**The Swedish National Monitoring Programme for Contaminants in Marine Biota (until 2018 year's data)**

- **Temporal trends and spatial variations**

Det svenska nationella miljöövervakningsprogrammet för miljögifter i biota (fram till 2018 års data)

- Temporal och spatial variation

Sara Danielsson, Suzanne Faxneld, Anne L. Soerensen

Överenskommelse: 2213-16-003

**2020-02-19**

**Preparation of samples and biological parameters:**
**Swedish Museum of Natural History**
**Henrik Dahlgren, Eva Kylberg, Jill Staveley Öhlund, Per-Arvid Berglund**

**Computational support (figures, tables and appendix):**
**Swedish Museum of Natural History**
**Martin Sköld**

**Chemical analysis and description of the analytical methodology of the specific compound:**

**Chlorinated pesticides, polychlorinated biphenyls and brominated flame retardants:**
**Department of Environmental Science and Analytical Chemistry, Stockholm University**
**Project leader: Cynthia de Wit**
**Chemists: Ulla Eriksson, Anna-Lena Egebäck, Martin Kruså**

**Perflourinated substances:**
**Department of Environmental Science and Analytical Chemistry, Stockholm University**
**Project leader: Jon Benskin**
**Chemists: Merle Plassman, Oskar Sandblom, Nathan Charlton**

**Trace metals:**
**Department of Environmental Science and Analytical Chemistry, Stockholm University**
**Project leader: Marcus Sundbom**
**Chemists: Pär Hjelmquist, Ann-Marie Johansson, Frida Edberg, Anna Hägglund**

**Stabile isotopes:**
**Department of Environmental Science and Analytical Chemistry, Stockholm University**
**Project leader: Marcus Sundbom**
**Chemists: Anna Hägglund, Monika Gineityte**
**Analyses by: SIF UC Davis, California, USA**

**PCDD/PCDF and dl-PCBs:**
**Department of Chemistry, Umeå University**
**Project leader: Peter Haglund**
**Chemist: Peter Haglund**

**PAHs:**
**IVL Swedish Environmental Research Institute**
**Project leader: Erika Rehngren**
**Chemists: Erika Rehngren, Jenny Friedrichen**

**Organotin compounds:**
**IVL Swedish Environmental Research Institute**
**Project leader: Elin Paulsson**
**Chemists: Raed Awad**

# The Swedish National Monitoring Programme for Contaminants in marine biota (until 2018 year's data) – Temporal trends and spatial variations

**Report authors**
Sara Danielsson, Suzanne Faxneld, Anne Sørensen

The Department of Environmental Research and Monitoring, Swedish Museum of Natural History

**Report title and subtitle**
Det svenska nationella miljöövervakningsprogrammet för miljögifter i marin biota (fram till 2018 års data)
- Temporal- och spatial variation

The Swedish National Monitoring Programme for Contaminants in marine biota (until 2018 year's data)
– Temporal trends and spatial variations

**Keywords for location (specify in Swedish)**
Östersjön, Västkusten, Bottenviken, Bottenhavet, Egentliga Östersjön, Skagerrak, Kattegatt, Rånefjärden, Harