .99-1.2</td>
<td>6</td>
<td>.96 .89-1.0</td>
<td>4</td>
<td>1.5 1.1-2.0</td>
<td>4</td>
<td>1.1 .98-1.2</td>
</tr>
<tr>
<td>Ångskärsklubb</td>
<td>16</td>
<td>1.1 1.0-1.2</td>
<td>-</td>
<td>-</td>
<td>15</td>
<td>.63 .55-.70</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Spring</td>
<td>24</td>
<td>1.0 1.0-1.1</td>
<td>1</td>
<td>.62</td>
<td>21</td>
<td>.77 .68-.85</td>
<td>1</td>
<td>.75</td>
</tr>
<tr>
<td>Landsort</td>
<td>28</td>
<td>.79 .76-.82</td>
<td>28</td>
<td>.75 .67-.81</td>
<td>28</td>
<td>.87 .77-.96</td>
<td>27</td>
<td>.79 .77-.82</td>
</tr>
<tr>
<td>Utlängan</td>
<td>20</td>
<td>1.1 1.0-1.1</td>
<td>20</td>
<td>1.0 .98-1.1</td>
<td>20</td>
<td>1.1 1.1-1.2</td>
<td>20</td>
<td>1.1 1.0-1.1</td>
</tr>
<tr>
<td>Spring</td>
<td>20</td>
<td>1.1 1.1-1.1</td>
<td>10</td>
<td>.81 .74-.88</td>
<td>10</td>
<td>1.1 1.0-1.1</td>
<td>10</td>
<td>1.0 .98-1.1</td>
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<td>Fladen</td>
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<td>1.4 1.3-1.4</td>
<td>5</td>
<td>.90 .77-1.0</td>
<td>6</td>
<td>1.1 1.94-1.3</td>
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<td>1.2 1.1-1.3</td>
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<tr>
<td>Cod liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>6</td>
<td>1.0 .95-1.1</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
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<td>-</td>
</tr>
<tr>
<td>Fladen</td>
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<td>1.1 1.0-1.1</td>
<td>-</td>
<td>-</td>
<td>-</td>
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<tr>
<td>Dab muscle</td>
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<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>9</td>
<td>1.0 .92-1.1</td>
<td>-</td>
<td>-</td>
<td>-</td>
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<td>Flounder muscle</td>
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</tr>
<tr>
<td>Väderöarna</td>
<td>1.0</td>
<td>.86-1.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Guillemot egg</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>30</td>
<td>1.2 1.1-1.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

The quantification limit (capillary column, GC) is estimated to approximately 7 ng/g fat weight for DDE, 4 ng/g for DDD and 3 ng/g for DDT.
23.3 Results

23.3.1 Spatial variation

Figure 23.1. Spatial variation of DDE concentration (ug/g lipid weight) in herring muscle.

The highest concentration of DDE in herring (lipid weight) was detected at Hanöbüken (Figure 23.1) in the south Baltic Proper, and was significantly higher than concentrations detected in the Bothnian Bay and at locations on the Swedish west coast. However, only two years are presented for the offshore sampling sites.

DDE concentrations in cod from the Baltic Proper (south east of Gotland) were about twice as high compared to cod from Fladen on the Swedish west coast (table 23.2).

23.3.2 Temporal variation

DDE concentrations in herring muscle (Fig. 23.2, 23.3), cod and perch (Fig. 23.4), in eelpout from Kvädöfjärden (Fig. 23.5) and in blue mussels (Fig. 23.6) decreased significantly between 1980 - 2009. This decrease varied between 2 - 11% per year (table 23.2). The time series of guillemot eggs (1969 - 2009) showed a significant decrease of -10% per year for DDE (Fig. 23.7). The ratio of DDT/sDDT is significantly decreasing at all herring sites, except for Väderöarna where there are not enough data points to detect a change.
Figure 23.2. DDE concentration (ug/g lipid weight) in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlänget (time series starting in 1978, 1978, 1978 and 1980 respectively). The green area denotes the levels below the suggested target value for DDE in fish.

DDE, ug/g lipid w., herring muscle
Fat adjusted spring herring samples

Figure 23.3. DDE concentration (ug/g lipid weight) in herring muscle from Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1972, 1972, 1980 and 1995 respectively). The green area denotes the levels below the suggested target value for DDE in fish.
DDE, ug/g lipid w., cod liver and perch muscle.

Geometric means, fat adjusted for cod

Figure 23.4. DDE concentration (ug/g lipid weight) in cod liver from south east Gotland, Fladen; and perch muscle from Holmöarna and Kvädöfjärden (time series starting in 1980). The green area denotes the levels below the suggested target value for DDE in fish.
Figure 23.5. DDE concentration (ug/g lipid weight) in eelpout muscle at Holmöarna, Kvädöfjärden and Väderöarna (time series starting in 1995). The green area denotes the levels below the suggested target value for DDE in fish.

Figure 23.6. DDE concentration (ug/g lipid weight) in blue mussel at Fladen, Väderöarna and Kvädöfjärden (time series starting in 1982, 1984 and 1995 respectively).
DDE, ug/g lipid w., guillemot egg, early laid

<table>
<thead>
<tr>
<th>n(tot)</th>
<th>n(ys)</th>
<th>1100</th>
</tr>
</thead>
<tbody>
<tr>
<td>410</td>
<td>39</td>
<td>1000</td>
</tr>
</tbody>
</table>

Figure 23.7. DDE concentration (ug/g lipid weight) in guillemot eggs at Stora Karlsö (time series starting in 1969).

The discharge of fresh DDT during 1983 - 84 (Bignert et al. 1990) is clearly noticeable in the time series from Landsort and Utlängan in the Baltic proper, and Fladen on the Swedish west coast.

The number of years required to detect an annual change of 10% for DDE in herring varied between 11 - 14 years. In general, DDE varies somewhat less between years compared to DDT and DDD. When comparing the power of the DDT time series with other contaminants, it should be noted that the DDT incident of 1983 - 84 decreased the power of the time series calculated from the log-linear regression lines.

23.4 Conclusion

The concentration of DDEs in herring and cod are higher from sites in the Baltic Proper compared to sites on the west coast of Sweden.

The concentration of DDE in herring, perch, cod and blue mussels has decreased at a rate of between 2 - 11% per year from all investigated sites between 1980 - 2009. DDE concentration has decreased by 10% per year in guillemot eggs. DDT has generally decreased faster than the sum of DDTs.

The measured DDE concentrations are above the suggested target level for cod from both the Baltic and the Swedish west coast and for herring from the Baltic Proper and spring caught herring from the south Bothnian Sea. The target level is based on OSPAR EAC (Ecological Assessment Criteria) of 5 ng/g wet weight.
Table 23.2. Trend (in %) for DDE (µg/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s DDE concentration values are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI). For dab and flounder sDDT is estimated (µg/g lipid weight).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>DDE concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring msc.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>477</td>
<td>30</td>
<td>78-09</td>
<td>-9.2 (-11,-7.3)*</td>
<td>.023 (.017-.033)</td>
</tr>
<tr>
<td>Ångskärskl. aut.</td>
<td>3-5</td>
<td>464</td>
<td>29</td>
<td>78-09</td>
<td>-8.2 (-9.4,-6.9)*</td>
<td>.058 (.047-.072)</td>
</tr>
<tr>
<td>” spring</td>
<td>2-5</td>
<td>634</td>
<td>36</td>
<td>72-09</td>
<td>-5.4 (-6.7,-4.2)*</td>
<td>.24 (.18-.31)</td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>456</td>
<td>31</td>
<td>78-09</td>
<td>-5.1 (-6.4,-3.8)*</td>
<td>.17 (.14-.22)</td>
</tr>
<tr>
<td>Utlångan, aut.</td>
<td>3-4</td>
<td>368</td>
<td>30</td>
<td>80-09</td>
<td>-4.2 (-5.5,-2.9)*</td>
<td>.17 (.14-.22)</td>
</tr>
<tr>
<td>” spring</td>
<td>2-4</td>
<td>626</td>
<td>35</td>
<td>72-09</td>
<td>-10 (-11,-9.2)*</td>
<td>.17 (.14-.21)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>523</td>
<td>30</td>
<td>80-09</td>
<td>-8.5 (-9.9,-7.0)*</td>
<td>.020 (.016-.026)</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>254</td>
<td>14</td>
<td></td>
<td>95-09</td>
<td>-6.8 (-11.2,5)*</td>
<td>.014 (.009-.020)</td>
</tr>
<tr>
<td>Cod liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>306</td>
<td>29</td>
<td>80-09</td>
<td>-4.5 (-5.9,-3.1)*</td>
<td>.35 (.28-.44)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>342</td>
<td>29</td>
<td>80-09</td>
<td>-5.1 (-6.5,-3.6)*</td>
<td>.18 (.14-.23)</td>
</tr>
<tr>
<td>Perch muscle</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Holmöarna</td>
<td>289</td>
<td>23</td>
<td></td>
<td>80-09</td>
<td>-11 (-13,-8.8)*</td>
<td>.022 (.016-.030)</td>
</tr>
<tr>
<td>Kvädöfjärden</td>
<td>253</td>
<td>27</td>
<td></td>
<td>80-09</td>
<td>-10 (-13,-7.4)*</td>
<td>.021 (.013-.034)</td>
</tr>
<tr>
<td>Eelpout muscle</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Holmöarna</td>
<td>95</td>
<td>11</td>
<td>95-07</td>
<td></td>
<td>.10 (.057-.18)</td>
<td></td>
</tr>
<tr>
<td>Kvädöfjärden</td>
<td>124</td>
<td>15</td>
<td>95-09</td>
<td>-11 (-17,-4.8)*</td>
<td>.073 (.045-.12)</td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>123</td>
<td>15</td>
<td>95-09</td>
<td></td>
<td>.077 (.050-.12)</td>
<td></td>
</tr>
<tr>
<td>Dab muscle</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>3-5</td>
<td>184</td>
<td>14</td>
<td>81-94</td>
<td>.12 (.062-.23)</td>
<td></td>
</tr>
<tr>
<td>Flounder msc</td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>4-6</td>
<td>163</td>
<td>15</td>
<td>80-94</td>
<td>.11 (.060-.20)</td>
<td></td>
</tr>
<tr>
<td>Blue mussel**</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>92</td>
<td>25</td>
<td>82-09</td>
<td>-7.3 (-9.5,-5.0)*</td>
<td>.020 (.014-.028)</td>
<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>94</td>
<td>25</td>
<td>84-09</td>
<td>-7.8 (-9.7,-6.0)*</td>
<td>.013 (.010-.017)</td>
<td></td>
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<tr>
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<td>15</td>
<td>95-09</td>
<td>-2.3 (-3.9,-.68)</td>
<td>.058 (.050-.066)</td>
<td></td>
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<tr>
<td>Guillemot egg</td>
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<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>410</td>
<td>39</td>
<td>69-09</td>
<td>-10 (-11,-9.3)*</td>
<td>8.1 (6.7-9.7)</td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
** Pooled samples
### Table 23.3. The estimated proportion of DDT, DDE, DDD (%) in various matrices and sites.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n yrs</th>
<th>year</th>
<th>DDT</th>
<th>DDE</th>
<th>DDD</th>
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</thead>
<tbody>
<tr>
<td><strong>Herring msc.</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-4</td>
<td></td>
<td>78-95</td>
<td>33</td>
<td>60</td>
<td>7</td>
</tr>
<tr>
<td>Ängskärskl. aut.</td>
<td>3-5</td>
<td></td>
<td>78-95</td>
<td>17</td>
<td>64</td>
<td>18</td>
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<tr>
<td>Landsort</td>
<td>3-5</td>
<td></td>
<td>78-95</td>
<td>17</td>
<td>51</td>
<td>32</td>
</tr>
<tr>
<td>Utlångan, aut.</td>
<td>2-4</td>
<td></td>
<td>80-95</td>
<td>19</td>
<td>49</td>
<td>32</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td></td>
<td>80-95</td>
<td>22</td>
<td>55</td>
<td>23</td>
</tr>
<tr>
<td><strong>Cod liver</strong></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td></td>
<td>80-95</td>
<td>17</td>
<td>56</td>
<td>27</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-4</td>
<td></td>
<td>80-95</td>
<td>10</td>
<td>76</td>
<td>14</td>
</tr>
<tr>
<td><strong>Perch muscle</strong></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Holmöarna</td>
<td></td>
<td></td>
<td>80-95</td>
<td>5</td>
<td>82</td>
<td>13</td>
</tr>
<tr>
<td>Kvådöfjärden</td>
<td>3-5</td>
<td></td>
<td>80-95</td>
<td>6</td>
<td>85</td>
<td>9</td>
</tr>
<tr>
<td><strong>Blue mussel</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td></td>
<td></td>
<td>81-95</td>
<td>17</td>
<td>63</td>
<td>20</td>
</tr>
<tr>
<td>Väderöarna</td>
<td></td>
<td></td>
<td>80-95</td>
<td>18</td>
<td>65</td>
<td>17</td>
</tr>
</tbody>
</table>
24 HCHs, Hexachlorocyclohexanes

The isomers α-HCH, β-HCH and γ-HCH i.e. lindane, have been analysed in muscle tissue for various fish species (liver tissue for cod), blue mussel soft body and guillemot eggs since 1988 (Table 24.1). Samples from 1987 at Harufjärden and Landsort have been retrospectively analysed. The concentrations of β-HCH are in many cases close to the quantification limit, which implies analytical problems.

24.1 Introduction

24.1.1 Uses, Production and Sources
Technical HCH contains various isomers - 60 - 75% α-HCH; 15% γ-HCH (lindane); 7 - 10% β-HCH; δ-HCH 7%; ε-HCH 1 - 2% - and came into general use in 1950 (Gaul, 1992). The γ-isomer is the most toxic isomer of the HCHs, being 500 - 1000 times as potent as the α-isomer (White-Stevens 1971).

24.1.2 Conventions, Aims and Restrictions
The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that the discharge of HCHs are to be reduced by 50% between 1985 and 1995, using 1985 as the base year.

The Minister Declaration from 1988, within HELCOM, calls for a reduction of stable organic substances by 50% by 1995, with 1987 as the base year.

HCHs are three of the initial 12 Persistent Organic Pollutants (POPs) included in The Stockholm Convention on POPs, an international agreement requiring measures for reducing or preventing release of dangerous substances into the environment. The Stockholm Convention was adopted in 2001 and entered into force in 2004.

In Sweden, the use of lindane was severely restricted in 1970, and subsequently prohibited for use in agriculture in 1978 because of its suspected carcinogenic properties and persistence. Remaining use was banned in 1988/89.

The use of technical HCH stopped in countries around the Baltic between 1970 - 1980. Since 1980, use of lindane in Europe has been allowed only as an insecticide. It was still used to a great extent in France and Italy as recently as 1990 (Yi-Fan et al. 1996).

24.1.3 Target Levels

The target level used for α-HCH and lindane in the time series for fish is 2.6 ug/kg wet weight. For further information on target levels and selection of target level see chapter 10. The original target level has been recalculated for each time series based on the lipid percentage. The recalculated target level (Tv) together with the lipid percentage (lp) is shown above the statistical information in each time series.
24.2 Methods

24.2.1 Analytical Information
See chapter 6, section 6.2 for further information on analysis methods for HCHs.
The quantification limit is estimated to approximately 2 ng/g fat weight for α-HCH, 3 ng/g for β-HCH and 3 ng/g for γ-HCH.

24.3 Results

24.3.1 Spatial Variation

Figure 24.1. Spatial variation of β-HCH concentration (ng/g lipid weight.) in herring muscle.

Somewhat higher concentrations of HCHs (lipid weight) are found in herring samples from the Baltic Proper compared to Bothnian Bay and the Kattegatt, even after the rapid decrease mentioned below (table 24.1, 24.2).

Figure 24.1 shows higher concentrations of β-HCH in herring from the Baltic Proper than in herring from the Bothnian Sea and the Swedish west coast. However, results from the offshore sites are based on analyses from two years only.

The ratio of lindane/α-HCH is higher in the Kattegatt compared to the Baltic in both herring and cod. This could reflect that in the former east-bloc countries, mainly technical HCHs were used, whereas the use of lindane (γ-HCH) was more common in western countries.

24.3.2 Temporal Variation

The variation for α-HCH concentrations in herring muscle was generally low (Fig. 24.2, 24.3). An annual decreasing trend of 13 - 20% was found for herring from all sites (table 24.1). Concentrations in cod liver (Fig. 24.4) have significantly decreased in the time series...
from south east of Gotland and Fladen (in the Kattegatt on the Swedish west coast). Concentrations of α-HCH have also decreased significantly in perch (Fig. 24.4) and eelpout from Kvädöfjärden, Holmöarna and Väderöarna, guillemot eggs from St Karlsö, and in blue mussels (Fig. 24.5) from all sites sampled (Baltic and West coast).

**α-HCH, μg/g lipid w., herring muscle**

**Figure 24.2.** α-HCH concentration (μg/g lipid weight) in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1987, 1989, 1987 and 1988 respectively). The green area denotes the levels below the suggested target value for α-HCH in fish.
### a-HCH, ug/g lipid w., herring muscle

<table>
<thead>
<tr>
<th>Location</th>
<th>Year</th>
<th>n(tot)</th>
<th>n(yrs)</th>
<th>Mean (95% CI)</th>
<th>Slope (95% CI)</th>
<th>Power (95% CI)</th>
<th>R²</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ångskärsklubb, spring (2-5)</td>
<td>2019</td>
<td>260</td>
<td>21</td>
<td>0.00 (0.00, 0.00)</td>
<td>1.01 (0.01, 0.01)</td>
<td>1.00 (1.00, 1.00)</td>
<td>0.98</td>
<td>&lt;0.001 *</td>
</tr>
<tr>
<td>Karlskrona, spring (2-4)</td>
<td>2019</td>
<td>255</td>
<td>20</td>
<td>0.01 (0.00, 0.02)</td>
<td>1.02 (0.02, 0.02)</td>
<td>1.00 (1.00, 1.00)</td>
<td>0.87</td>
<td>&lt;0.001 *</td>
</tr>
<tr>
<td>Fladen (2-3)</td>
<td>2019</td>
<td>348</td>
<td>22</td>
<td>0.00 (0.00, 0.00)</td>
<td>1.01 (0.01, 0.01)</td>
<td>1.00 (1.00, 1.00)</td>
<td>0.61</td>
<td>&lt;0.026 *</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>2019</td>
<td>234</td>
<td>13</td>
<td>0.00 (0.00, 0.00)</td>
<td>1.01 (0.01, 0.01)</td>
<td>1.00 (1.00, 1.00)</td>
<td>0.89</td>
<td>&lt;0.001 *</td>
</tr>
</tbody>
</table>

### a-HCH, ug/g lipid w., cod liver and perch muscle

<table>
<thead>
<tr>
<th>Location</th>
<th>Year</th>
<th>n(tot)</th>
<th>n(yrs)</th>
<th>Mean (95% CI)</th>
<th>Slope (95% CI)</th>
<th>Power (95% CI)</th>
<th>R²</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cod, SE Gotland</td>
<td>2019</td>
<td>185</td>
<td>21</td>
<td>0.00 (0.00, 0.00)</td>
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<td>1.00 (1.00, 1.00)</td>
<td>0.88</td>
<td>&lt;0.001 *</td>
</tr>
<tr>
<td>Cod (2-3), Fladen</td>
<td>2019</td>
<td>178</td>
<td>21</td>
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<td>1.00 (1.00, 1.00)</td>
<td>0.59</td>
<td>&lt;0.026 *</td>
</tr>
<tr>
<td>Perch, Holmöarna</td>
<td>2019</td>
<td>149</td>
<td>15</td>
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<td>1.01 (0.01, 0.01)</td>
<td>1.00 (1.00, 1.00)</td>
<td>0.78</td>
<td>&lt;0.001 *</td>
</tr>
<tr>
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<td>2019</td>
<td>214</td>
<td>21</td>
<td>0.00 (0.00, 0.00)</td>
<td>1.01 (0.01, 0.01)</td>
<td>1.00 (1.00, 1.00)</td>
<td>0.78</td>
<td>&lt;0.001 *</td>
</tr>
</tbody>
</table>

---

**Figure 24.3.** α-HCH concentration (ug/g lipid weight) in herring muscle from Ångskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1989, 1987, 1988 and 1995 respectively). The green area denotes the levels below the suggested target value for a-HCH in fish.

**Figure 24.4.** α-HCH concentration (ug/g lipid weight) in cod liver from south east Gotland and Fladen; and in perch muscle from Holmöarna and Kvädöfjärden (time series starting in 1989 (cod); 1989 and 1984 (perch)). The green area denotes the levels below the suggested target value for a-HCH in fish.
Figure 24.5. α-HCH concentration (ug/g lipid weight) in blue mussel from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1995, 1988 and 1988 respectively).

The number of years required to detect an annual change of 10% is about 8 - 9 years for cod, and varies between 6 - 9 years for the herring time series.

Concentrations of β-HCH are generally decreasing, and are now approaching the quantification limit, making it less suitable for use in this kind of study. The concentrations of β-HCH in some matrices are, however, still detectable and show significant decreasing trends, for example in herring from Ångskärsklubb, Landsort and Utlängan, in cod from south east of Gotland, and in guillemot eggs from St Karlsö (Fig. 24.6).
The concentration of lindane (γ-HCH) has decreased significantly in all analysed matrices at all sampling sites (Fig. 24.7, 24.8, 24.9, 24.10), except for guillemot eggs from St Karlsö. This decrease is in the magnitude of 11 - 15% for herring and blue mussels, 15 - 18% for cod (table 24.2).

The ratio of α-HCH/lindane in herring showed significant decreasing trends from Harufjärden, Landsort and Utlängan.
Figure 24.7. γ-HCH concentration (ug/g lipid weight) herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utšängan (time series from 1987, 1989, 1987 and 1998 respectively). The green area denotes the levels below the suggested target value for lindane in fish.

Figure 24.8. γ-HCH concentration (ug/g lipid weight) herring muscle from Ängskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1989, 1986, 1988 and 1995 respectively). The green area denotes the levels below the suggested target value for lindane in fish.
Figure 24.9. γ-HCH concentration (µg/g lipid weight) in cod liver from south east Gotland and Fladen; and in perch muscle from Holmöarna and Kvädöfjärden (time series starting in 1989 (cod); and 1989 (perch)). The green area denotes the levels below the suggested target value for lindane in fish.

Figure 24.10. γ-HCH concentration (µg/g lipid weight) in blue mussel from south east Gotland and Fladen; and in perch muscle from Kvädöfjärden, Fladen and Väderöarna (time series starting in 1995; 1981, 1983 and 1983).
24.4 Conclusion

There is some variation in HCH concentrations between sites; however as offshore sites have only been sampled for two years, one must treat this trend with caution.

In general, the concentration of HCHs seem to have decreased at a rate of about 9.5% or more per year in various species from the Baltic as well as the Swedish west coast, since the end of the 1980s. From 10 time series on herring, cod and guillemot eggs for 1987 - 95, a median decrease of 65% (38 - 88%) could be estimated. In general, α-HCH is decreasing faster than lindane.

Unlike PCBs, DDTs and HCB, HCHs showed no significant seasonal difference in concentrations between herring caught in spring and autumn.

In all areas, the measured α-HCH and Lindane concentrations, at least during the last 5 years, are below the suggested target level based on EQS set for water translated to concentration in biota, 2.6 ng/g wet weight.

Measures taken to fulfil the aim of the North Sea Conference and the HELCOM Convention of a 50% reduction in HCH discharges by 1995, with 1985 and 1987 as base years, seem to have had a measurable effect in biota.
Table 24.1. Trend (in %) for α-HCH (µg/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s α-HCH concentrations are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>α-HCH concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring msc.</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>294</td>
<td>21</td>
<td>87, 90-99#</td>
<td>-17 (-18,-16)*</td>
<td>**</td>
</tr>
<tr>
<td>Ångskärskl. aut.</td>
<td>3-5</td>
<td>295</td>
<td>20</td>
<td>89-04#</td>
<td>-19 (-21,-17)*</td>
<td>**</td>
</tr>
<tr>
<td>” spring</td>
<td>2-4</td>
<td>260</td>
<td>21</td>
<td>89-09</td>
<td>-18 (-19,-17)*</td>
<td>.002 (.002-.002)</td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>324</td>
<td>23</td>
<td>87-09</td>
<td>-17 (-19,-16)*</td>
<td>.003 (.002,.003)</td>
</tr>
<tr>
<td>Utlångan, aut.</td>
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<td>-18 (-19,-17)*</td>
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<tr>
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<td>2-3</td>
<td>255</td>
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<td>87-09</td>
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<td>.002 (.002-.003)</td>
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<tr>
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<td>88-01#</td>
<td>-14 (-15,-12)*</td>
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<tr>
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<td>13</td>
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<td>Cod liver</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>185</td>
<td>21</td>
<td></td>
<td>89-09</td>
<td>-18 (-20,-17)*</td>
<td>.003 (.002-.003)</td>
</tr>
<tr>
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<td></td>
<td>89-09</td>
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<td>.001 (.001-.001)</td>
</tr>
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<td>Perch muscle</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Holmöarna</td>
<td>149</td>
<td>15</td>
<td></td>
<td>89,95-98#</td>
<td>-7.9 (-11,-4.8)*</td>
<td>**</td>
</tr>
<tr>
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<td>84, 89-01,06#</td>
<td>-12 (-15,-9.4)*</td>
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<tr>
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</tr>
<tr>
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<td>.004 (.001-.013)</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kvädojärden</td>
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</tr>
<tr>
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<td>22</td>
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</table>

# All values at or below quantification limit during recent years
* significant trend, p < 0.05
** No estimated value because of concentrations at or below quantification limit
*** Pooled samples
**Table 24.2.** Trend (in %) for γ-HCH (µg/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s γ-HCH concentrations are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>γ-HCH concentration last year (95% CI)</th>
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<td>Herring msc.</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>297</td>
<td>21</td>
<td>87, 90-01#</td>
<td>-11 (-12,-9.7)*</td>
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<tr>
<td>Ångskärskl. aut.</td>
<td>3-5</td>
<td>295</td>
<td>20</td>
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<td>**</td>
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<tr>
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<td>2-5</td>
<td>261</td>
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<td>89-05#</td>
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<td>**</td>
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<td>23</td>
<td>87-09</td>
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<td>.003 (.003-.004)</td>
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<tr>
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<td></td>
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<td>89-09</td>
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<td>**</td>
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<td>Perch muscle</td>
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<td>149</td>
<td>15</td>
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<td>89,95-98#</td>
<td>-4.5 (-7.5,-1.6)*</td>
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<td>Kvädöfjärden</td>
<td>167</td>
<td>17</td>
<td></td>
<td>89,94-01#</td>
<td>-6.1 (-9.2,-3.0)*</td>
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<td>Eelpout</td>
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<td>**</td>
<td></td>
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<tr>
<td>Fladen</td>
<td>93</td>
<td>24</td>
<td>81,83,88-01#</td>
<td>-13 (-15,-11)*</td>
<td>**</td>
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</tr>
<tr>
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<td>22</td>
<td>83,88-04#</td>
<td>-15 (-17,-12)*</td>
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<td></td>
</tr>
<tr>
<td>Guillemot egg</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>96</td>
<td>13</td>
<td>88-91,93-97,00-01,07#</td>
<td></td>
<td>**</td>
<td></td>
</tr>
</tbody>
</table>

# all values below quantification limit during recent years
* significant trend, p < 0.05
** no estimated value because of concentrations at or below quantification limit
*** Pooled samples

**Table 24.3.** The estimated proportion of α-, β-, γ- HCH (%) in various matrices and sites.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>yrs</th>
<th>year</th>
<th>α</th>
<th>β</th>
<th>γ</th>
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</thead>
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<tr>
<td>Herring msc.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-4</td>
<td>7</td>
<td>87</td>
<td>87,90-95</td>
<td>57</td>
<td>16</td>
<td>27</td>
</tr>
<tr>
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<td>89</td>
<td>89-95</td>
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<td>22</td>
<td>28</td>
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<tr>
<td>” spring</td>
<td>2-5</td>
<td>7</td>
<td>89</td>
<td>89-95</td>
<td>48</td>
<td>26</td>
<td>26</td>
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<tr>
<td>Landsort</td>
<td>3-5</td>
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<td>87</td>
<td>87-95</td>
<td>47</td>
<td>25</td>
<td>28</td>
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<tr>
<td>Utlångan, aut.</td>
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<td>88</td>
<td>88-95</td>
<td>43</td>
<td>27</td>
<td>30</td>
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<tr>
<td>” spring</td>
<td>2-3</td>
<td>7</td>
<td>87</td>
<td>87-95</td>
<td>43</td>
<td>24</td>
<td>33</td>
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<tr>
<td>Fladen</td>
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<td>87</td>
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<tr>
<td>Cod liver</td>
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<tr>
<td>SE Gotland</td>
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<td>87-95</td>
<td>45</td>
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<td>27</td>
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<td>87</td>
<td>87-95</td>
<td>37</td>
<td>11</td>
<td>52</td>
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<tr>
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</tr>
<tr>
<td>Fladen</td>
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<td>81-95</td>
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<td>9</td>
<td>57</td>
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<tr>
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<td>83-95</td>
<td>31</td>
<td>31</td>
<td>9</td>
<td>60</td>
<td></td>
</tr>
</tbody>
</table>
Since 1988, HCB has been analysed in various species (Table 25.1). At Harufjärden and Landsort, samples from 1987 have been retrospectively analysed.

25.1 Introduction

25.1.1 Uses, Production and Sources
The use of the highly persistent HCB as a fungicide is banned in the Baltic countries. Although it may still reach the environment as a by-product of many chlorinating processes, for example pentachlorophenol and vinyl chloride monomer production, we have reason to expect a decrease in biological samples from the Baltic.

25.1.2 Conventions, Aims and Restrictions

The North Sea Conference (1984, 1987, 1990) that covers all routes of pollution into the North Sea, states that HCB discharge was to be reduced by 50% between 1985 and 1995, using 1985 as the base year.

The Minister Declaration from 1988, within HELCOM, calls for a reduction of stable organic substances by 50% by 1995, with 1987 as the base year.

HCB is one of the initial 12 Persistent Organic Pollutants (POPs) included in The Stockholm Convention on POPs, an international agreement requiring measures for reducing or preventing release of dangerous substances to the environment. The Stockholm Convention was adopted in 2001 and entered into force in 2004.

In 1980, HCB was withdrawn from the market in Sweden because of its carcinogenic effects on experimental animals and its persistence.

The use of the highly persistent HCB as a fungicide is banned in the Baltic countries.

25.1.3 Target Levels

The target level used for HCB in the time series for fish is 9.7 ug/kg wet weight. For further information on target levels and selection of target level see chapter 10. The original target level has been recalculated for each time series based on the lipid percentage. The recalculated target level (Tv) together with the lipid percentage (lp) is shown above the statistical information in each time series.
25.2 Methods

25.2.1 Analytical Information

See chapter 6, section 6.2 for further information on analysis methods for HCBs. The quantification limit is estimated to approximately 1 ng/g fat weight.

25.3 Results

25.3.1 Spatial variation

Figure 25.1. Spatial variation of HCB concentration (ng/g lipid weight) in herring muscle.

Herring muscle from Landsort and Utlängan in the Baltic Proper represents the highest HCB concentrations of the herring samples (Fig. 25.1), significantly higher when compared to the other sites during the late 1980s. However, since the concentrations had decreased considerably in samples from the Baltic Proper, and the variance from the Bothnian Bay and the Baltic Sea were large, no significant differences could be seen in the estimated concentrations for 2007-2009 in the autumn-caught herring from the various sites in the Baltic. The estimated concentrations from 2007-2009 were more than three times as high in herring from most of the sites in the Baltic compared to herring from the Swedish west coast.
The results from eelpout and blue mussel samples from Kvädöfjärden that were analysed for HCB for the first time in 1995, indicated that concentrations were at least twice as high in the Baltic compared to the Kattegatt and the Skagerrak. This difference was significant for blue mussels and for eelpout when comparing Holmöarna and Väderöarna.

25.3.2 Temporal Variation

There were significant decreases in HCB concentration in all analysed fish species (Fig. 25.2, 25.3, 25.4, 25.5) and in guillemot eggs (Fig. 25.7).

This decrease is in the magnitude of 4 - 9% for herring, cod, perch and guillemot egg, and 2 - 9% for eelpout (table 25.1).

The number of years required to detect an annual change of 10% is about 12 - 13 years for cod and perch, and varies between 10 - 13 years for the herring time series.
### HCB, ug/g lipid w., herring muscle

#### Harufjärden (3-5)
- HCB, ug/g lipid w., herring muscle
- Harufjärden (3-5)
- TV=.33, lp%=3.0
- y(09)=.02
- r2=.36, p=.003

#### Angskärsklubb (3-5)
- HCB, ug/g lipid w., herring muscle
- Angskärsklubb (3-5)
- TV=.34, lp%=2.9
- y(09)=.02
- r2=.36, p=.003

#### Landsort (3-5)
- HCB, ug/g lipid w., herring muscle
- Landsort (3-5)
- TV=.25, lp%=3.9
- y(09)=.02
- r2=.36, p=.003

#### Utlangan (3-4)
- HCB, ug/g lipid w., herring muscle
- Utlangan (3-4)
- TV=.30, lp%=3.2
- y(09)=.02
- r2=.36, p=.003

### Figure 25.2.
HCB concentration (ug/g lipid weight) in herring muscle from Harufjärden, Angskärsklubb, Landsort and Utlangan (time series starting in 1987, 1989, 1987 and 1988 respectively). The green area denotes the levels below the suggested target value for HCB in fish.

### HCB, ug/g lipid w., herring muscle

#### Ångskärsklubb, spring (2-5)
- HCB, ug/g lipid w., herring muscle
- Ångskärsklubb, spring (2-5)
- TV=.34, lp%=2.9
- y(09)=.02
- r2=.36, p=.003

#### Karlskrona, spring (2-4)
- HCB, ug/g lipid w., herring muscle
- Karlskrona, spring (2-4)
- TV=.34, lp%=2.9
- y(09)=.02
- r2=.36, p=.003

#### Fladen (2-3)
- HCB, ug/g lipid w., herring muscle
- Fladen (2-3)
- TV=.22, lp%=4.5
- y(09)=.02
- r2=.36, p=.003

#### Väderöarna
- HCB, ug/g lipid w., herring muscle
- Väderöarna
- TV=.17, lp%=5.9
- y(09)=.02
- r2=.36, p=.003

### Figure 25.3.
HCB concentration (ug/g lipid weight) in herring muscle from Ångskärsklubb (spring), Karlskrona (spring), Fladen and Väderöarna (time series starting in 1987, 1989, 1988 and 1995 respectively). The green area denotes the levels below the suggested target value for HCB in fish.
Figure 25.4. HCB concentration (ug/g lipid weight) in cod liver from south east Gotland and Fladen, and in perch muscle from Holmöarna and Kvädöfjärden (time series starting in 1989 (cod); 1989 and 1984 perch). The green area denotes the levels below the suggested target value for HCB in fish.

Figure 25.5. HCB concentration (ug/g lipid weight) in eelpout muscle from Holmöarna, Kvädöfjärden and Väderöarna (time series starting in 1995). The green area denotes the levels below the suggested target value for HCB in fish.
Figure 25.6. HCB concentration (ug/g lipid weight) in blue mussel soft tissue from Kvädöfjärden, Fladen and Väderöarna (time series starting in 1988, 1988 and 1995 respectively).

In blue mussels from the Swedish west coast, the concentrations were very low (Fig. 25.6). Since 2000, values were at or below the quantification limit, hence blue mussels are not considered to be a good matrix for monitoring of HCBs in this region.

Figure 25.7. HCB concentration (ug/g lipid weight) in guillemot eggs from Stora Karlsö (time series from 1979).
The number of years required to detect an annual change of 10% varied between 14 - 20 years for the blue mussel series.

25.3.3 Species Differences
At some of the sampling sites, specimens of different species were collected within the same area. HCB was analysed in fish muscle tissue, except for cod where the liver was used, whereas whole soft body was analysed in blue mussels. The mean concentrations (last year) (ng/g lipid weight) found are listed in decreasing order below. Differences in geometric mean HCB concentration among the species samples from the same area are marked with a ‘>’:

Holmöarna: Eelpout (16) > Perch (7)
Kvädöfjärden: Eelpout (11) > Perch (6)- Blue mussel (5)
Fladen: Cod (9) – Herring (6) > Blue mussel (<3)
Väderöarna: Eelpout (8) – Herring (5) > Blue mussel (<3)

The lowest concentrations were found in blue mussels, and the highest were found in guillemot eggs.

Herring caught in the spring showed two to three times higher HCB concentrations on a lipid-weight basis compared to samples collected in autumn.
25.4 Conclusion
Concentrations of HCBs are higher at sites from the Baltic Proper for herring, eelpout, cod and blue mussels, compared to the Swedish west coast, although a considerable decrease in HCBs in herring muscle from the Baltic Proper has been observed.

All time series, where concentrations were compared with the target value based on food uptake by human via fish products of 9.7 ng/g wet weight, were below the suggested level except in cod from south east Gotland.

Since 1988, the concentration of HCB in herring, cod, dab and guillemot egg has decreased at a rate of about 5 - 10% per year from the Baltic Proper. The aim of the North Sea Conference and the HELCOM Convention of a 50% reduction of HCB by 1995, with 1985 and 1987 respectively as base years, thus seems to have been fulfilled.

Table 25.1. Trend (in %) for HCB (µg/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s HCB concentrations are estimated from trends if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>HCB concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Herring msc.</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-5</td>
<td>297</td>
<td>21</td>
<td>87-09</td>
<td>-4.0 (-5.7,-2.2)*</td>
<td>.014 (.011-.017)</td>
</tr>
<tr>
<td>Ångsèlearl. aut.</td>
<td>3-5</td>
<td>283</td>
<td>19</td>
<td>89-09</td>
<td>-7.4 (-11.4,-4.0)*</td>
<td>.009 (.006-.014)</td>
</tr>
<tr>
<td>” spring</td>
<td>262</td>
<td>21</td>
<td></td>
<td>89-09</td>
<td>-5.5 (-8.0,-2.9)*</td>
<td>.040 (.030-.054)</td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>312</td>
<td>22</td>
<td>87-09</td>
<td>-5.4 (-7.9,-3.0)*</td>
<td>.019 (.014-.026)</td>
</tr>
<tr>
<td>Utlingen, aut.</td>
<td>3-4</td>
<td>286</td>
<td>22</td>
<td>88-09</td>
<td>-6.6 (-9.2,-3.9)*</td>
<td>.016 (.011-.022)</td>
</tr>
<tr>
<td>” spring</td>
<td>265</td>
<td>21</td>
<td></td>
<td>87-09</td>
<td>-8.7 (-11.6,-5.6)*</td>
<td>.021 (.016-.028)</td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>347</td>
<td>22</td>
<td>88-09</td>
<td>-7.4 (-9.1,-5.7)*</td>
<td>.005 (.004-.006)</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>234</td>
<td>13</td>
<td></td>
<td>95-09</td>
<td>-5.0 (-9.2,-6.8)*</td>
<td>.005 (.004-.007)</td>
</tr>
<tr>
<td><strong>Cod liver</strong></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>173</td>
<td>21</td>
<td>89-09</td>
<td>-6.2 (-8.8,-3.6)*</td>
<td>.019 (.014-.025)</td>
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<tr>
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<td>2-3</td>
<td>165</td>
<td>20</td>
<td>89-09</td>
<td>-5.3 (-7.9,-2.7)*</td>
<td>.009 (.007-.012)</td>
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<td><strong>Perch muscle</strong></td>
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<tr>
<td>Holmöarna</td>
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<td>89,95-09</td>
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<td>.004 (.003-.006)</td>
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</tr>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Holmöarna</td>
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<td>11</td>
<td>95-07</td>
<td>-9.7 (-18.1,-1.1)*</td>
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<td>Kvädöfjärden</td>
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<td>15</td>
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</tr>
<tr>
<td>Väderöarna</td>
<td>125</td>
<td>15</td>
<td>95-09</td>
<td>-2.3 (-4.9,-2.3)*</td>
<td>.007 (.006-.009)</td>
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<tr>
<td><strong>Dab muscle</strong></td>
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<td></td>
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<td>6</td>
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<td>.004 (.003-.006)</td>
</tr>
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<td></td>
</tr>
<tr>
<td>Väderöarna</td>
<td>4-6</td>
<td>6</td>
<td></td>
<td>89-94</td>
<td></td>
<td>.004 (.001-.028)</td>
</tr>
<tr>
<td><strong>Blue mussel</strong>*</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>83</td>
<td>19</td>
<td>88-00#</td>
<td></td>
<td></td>
<td>**</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>82</td>
<td>19</td>
<td>88-00#</td>
<td></td>
<td>-7.9 (-16.05)*</td>
<td>**</td>
</tr>
<tr>
<td>Kvädöfjärden</td>
<td>75</td>
<td>15</td>
<td>95-09</td>
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<td></td>
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<tr>
<td><strong>Guillemot egg</strong></td>
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<td></td>
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<tr>
<td>St. Karlsö</td>
<td>228</td>
<td>23</td>
<td>79.88-09</td>
<td>-6.4 (-7.9,-4.8)*</td>
<td>.56 (.46-.68)</td>
<td></td>
</tr>
</tbody>
</table>

# all values below quantification limit during recent years
* significant trend, p < 0.05
** no estimated value because of concentrations at or below quantification limit
*** Pooled samples
Dioxins in guillemot eggs from St. Karlsö have been retrospectively analysed in a time series dating back to 1968. Herring muscle tissue has been analysed since 1989.

26.1 Introduction

26.1.1 Uses, Production and Sources

“Dioxins” refer to polychlorinated dibenzo-\(p\)-dioxin (PCDD) and dibenzofuran (PCDF) compounds. Seventeen (10 furans, 7 dioxins) of the 210 possible congeners, substituted in the positions 2,3,7,8, are considered to be of toxicological importance. Some polychlorinated biphenyls (PCBs) are called dioxin-like PCBs (dl-PCBs) because they have a structure similar to that of dioxins and have dioxin-like effects, however, they are not included in this chapter. PCDD/Fs are characterized by low water solubility and low vapor pressure. In the environment, they can undergo photolysis, however, they are generally very resistant to chemical and biological degradation. Due to their persistent and hydrophobic properties, PCDD/Fs accumulate in sediments and organisms in the aquatic environment.

PCDD/Fs are not produced intentionally. They are formed as by-products in several industrial processes and from most combustion processes, such as municipal waste incineration and small-scale burning under poorly controlled conditions. They are also minor impurities in several chlorinated chemical products (e.g. PCBs, chlorophenols, hexachlorophene etc.). Formerly, pulp bleaching using chlorine gas was an important source of PCDD/Fs.

26.1.2 Toxicological Effects

PCDD/Fs can cause a variety of biological and toxicological effects in animals and humans. The most relevant toxic effects are developmental toxicity, carcinogenity and immunotoxicity. Most toxic effects are explained by the binding of PCDD/Fs to the aryl hydrocarbon (Ah) receptor. The sensitivity of various species to the toxic effects of PCDD/Fs vary significantly. 2,3,7,8-TCDD is the most toxic and well-studied congener and is used as a reference for all other related chemicals.

Each of the 17 relevant congeners is assigned a toxic equivalency factor (TEF), where 2,3,7,8-TCDD equals 1 (Van den Berg et al. 1998; Van den Berg et al. 2006). Dioxin concentrations are here reported as TCDD-equivalents (TEQ), which is the sum of the individual congener concentrations multiplied with its specific TEF.
26.1.3 Conventions, aims and restrictions

Dioxins are included in several international agreements, of which the Stockholm Convention and the Convention on Long Range Transboundary Air are among the most important for the control and reduction of sources to the environment. Several EU legislations regulate dioxins, e.g. the plan for integrated pollution prevention and control (IPPC) and directives on waste incineration (EC 2000, 2008). The EU has also adopted a Community Strategy for dioxins, furans and PCBs (EC 2001). PCDD/Fs are currently not included in the Water Framework Directive but are on the list of substances to be revised for adoption in the near future. HELCOM has listed PCDD/Fs and dl-PCBs as prioritized hazardous substances of specific concern for the Baltic Sea (HELCOM 2010), like OSPAR on the List of Chemicals for Priority Action (OSPAR 2010b).

WHO and FAO have jointly established a maximum tolerable human intake level of dioxins via food, and within the EU there are maximum allowable levels of dioxins in food and feed stuff (EC 2006). The European limit for dioxin levels in the muscle tissue of fish is 4 pg/g ww WHO98-TEQ (ΣPCDD/Fs) or 8 pg/g ww WHO98-TEQ (ΣPCDD/Fs + dl-PCBs). PCDD/F levels in fat fish, mainly herring and salmon, from the Baltic Sea often exceed this limit. Sweden and Finland have since 2002 been authorised a derogation from this directive, allowing to sell on the domestic market or to non-member states (EC 2375/2001, EC 201/2002, EC 199/2006, EC 1881/2006).

However, the TEQ levels in herring from the reference sites in this investigation do not exceed the prescribed maximum.

26.1.4 Target Levels

The target level used in the time series for fish (secondary poisoning) is 0.23 ng WHO98-TEQ/kg wet weight. For further information on target levels and selection of target level see chapter 10. When appropriate, the target level (ng/kg ww) has been recalculated to a lipid base (ng/kg lw) for each time series based on the lipid content of the fish muscle. The recalculated target level (Tv) together with the lipid percentage (lp) is shown above the statistical information in Fig. 26.4.

26.2 Methods

26.2.1 Analytical information

See chapter 6, section 6.3 for information on analysis methods for dioxins and dibenzofurans.

* NOTE: This regulation is under amendment and new target levels should be decided during 2011. The new levels will be based on the WHO-2005 TEF-values.
26.3 Results

26.3.1 Spatial Variation

TCDD-eqv

Figure 26.1. Spatial variation in TCDD-equivalents concentration (pg/g lipid weight) in herring muscle.

TCDD-equivalents (pg/g lw) in herring muscle (Figure 26.1) from 2007-2009 (only the two previous years are presented for the offshore sites) are higher in Gaviksfjärden, Kinnbäcksfjärden and Ängskärsklubb (Bothnian Sea), compared to locations in the Baltic Proper and on the Swedish west coast.

26.3.2 Temporal Variation

In guillemot eggs, significant decreasing trends were observed for TCDD, TCDF and total PCDD/Fs (TCDD-equivalents) during the period 1970-2009 (Fig. 26.2, table 26.1). However, contrary to the TCDDs, the TCDFs show no decreasing trend since 1990, which may explain the levelling off of the trend for total PCDD/Fs during the last 20 years (Fig. 26.2).
Figure 26.2. PCDD, PCDF (pg TCDD-eqv/g fat) and PCDD/F (ng TCDD-eqv/g fat) concentrations in guillemot eggs from Stora Karlsö (time series starting in 1970).

The number of years required to detect an annual change of 10% varied between 8 - 12 years in the time series of guillemot.

There were no significant changes in the PCDD/F concentrations over time in herring muscle at Harufjärden, Utlängan and Fladen, either on a wet weight or a lipid weight basis (Fig 26.3 and 26.4). At Ängskärsklubb, however, which had very high levels at the start of the sampling period, a significant decreasing trend is seen (Fig. 26.3 and 26.4). All time-series are above the suggested target value for fish (protection of predators against secondary poisoning). Between 2000-2007, an increasing trend was observed at Harufjärden (Fig. 26.4), but the very low level of TCDD-equivalents in herring from 2008/2009 (table 26.2) eliminated that trend. The low levels of TCDD-equivalents at can not be explained by fat content, weight or length (these parameters were normal) so further investigations are needed.

The number of years required to detect an annual change of 10% varied between 12 - 14 years for these time series.
Figure 26.3. PCDD/F concentrations (pg TCDD-eqv/g fresh weight) in herring muscle from Harufjärden, Ångskärsklubb, Utlängan and Fladen (time series starting in 1990, 1979, 1988 and 1990 respectively). The green area denotes the levels below the suggested target value for PCDD/Fs in fish (secondary poisoning).

Figure 26.4. PCDD/F concentrations (pg TCDD-eqv/g fat weight) in herring muscle from Harufjärden, Ångskärsklubb, Utlängan and Fladen (time series starting in 1990, 1979, 1988 and 1990 respectively). The green line denotes the suggested target value for PCDD/Fs in fish (secondary poisoning).
26.4 Conclusion

In guillemot eggs, significant decreasing trends were observed for TCDD, TCDF and PCDD/Fs during 1970-2009. For TCDFs, no trend can be observed between 1990-2009.

In all areas, the PCDD/F concentrations are above the suggested target level for concentrations in fish for protection of predators against secondary poisoning of 0.23 pg/g wet weight.

PCDD/F concentrations are higher in herring muscle from the Bothnian Sea compared to the Baltic Proper and the Swedish west coast. In all areas, the PCDD/F concentrations are above the suggested target level for concentrations in fish for protection of predators against secondary poisoning. Except from Ångskärsklubb, no decreasing trends of PCDD/Fs in herring muscle can be observed in the last 20 years.
Table 26.1. Trend (in %) of PCDD/F concentrations in herring (pg TCDD-eqv/g lipid weight) and guillemot eggs (ng TCDD-eqv/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval, the total number of samples and the number of years for the various time-series are shown in the first three columns. The last year’s PCDD/F concentrations are estimated from the trend if \( p < 0.05 \) or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>TDCC-equivalent concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring msc. **</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj. autumn</td>
<td>3-4</td>
<td>137</td>
<td>19</td>
<td>90-09</td>
<td>.72 (.50-.1.0)</td>
<td></td>
</tr>
<tr>
<td>Ångskärsklubb</td>
<td>3-4</td>
<td>29</td>
<td>26</td>
<td>79-09</td>
<td>.77 (.53-1.1)</td>
<td></td>
</tr>
<tr>
<td>Utlångan</td>
<td>3-4</td>
<td>177</td>
<td>19</td>
<td>90, 92-09</td>
<td>.58 (.42-.82)</td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>138</td>
<td>20</td>
<td>90-09</td>
<td>.45 (.33-.60)</td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, \( p < 0.05 \)
** Pooled samples

Table 26.2. Trend (in %) of PCDD/F concentrations in herring (pg TCDD-eqv/g fresh weight) assessed from the annual geometric mean in herring muscle. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first three columns. Last year’s PCDD/F concentrations are estimated from trends if \( p < 0.05 \) or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>TCDD-equivalent concentration last year (95% CI)</th>
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<td>19</td>
<td>90, 92-09</td>
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<td>Fladen</td>
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<td>138</td>
<td>20</td>
<td>90-09</td>
<td>.45 (.33-.60)</td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, \( p < 0.05 \)
** Pooled samples
Polybrominated flame retardants in guillemot eggs from St. Karlsö have been retrospectively analysed in a time series dating back to 1968. Herring muscle tissue has also been analysed during recent years. Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) were included in these trend studies.

27.1 Introduction

27.1.1 Uses, Production and Sources

PBDEs are produced as three different technical products; penta-, octa and deca BDE. Each of these products include a few major congeners. For pentaBDE these are BDE-47, -99, and-100. OcatBDE contains mainly BDE-183, while decaBDE includes almost exclusively BDE-209 (LaGuardia et al. 2006). HBCDD is produced as a mixture of three stereoisomers α-, β- and γ-HBCDD (Covaci et al. 2006). Both PBDE and HBCDD are used as additive flame retardants incorporated into materials such as plastics and textiles in products that need to be prevented from catching fire.

Leakage of these substances to the environment occurs from production and use of products, and long-range transport via air borne particles. The PBDE congeners that are most commonly found in fish are BDE-47, -99 and -100, while PBDE congeners with a higher degree of bromination are more common in the terrestrial environment.

27.1.2 Toxic effects

Several PBDE congeners and HBCDD have been shown to cause neurotoxic effects in rats and mice. Animals exposed to PBDEs and HBCDD during a sensitive stage of brain development have later shown reduced memory and learning abilities (Viberg 2004; Eriksson et al. 2006). Brominated flame retardants (BFR) are also considered to be endocrine disruptors, and in particular, effects on the thyroid hormone system are seen (Darnerud 2008).

27.1.3 Conventions, aims and restrictions

The PBDEs, tetrabromodiphenyl ether, pentabromodiphenyl ether, hexabromodiphenyl ether and heptabromodiphenyl ether are among the nine new Persistent Organic Pollutants (POPs) included in The Stockholm Convention on POPs. Within the EU, the penta- and octaBDE products were banned for use in 2004. A Swedish ban of decaBDE was established in 2007, but this ban was withdrawn when decaBDE was included in the RoHS directive in 2008. PBDEs are also on the list of prioritized substances within the Water Framework Directive.
HBCDD is under review by the **Persistent Organic Pollutants Review Committee (POPRC)** as a proposed substance to be listed under the Stockholm Convention (Arnot et al. 2009).

### 27.1.4 Target Levels

The target level used for HBCDD in the time series for fish is 167 ug/kg wet weight. For further information on target levels and selection of target level see chapter 10. The original target level has been recalculated for each time series based on the lipid percentage. The recalculated target level (Tv) together with the lipid percentage (lp) is shown above the statistical information in each time series.

### 27.2 Methods

#### 27.2.1 Analytical information

See chapter 6, section 6.2 for further information regarding analytical methods for BFRs.
27.3 Results

27.3.1 Spatial variation

Figure 27.1. Spatial variation of HBCDD concentration (ng/g lipid weight) in herring muscle.
Elevated concentrations of HBCDD (lipid weight) at the sampling sites in the south Baltic Proper (only two years are presented for the offshore sites) are seen (Fig. 27.1). Figure 27.2 and the herring time series indicate elevated concentrations of some of the substances at Karlskrona in the southern Baltic Proper. In general, PBDEs and HBCDD seem to be more evenly distributed among sites compared to e.g. PCB.

### 27.3.2 Temporal Variation

Significant increasing concentrations of BDE-47, BDE-100 and BDE-99 in guillemot eggs from the late 1960s until the early 1990s, are followed by decreasing values during the more recent period (Fig. 27.3).

Significant decreasing concentrations of BDE-47 are observed in herring from Harufjärden, Ängskärsklubb (autumn), Landsort, Utlängan (autumn) and Väderöarna, in cod and herring from Fladen and in blue mussels from all sampling sites (Fig. 27.4-7, Table 27.1). The number of years required to detect an annual change of 10% in the concentration of BDE-47 is 8 to 16 years for herring, cod and blue mussel.

Significant decreasing concentrations of BDE-99 are observed in herring from Harufjärden, Ängskärsklubb (autumn), Landsort, Utlängan (autumn), Fladen and Väderöarna (Fig. 27.8, 27.8). No trend in either direction is detected from cod at either sites sampled (Fig. 27.10). Blue mussels from Kvädöfjärden show decreasing concentrations (27.11).
Significant decreasing concentrations of BDE-153 are observed in herring from Utlängan (autumn) (27.12), Fladen and Väderöarna (Fig. 27.13); a non-significant decreasing trend is seen at Ångskärsklubb (spring) for herring (Fig. 27.13) and south east Gotland for cod (Fig. 27.14).

**Figure 27.3.** Temporal trends of BDE-47, -99, and 100 in guillemot eggs (time series starting in 1968).

**Figure 27.4.** Temporal trends of BDE-47 in herring muscle from Harufjärden, Ångskärsklubb, Landsort and Utlängan (time series starting in 1999).
Figure 27.5. Temporal trends of BDE-47 in herring muscle from Fladen, Väderöarna, Ångskärsklubb (spring) and Karlskrona (spring) (time series starting in 1999).

Figure 27.6. BDE-47 concentrations in cod liver from south east Gotland and Fladen (time series starting in 1999).
Figure 27.7. BDE-47 concentrations in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 2000).

Figure 27.8. BDE-99 concentrations in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1999).
Figure 27.9. BDE-99 concentrations in herring muscle from Fladen, Väderöarna, Ängskärsklubb and Karlskrona (spring) (time series starting in 1999).

Figure 27.10. BDE-99 concentrations in cod liver from south east Gotland and Fladen (time series starting in 1999).
Figure 27.11. BDE-99 concentrations in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 2000).

Figure 27.12. BDE-153 concentrations in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1999).
Figure 27.13. BDE-153 concentrations in herring muscle from Fladen, Väderöarna, Ängskärrsklubb and Karlskrona (spring) (time series starting in 1999, 1999, 2003 and 2002 respectively).

Figure 27.14. BDE-153 concentrations in cod liver from south east Gotland and Fladen (time series starting in 1999).

HBCD in herring is decreasing at Utlängan in the Baltic Proper (8.3% per year, table 27.2) (Fig. 27.15), Väderöarna (15% per year, table 27.2) and Fladen (16% per year, table 27.2) (Fig. 27.16) and in cod at Fladen (10% per year, table 27.2) (Fig. 27.17). A non-significant
A decreasing trend is seen in blue mussels at Fladen (Fig. 27.18). Concentrations of HBCDD are increasing in guillemot eggs by about 3% per year (table 27.2, Fig. 27.19).

The number of years required to detect an annual change of 10% in the concentration of HBCDD is 10 - 15 years for herring and cod, and 12 years for guillemot egg.

**HBCDD, ng/g lipid w., herring muscle**

<table>
<thead>
<tr>
<th>Location</th>
<th>Mean (95% CI)</th>
<th>SD (95% CI)</th>
<th>Power (95% CI)</th>
<th>Y(09) (95% CI)</th>
<th>R²</th>
<th>Tauo</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harufjärden</td>
<td>m=8.08 (6.03,12.2)</td>
<td>3.0%</td>
<td>0.30</td>
<td>0.40 (5.8,21.9)</td>
<td>0.12</td>
<td>NS</td>
</tr>
<tr>
<td>Ängskärsklubb</td>
<td>m=5.03 (3.52,7.2)</td>
<td>3.0%</td>
<td>0.30</td>
<td>0.38 (1.82,8.23)</td>
<td>0.09</td>
<td>NS</td>
</tr>
<tr>
<td>Landsort</td>
<td>m=12.6 (10.6,15.1)</td>
<td>3.0%</td>
<td>0.30</td>
<td>13.5 (8.6,18.1)</td>
<td>0.03</td>
<td>NS</td>
</tr>
<tr>
<td>Utlängan</td>
<td>m=15.2 (11.1,20.9)</td>
<td>3.0%</td>
<td>0.30</td>
<td>10.0 (6.0,16.7)</td>
<td>0.03</td>
<td>NS</td>
</tr>
</tbody>
</table>

**Figure 27.15.** HBCDD concentrations (ng/g lipid weight) in herring muscle from Harufjärden, Ängskärsklubb, Landsort and Utlängan (time series starting in 1999). The green area denotes the levels below the suggested target value for HBCDD in fish.
Figure 27.16. HBCDD concentrations (ng/g lipid weight) in herring muscle from Ängskärsklubb (spring), Karlskrona (spring), Fladen, and Väderöarna (time series starting in 2002, 2002, 1999 and 1999 respectively). The green area denotes the levels below the suggested target value for HBCDD in fish.

Figure 27.17. HBCDD concentrations (ng/g lipid weight) in cod liver from south east Gotland and Fladen (time series starting in 1996). The green area denotes the levels below the suggested target value for HBCDD in fish.
Figure 27.18. HBCDD concentrations (ng/g lipid weight) in blue mussel from Fladen, Väderöarna and Kvädöfjärden (time series starting in 2000).

Figure 27.19. HBCDD concentrations in guillemot eggs from Stora Karlsö (time series starting in 1969).
27.4 Conclusions

Elevated levels of HBCDD are seen in sites from the southern Baltic Proper. This includes data from the last 2 years for the offshore sites, where the trend should be treated with caution. PBDEs and HBCDD are more evenly distributed among sites compared to e.g. PCBs.

A significant increase in BDE-47, 99 and 100 has been seen in guillemot eggs since the late 1960s until the early 1990s, where concentrations then began to show a decrease. For herring, cod, and blue mussels, HBCDD and BDE-47, 99 and 153 decreased at some sites and showed inconsistent trends at other sites, whereas a 3% per year increase in HBCDD has been seen in guillemot eggs. Therefore, future changes in BFR concentrations should be examined carefully for temporal changes.

In all areas, HBCDD concentrations are below the suggested target level for concentrations in fish based on protection of predators against secondary poisoning of 167 ng/g wet weight.

Table 27.1. Trend (in %) for BDE-47 (ng/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s BDE-47 concentrations are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>BDE-47 concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring msc.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj.</td>
<td>3-5</td>
<td>130</td>
<td>11</td>
<td>99-09</td>
<td>4.7 (2.9-7.7)</td>
<td></td>
</tr>
<tr>
<td>Ångskärsklubb</td>
<td>3-5</td>
<td>120</td>
<td>10</td>
<td>99-09</td>
<td>-8.8 (-15,-2.4)*</td>
<td>3.5 (2.4-5.3)</td>
</tr>
<tr>
<td>Ångskärsklubb</td>
<td>2-5</td>
<td>70</td>
<td>8</td>
<td>02-09</td>
<td>13 (5.7-30)</td>
<td></td>
</tr>
<tr>
<td>Ångskärsklubb</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>131</td>
<td>11</td>
<td>99-09</td>
<td>-8.5 (-15,-2.4)*</td>
<td>4.3 (3.0-6.2)</td>
</tr>
<tr>
<td>Utlångan</td>
<td>3-4</td>
<td>132</td>
<td>11</td>
<td>99-09</td>
<td>-5.5 (-11,.11)</td>
<td>6.6 (4.8-9.2)</td>
</tr>
<tr>
<td>Utlångan</td>
<td>2-4</td>
<td>60</td>
<td>7</td>
<td>03-09</td>
<td>13 (6.6-24)</td>
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</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>132</td>
<td>11</td>
<td>99-09</td>
<td>-16 (-20,-13)*</td>
<td>1.5 (1.3-1.9)</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>176</td>
<td>10</td>
<td>99-09</td>
<td>-16 (-22,-11)*</td>
<td>1.6 (1.2-2.3)</td>
<td></td>
</tr>
<tr>
<td>Cod liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>107</td>
<td>11</td>
<td>99-09</td>
<td>18 (13-24)</td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>106</td>
<td>11</td>
<td>99-09</td>
<td>-20 (-27,-12)*</td>
<td>7.9 (5.2-12)</td>
</tr>
<tr>
<td>Guillemot egg</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>207</td>
<td>35</td>
<td>68-09</td>
<td>99-09</td>
<td>37 (18-76)</td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
Table 27.2. Trend (in %) for HBCDD (ng/g lipid weight) assessed from the annual geometric mean in various matrices. The age interval for fish, the total number of samples and the number of years for the various time-series are shown in the first four columns. Last year’s HBCDD concentrations are estimated from the trend if p<0.05 or from the mean if no trend is present. Numbers in brackets are 95% confidence intervals (CI).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>age</th>
<th>n</th>
<th>n yrs</th>
<th>year</th>
<th>trend % (95% CI)</th>
<th>HBCDD concentration last year (95% CI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Herring msc.</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Harufj.</td>
<td>3-5</td>
<td>130</td>
<td>11</td>
<td>99-09</td>
<td>11 (5.8-22)</td>
<td></td>
</tr>
<tr>
<td>Ängskärsklubb</td>
<td>3-5</td>
<td>116</td>
<td>10</td>
<td>99-09</td>
<td>3.9 (1.8-8.2)</td>
<td></td>
</tr>
<tr>
<td>Ängskärsklubb</td>
<td>2-5</td>
<td>70</td>
<td>8</td>
<td>02-09</td>
<td>21 (11-38)</td>
<td></td>
</tr>
<tr>
<td>spring</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Landsort</td>
<td>3-5</td>
<td>131</td>
<td>11</td>
<td>99-09</td>
<td>14 (9.6-19)</td>
<td></td>
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<tr>
<td>Utlångan</td>
<td>3-4</td>
<td>131</td>
<td>11</td>
<td>99-08</td>
<td>-8.3 (-17.33)</td>
<td>10 (6.0-17)</td>
</tr>
<tr>
<td>autumn</td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Utlångan</td>
<td>2-4</td>
<td>68</td>
<td>8</td>
<td>02-09</td>
<td>24 (14-41)</td>
<td></td>
</tr>
<tr>
<td>autumn</td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>125</td>
<td>11</td>
<td>99-09</td>
<td>-16 (-22,-9.7)*</td>
<td>2.4 (1.7-3.4)</td>
</tr>
<tr>
<td>Väderöarna</td>
<td>166</td>
<td>10</td>
<td></td>
<td>02-09</td>
<td>-15 (-22,-8.1)*</td>
<td>2.1 (1.3-3.2)</td>
</tr>
<tr>
<td>Cod liver</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SE Gotland</td>
<td>3-4</td>
<td>106</td>
<td>11</td>
<td>99-09</td>
<td>22 (12-41)</td>
<td></td>
</tr>
<tr>
<td>Fladen</td>
<td>2-3</td>
<td>70</td>
<td>8</td>
<td>99-09</td>
<td>-10 (-20,-29)*</td>
<td>5.1 (2.9-9.0)</td>
</tr>
<tr>
<td>Gilliemot egg</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>St. Karlsö</td>
<td>217</td>
<td>36</td>
<td>68-09</td>
<td>2.7 (1.7,3.7)*</td>
<td>170 (140-210)</td>
<td></td>
</tr>
</tbody>
</table>

* significant trend, p < 0.05
Polyaromatic hydrocarbons were retrospectively analysed in blue mussels from Kvädöfjärden in the Baltic, and Fladen and Väderöarna on the Swedish west coast, in time series from 1987 - 2003, 1985 - 2003 and 1984 - 2003, respectively. Since 2003, PAHs have been analysed on a yearly basis from these three blue mussel sites. Other species are not analysed as the extent to which PAHs metabolise in other species is not known.

28.1 Introduction

28.1.1 Uses, Production and Sources
PAH sources are either pyrolytic or petrogenic. They can be evaluated by molecule indexes and are based on concentration relationships between individual PAHs (Pikkarainen 2004).

28.1.2 Target Levels
The target levels used for PAHs in blue mussels are listed below in ug/kg dry weight. For further information on target levels and selection of target level see chapter 10.

Fluoranthene 110 ug/kg d.w.; Anthracene 290 ug/kg d.w.; Naphtalene 340 ug/kg d.w.; Phenantrene 1700 ug/kg d.w.; Pyrene 100 ug/kg d.w.; Benzo(a)antracene 80 ug/kg d.w.; benzo(a)pyrene 600 ug/kg d.w.; and Benzo(g,h,i)perylene 110 ug/kg d.w..

28.2 Methods

28.2.1 Analytical Information
The PAHs analysed are: naphthalen, acenaphtene, fluorene, phenantrene, antracene, fluoranthene, pyrene, benzo(a)antracene, chryene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)antracene, benzo(g,h,i)perylene and indeno(1,2,3-cd)pyrene. Metabolic capacity of the species sampled has to be considered.

See chapter 6, section 6.4 for further information regarding analytical methods for PAHs.
28.3 Results

28.3.1 Spatial Variation

Figure 28.1. Spatial variation in sPAH concentration (ng/g dry weight) in blue mussel soft body.

Blue mussel soft body from Kvädöfjärden in the Baltic Proper represent the highest sum of PAHs (sPAH) concentrations of the blue mussel samples (Fig. 28.1). Retrospective studies showed that the PAHs were not all systematically higher at Kvädöfjärden, for example fluoranthene and pyrene showed higher concentrations at Väderöarna.

28.3.2 Temporal Variation

All PAHs analysed (except acenaphthene, which was rarely found above the quantification limit) are presented as time series below (Fig. 28.2 – 28.16). Decreasing trends were found at some sites only e.g in the time series for sPAH at Väderöarna (Fig. 28.2); for chrysene at Fladen and Väderöarna (Fig. 28.9); for fluoranthene at Väderöarna (Fig. 28.12); for naphthalene at Kvädöfjärden and Väderöarna (Fig. 28.14); and for pyrene at Fladen and Väderöarna (Fig. 28.16). Increasing trends were observed for benzo(a)anthracene at Kvädöfjärden (Fig. 28.4), for fluorene at Kvädöfjärden, and for the last 10 years also at Väderöarna (Fig. 28.11).

The number of years required to detect an annual change of 10% in concentration varied a lot depending on the type of PAH and sampling site. Generally the statistical power to detect trends is low compared to other contaminants, and is between 14 - 24 years. Some PAHs (e.g. anthracene and fluoranthene (Fig. 28.3, 28.12) show extremely high
concentrations in certain years compared to the average concentrations. These results could possibly be outliers. The power and number of years required to detect a trend would improve if these outliers were excluded.
Figure 28.2. sPAH concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1986, 1983 and 1987 respectively).

Figure 28.3. Anthracene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1986, 1984 and 1987 respectively). The green area denotes the levels below the suggested target value for anthracene in blue mussels.
**Figure 28.4.** Benzo(a)anthracene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1989, 1984 and 1986 respectively). The green area denotes the levels below the suggested target value for benzo(a)anthracene in blue mussels.

**Figure 28.5.** Benzo(a)pyrene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively). The green area denotes the levels below the suggested target value for benzo(a)pyrene in blue mussels.
Figure 28.6. Benzo(b)fluoranthene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1986, 1984 and 1986 respectively).

Figure 28.7. Benzo(g, h, i)perylene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively). The green area denotes the levels below the suggested target value for benzo(g, h, i)perylene in blue mussels.
Benzo(k)fluoranthene, ng/g dry w., blue mussel

Vaderoarna

n(tot)=18, n(yrs)=17
m=6.19 (3.80, 10.1)
slope=-6.4% (-12, -.63)
SD(n)=100%, 53%, 23 yr
SD(sm)=40, N5, 29%
slope=6.3% (17, 29)
SD(lr)=88%, 37%, 19 yr
power=13, 16/30%
r2=0.09

Kvadofjarden

n(tot)=18, n(yrs)=18
m=8.94 (6.91, 11.6)
slope=-1.8% (-5.4, 1.9)
SD(n)=55%, 7.2%, 15 yr
power=98, 30/20%
y(09)=7.3 (4.5, 11.9)r2=0.11

Fladen

n(tot)=20, n(yrs)=17
m=4.66 (2.96, 7.33)
slope=4.9% (11, 81)
SD(n)=99%, 13%, 20 yr
power=58, 15/33%
y(09)=8.83 (1.38, 5.82)r2=0.18

Chrysene, ng/g dry w., blue mussel

Vaderoarna

n(tot)=18, n(yrs)=17
m=6.19 (3.80, 10.1)
slope=6.4% (17, 29)
SD(n)=100%, 43%, 20 yr
power=58, 14/34%
y(09)=3.25 (1.58, 6.69)r2=0.09
tao=37, p<0.001

Kvadofjarden

n(tot)=18, n(yrs)=18
m=8.94 (6.91, 11.6)
slope=1.8% (5.4, 1.9)
SD(n)=55%, 7.2%, 15 yr
power=98, 30/20%
y(09)=7.3 (4.5, 11.9)
r2=0.11

Fladen

n(tot)=20, n(yrs)=17
m=1.27 (0.81, 1.98)
slope=-2.0% (-8.1, 4.1)
SD(n)=141%, 14%, 21 yr
power=.53/.13/36%
y(09)=1.04 (0.48, 2.22)

Chrysene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively).

Figure 28.8.
Figure 28.10. Dibenzo(a,h)anthracene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively).

Figure 28.11. Fluorene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively).
### Fluoranthene, ng/g dry w., blue mussel

**Fladen**

- \( n(\text{tot})=20, n(\text{yrs})=17 \)
- \( m=6.93 (3.98, 12.1) \)
- Slope= 5.7\%  (13.1, 4)
- Power= 41.1\%  (8.95, 9.46)
- \( r^2=0.16, \text{NS} \)

**Väderöarna**

- \( n(\text{tot})=17, n(\text{yrs})=16 \)
- \( m=6.93 (16.7, 24.2) \)
- Slope= 5.5\%  (11.9, -9.6)
- Power= 72.2\%  (5.0, 18.3)
- \( r^2=0.32, \text{NS} \)

**Kvädöfjärden**

- \( n(\text{tot})=18, n(\text{yrs})=16 \)
- \( m=6.93 (8.95, 12.1) \)
- Slope= 7.5\%  (46.31)
- Power= 0.07 (0.09, 0.50)
- \( r^2=0.02, \text{NS} \)

**Figure 28.12.** Fluoranthene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively). The green area denotes the levels below the suggested target value for fluoranthene in blue mussels.

### Indeno(1,2,3-cd)pyrene, ng/g dry w., blue mussel

**Fladen**

- \( n(\text{tot})=16, n(\text{yrs})=15 \)
- \( m=1.29 (0.795, 2.08) \)
- Slope= -0.51\%  (7.5, 6.5)
- Power= 36.13\%  (48.3, 0.09)
- \( r^2=0.00, \text{NS} \)

**Väderöarna**

- \( n(\text{tot})=15, n(\text{yrs})=14 \)
- \( m=1.59 (1.02, 2.47) \)
- Slope= -0.01\%  (-6.3, 6.2)
- Power= 42.15\%  (69.3, 6.7)
- \( r^2=0.00, \text{NS} \)

**Kvädöfjärden**

- \( n(\text{tot})=18, n(\text{yrs})=18 \)
- \( m=7.93 (6.19, 10.2) \)
- Slope= -2.1\%  (-40, 36)
- Power= 42.15\%  (69.3, 6.7)
- \( r^2=0.00, \text{NS} \)

**Figure 28.13.** Indeno(1, 2, 3-cd)pyrene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively).
Naphthalene, ng/g dry w., blue mussel

Fladen

- n(tot)=20, n(yrs)=17
- m=2.65 (1.96,3.71)
- slope=-3.1% (-7.5,1.2)
- SD(lr)=69%, 9.6%, 17 yr
- power=.83/.22/24%

Väderöarna

- n(tot)=18, n(yrs)=17
- m=2.08 (1.19,3.62)
- slope=-6.4%(-13,.42)
- SD(lr)=129%,16%,23 yr
- power=.43/.12/41%

Kvädöfjärden

- n(tot)=18, n(yrs)=18
- m=10.4 (6.04,17.8)
- slope=-13%(-17,-8.5)
- SD(lr)=64%,8.2%,16 yr
- power=.93/.25/23%

Figure 28.14. Naphthalene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively). The green area denotes the levels below the suggested target value for naphthalene in blue mussels.

Phenanthrene, ng/g dry w., blue mussel

Fladen

- n(tot)=20, n(yrs)=17
- m=7.35 (4.49,12.0)
- slope=-4.1%(-11,2.3)
- SD(lr)=118%,15%,22 yr
- power=.48/.12/38%

Väderöarna

- n(tot)=18, n(yrs)=17
- m=7.19 (4.89,10.6)
- slope=-.87%(-6.2,4.4)
- SD(lr)=90%,12%,19 yr
- power=.65/.16/31%

Kvädöfjärden

- n(tot)=18, n(yrs)=18
- m=7.04 (4.82,10.3)
- slope=-1.2%(-6.7,4.3)
- SD(lr)=92%,11%,20 yr
- power=.72/.16/37%

Figure 28.15. Phenanthrene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively). The green area denotes the levels below the suggested target value for phenanthrene in blue mussels.
**Pyrene, ng/g dry w., blue mussel**

Figure 28.16. Pyrene concentration (ng/g dry weight) in blue mussels from Fladen, Väderöarna and Kvädöfjärden (time series starting in 1987, 1984 and 1986 respectively). The green area denotes the levels below the suggested target value for pyrene in blue mussels.

### 28.4 Conclusion

Only blue mussels have been examined for spatial differences in PAH concentrations; concentration of sPAH was found to be higher from Kvädöfjärden in the Baltic Proper. However, not all PAHs were systematically higher at Kvädöfjärden, for example fluoranthene and pyrene showed higher concentrations at Väderöarna.

Over time, acenaphtene was rarely found above the quantification limit.

The variation in the time series for PAHs is most often large with many extreme values, so one should interpret the trends with caution. Significant increasing trends were observed for sPAH, chrysene, fluoranthene, pyrene, naphthalene at Väderöarna, and naphthalene at Kvädöfjärden and chrysene, pyrene at Fladen. Significant increasing trends were seen for the whole time period, or in the last 10 years, for fluorene at Väderöarna, and benzo(a)anthracene and fluorene at Kvädöfjärden.

All time series where concentration of various PAHs were compared with the target value based on OSPAR EAC (Ecological Assessment Criteria), were below the target value.
PFOS was retrospectively analysed in guillemot eggs from St. Karlsö in a time series starting from 1968. Additionally, a selection of perfluorinated substances (see 29.2.1) were analysed in herring liver tissue over the last five years.

29.1 Introduction

29.1.1 Uses, Production and Sources

Perfluorinated substances have been used industrially and commercially since the beginning of the 1950s. However it was not until 2000 that the main producer, 3M, started to phase out their production of the main compound of concern, perfluorooctane sulfonate (PFOS) and PFOS-related chemicals (Key et al. 1997; Holmström et al. 2005).

PFCAs (perfluorinated carboxylates) in the environment can have two sources - direct sources from manufacturing and use of PFCAs, and indirect sources from degradation of volatile precursor compounds (Prevedouros et al. 2006). PFNA (perfluorononanoate) is intentionally produced and therefore probably originates mainly from direct sources (production and use of consumer products containing PFNA, such as PTFE products), and waterborne transport to remote locations. Therefore, sewage treatment plant effluent from industry or larger cities could represent hot-spots. In contrast, PFUnA (perfluoroundecanoate) and PFTrA (perfluorotridecanoate) are unintentionally produced substances, and their presence in the environment is probably due to both direct sources (impurities in PFOA (perfluorooctanoate) and PFNA productions) and indirect sources (atmospheric transport and degradation of precursors).

29.1.2 Conventions, aims and restrictions

Perfluorooctane sulfonic acid, its salts, and perfluorooctane sulfonyl fluoride are among the nine new Persistent Organic Pollutants (POPs) included in The Stockholm Convention on POPs, an international agreement requiring measures for reducing or preventing release of dangerous substances to the environment. The Stockholm Convention was adopted in 2001 and entered into force in 2004. The nine new POPs were adopted in 2009 and the amendments entered into force in 2010.

29.1.3 Target Levels

The target levels used for PFOS in herring liver is 9.1 ug/kg wet weight. For further information on target levels and selection of target level see chapter 10.
29.2 Methods

29.2.1 Analytical Information

Comprised PFCAs analysed included: perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), perfluoroctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDoA), perfluoroundecanoate (PFUnA), perfluorododecanoate (PFPeDA), perfluorotridecanoate (PFTriA), perfluorotetradecanoate (PFTeA), perfluoropentadecanoate (PFPeDA) as well as perfluorinated sulfonates (PFSs): perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS), perfluorodecane sulfonate (PFDoS), perfluorooctane sulfonamide (PFOSA) and 6:2 fluorotelomer sulfonate (6:2 FTS).

See chapter 6, section 6.5 for further details regarding analytical methods for PFCs.

29.3 Results

29.3.1 Spatial Variation

So far analysis of herring liver from only five years (2005 - 09) (pooled samples, 12 fish in each) from the old sampling sites, and three years from the new sampling sites (two years at the offshore sites) are available. Therefore, the results should be treated with caution. However, it has been shown that the individual variation of perfluorinated substances is relatively small compared to classical POPs (Verreault et al. 2007). The spatial variation of seven perfluorinated substances (three PFSs in figure 29.1: PFHxS, PFOS and PFOSA; and four PFCAs in figure 29.2: PFNA, PFDoA, PFUnA and PFTriA) are presented below. The selection was based on a number of results above LOQ.
Figure 29.1 Spatial variation in concentration (wet weight) of A) PFHxS, B) PFOS and C) PFOSA in herring liver. Highest concentration of PFHxS and PFOS were 1.3 ng/g and 18.7 ng/g, respectively at Lagnö in the north Baltic Proper. Highest PFOSA concentration (7.1 ng/g) was found at Fladen in the Kattegatt.
A) PFNA

B) PFDcA

C) PFUnA

D) PFTriA

**Figure 29.2** Spatial variation in PFCA concentrations (wet weight) of A) PFNA, B) PFDcA, C) PFUnA and D) PFTriA in herring liver. Highest concentrations of PFNA (2.9 ng/g)) were found at Utlängan in the south Baltic Proper. Highest concentrations of PFUnA (3.0 ng/g), PFDcA (2.1 ng/g and PFTriA (3.3 ng/g) were found at Rånefjärden in the Bothnian Bay.

### 29.3.2 Temporal Variation

A significant increasing trend of 8% per year was observed for PFOS in guillemot eggs (Fig. 29.3), which is equivalent to an increase 25 - 30 times higher in the early 2000s compared to the late 1960s.

**The time series for PFCs in herring liver are based on only five years of sampling and many of the values are below the Level of Quantification (LOQ), thus it is too early to interpret the time trends.**

Most of the time series in herring liver for the PFCs show no significant decreases or increases, and it is too early to interpret those that do show a trend due to the short period of monitoring and the fact that some of the yearly concentrations are based on values below LOQ (Fig.29.4 – 29.12).
**Figure 29.3** Temporal trend of PFOS concentrations in guillemot eggs (ng/g wet weight) (time series starting in 1968). The mean annual PFOS value shown as red dots in the figure is based on pooled samples or mean values of individual samples.

**PFOS, ng/g wet w., herring liver**

<table>
<thead>
<tr>
<th>Location</th>
<th>n(tot)</th>
<th>n(yrs)</th>
<th>m</th>
<th>slope</th>
<th>SD(in)</th>
<th>SD(sm)</th>
<th>SD(lr)</th>
<th>power</th>
<th>y(09)</th>
<th>r2</th>
<th>tao</th>
<th>SD(sm)</th>
<th>p &lt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harufjarden</td>
<td>28</td>
<td>24</td>
<td>11.0 (7.9, 15.1)</td>
<td>15%</td>
<td>78,300</td>
<td>16%</td>
<td>23%</td>
<td>7%</td>
<td>48%</td>
<td>5.0 %</td>
<td>26</td>
<td>0.6</td>
<td>33</td>
</tr>
<tr>
<td>Angskarsklubb</td>
<td>24</td>
<td>20</td>
<td>8.4 (3.9, 11.7)</td>
<td>14%</td>
<td>52,800</td>
<td>11%</td>
<td>8%</td>
<td>7%</td>
<td>45%</td>
<td>12.6</td>
<td>6.8,23%</td>
<td>22</td>
<td>0.6</td>
</tr>
<tr>
<td>Landsort</td>
<td>20</td>
<td>16</td>
<td>10.6 (7.2, 14.6)</td>
<td>17%</td>
<td>14,870</td>
<td>11%</td>
<td>1%</td>
<td>7%</td>
<td>45%</td>
<td>12.6</td>
<td>6.8,23%</td>
<td>22</td>
<td>0.6</td>
</tr>
<tr>
<td>Ulltangan</td>
<td>24</td>
<td>20</td>
<td>8.1 (8.1, 19.1)</td>
<td>21%</td>
<td>19,380</td>
<td>13%</td>
<td>9%</td>
<td>7%</td>
<td>45%</td>
<td>12.6</td>
<td>6.8,23%</td>
<td>22</td>
<td>0.6</td>
</tr>
<tr>
<td>Fladen</td>
<td>24</td>
<td>20</td>
<td>6.9 (5.5, 8.3)</td>
<td>30%</td>
<td>12,550</td>
<td>12%</td>
<td>6%</td>
<td>7%</td>
<td>45%</td>
<td>12.6</td>
<td>6.8,23%</td>
<td>22</td>
<td>0.6</td>
</tr>
<tr>
<td>Vaderoarna</td>
<td>24</td>
<td>20</td>
<td>3.8 (2.4, 6.1)</td>
<td>38%</td>
<td>9,600</td>
<td>13%</td>
<td>1%</td>
<td>7%</td>
<td>45%</td>
<td>12.6</td>
<td>6.8,23%</td>
<td>22</td>
<td>0.6</td>
</tr>
</tbody>
</table>

**Figure 29.4.** Temporal trend of PFOS in herring liver (ng/g wet weight) (time series starting in 2005). The green area denotes the levels below the suggested target value for PFOS in fish.
PFOSA concentration in herring liver shows a decreasing trend at three sites, but is inconsistent at the other three sites (Fig. 29.5); this is the same as for PFHpA concentration (Fig. 29.6). By contrast, an increasing trend is seen at Harufjärden for PFHxS (Fig. 29.7), and at Ångskärrsklubb, Landsort and Utlängan for PFOA (Fig. 29.8).

**Figure 29.5.** Temporal trend of PFOSA in herring liver (ng/g wet weight) (time series starting in 2005).

**Figure 29.6.** Temporal trend of PFHpA concentrations in herring liver (ng/g wet weight) (time series starting in 2005).
### PFHxS, ng/g wet w., herring liver

<table>
<thead>
<tr>
<th>Location</th>
<th>n(tot)</th>
<th>n(yrs)</th>
<th>m (ng/g)</th>
<th>SD (ng/g)</th>
<th>SD (%)</th>
<th>Slope (%)</th>
<th>Power (%)</th>
<th>y(09) (ng/g)</th>
<th>r²</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harufjarden</td>
<td>8</td>
<td>5</td>
<td>0.145</td>
<td>0.083</td>
<td>16</td>
<td>15</td>
<td>16</td>
<td>0.234</td>
<td>0.71</td>
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</tr>
<tr>
<td>Angskarsklubb</td>
<td>5</td>
<td>5</td>
<td>0.171</td>
<td>0.152</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>0.34</td>
<td>0.55</td>
<td>&lt;0.050</td>
</tr>
<tr>
<td>Landsort</td>
<td>8</td>
<td>5</td>
<td>0.320</td>
<td>0.152</td>
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<td>33</td>
<td>11</td>
<td>0.51</td>
<td>0.36</td>
<td>NS</td>
</tr>
<tr>
<td>Utlangan</td>
<td>8</td>
<td>5</td>
<td>0.294</td>
<td>0.146</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>0.44</td>
<td>0.32</td>
<td>NS</td>
</tr>
<tr>
<td>Fladen</td>
<td>8</td>
<td>5</td>
<td>0.206</td>
<td>0.135</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>0.284</td>
<td>0.55</td>
<td>&lt;0.050</td>
</tr>
<tr>
<td>Vaderoarna</td>
<td>8</td>
<td>5</td>
<td>0.121</td>
<td>0.083</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>0.138</td>
<td>0.15</td>
<td>NS</td>
</tr>
</tbody>
</table>

### PFOA, ng/g wet w., herring liver

<table>
<thead>
<tr>
<th>Location</th>
<th>n(tot)</th>
<th>n(yrs)</th>
<th>m (ng/g)</th>
<th>SD (ng/g)</th>
<th>SD (%)</th>
<th>Slope (%)</th>
<th>Power (%)</th>
<th>y(09) (ng/g)</th>
<th>r²</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Angskarsklubb</td>
<td>5</td>
<td>4</td>
<td>0.409</td>
<td>0.248</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>0.56</td>
<td>0.55</td>
<td>&lt;0.050</td>
</tr>
<tr>
<td>Landsort</td>
<td>8</td>
<td>5</td>
<td>0.584</td>
<td>0.164</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>1.59</td>
<td>0.91</td>
<td>&lt;0.050</td>
</tr>
<tr>
<td>Utlangan</td>
<td>8</td>
<td>5</td>
<td>0.903</td>
<td>0.314</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>2.20</td>
<td>0.60</td>
<td>&lt;0.050</td>
</tr>
<tr>
<td>Fladen</td>
<td>8</td>
<td>5</td>
<td>1.000</td>
<td>0.311</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>2.70</td>
<td>0.47</td>
<td>&lt;0.050</td>
</tr>
<tr>
<td>Vaderoarna</td>
<td>8</td>
<td>5</td>
<td>0.424</td>
<td>0.300</td>
<td>15</td>
<td>33</td>
<td>11</td>
<td>0.47</td>
<td>0.37</td>
<td>&lt;0.050</td>
</tr>
</tbody>
</table>

---

**Figure 29.7.** Temporal trend of PFHxS in herring liver (ng/g wet weight) (time series starting in 2005).

**Figure 29.8.** Temporal trend of PFOA in herring liver (ng/g wet weight) (time series starting in 2005).

All other compounds examined (PFDcA, PFNA, PFTriA and PFUnA) show inconsistent trends over the examined time series (Fig. 29. 9 – 12).
Figure 29.9. Temporal trend of PFDcA concentrations in herring liver (ng/g wet weight) (time series starting in 2005).

Figure 29.10. Temporal trend of PFNA in herring liver (ng/g wet weight) (time series starting in 2005).
Figure 29.11. Temporal trend of PFTriA in herring liver (ng/g wet weight) (time series starting in 2005).

Figure 29.12. Temporal trend of PFUnA in herring liver (ng/g wet weight) (time series starting in 2005).
29.4 Conclusion

The time series of PFOS concentration in herring that were compared with the target value based on food uptake by human via fish products of 9.1 ng/g wet weight, were all close to the target level. At Harufjärden, Landsort and Utlängan, the majority of the measured concentrations were above the suggested target level.

PFHxS and PFOS show a similar spatial pattern, but PFOS concentrations were approximately 45 times higher than PFHxS levels. This was expected, since PFHxS has not been produced intentionally, but only occurs as a by-product of technical PFOS.

Furthermore, the distribution of PFOS is quite homogenous along the Swedish coast (with the exception of Lagnö and the offshore sampling site in the Baltic Proper), which is a result of the extraordinary persistency of the compound and the long history of use (three decades). Elevated levels may be expected at sites with a higher population density and associated current emissions from consumer products still containing technical PFOS. PFOSA, however, is not persistent, but a precursor compound to PFOS. The high concentrations on the west coast reflect a current source probably located around the North Sea. The relatively short environmental half-life of PFOSA does not allow it to diffuse into the Baltic, due to the low water exchange between the two seas. Degradation of PFOSA to PFOS might also contribute to higher PFOS concentrations. Taking into account that liver generally contains about five times higher concentrations than fish muscle, PFOS levels in herring liver are compatible with levels found in other fish species from the Baltic (Swedish Environmental Protection Agency, 2007). PFOS concentrations in guillemot eggs from 2005, however, are about 200 times higher than in herring liver (herring and sprat being the main prey of guillemot), showing the high retention of this compound in guillemot and the transport potential to the forming egg.

PFCAs in the environment can have two sources - direct sources from manufacturing and use of PFCAs, and indirect sources from degradation of volatile precursor compounds (Prevedouros et al. 2006). PFNA is intentionally produced and therefore probably originates mainly from direct sources (production and use of consumer products containing PFNA, such as PTFE products), and waterborne transport to remote locations. This may partly explain the spatial variations of PFNA in this study, as sewage treatment plant effluent from industry or larger cities could represent hot-spots. In contrast, PFUnA and PFTriA are unintentionally produced substances, and their presence in the environment is probably due to both direct sources (impurities in PFOA and PFNA productions) and indirect sources (atmospheric transport and degradation of precursors). The fact that the odd-chained PFUnA and PFTriA are more highly concentrated than PFDoA, and the homogenous spatial distribution of these compounds, supports the theory that indirect sources are important for these long-chain PFCAs. Also levels and compound patterns of PFCAs are in good agreement with concentrations in other Baltic fish (Swedish Environmental Protection Agency, 2007).
A consistently increasing trend in PFOS in guillemot eggs has been observed throughout the whole examined time period, although this trend has increased as a faster rate since the mid 1990s compared to earlier years. Due to a change in the analytical method between 2003 - 2004 and relatively high inter-annual variations, the future trend for temporal PFOS concentrations in the Baltic marine environment cannot be predicted. Further monitoring will reveal if the phase out by 3M will make a difference for the PFOS concentrations in biota.
30 References


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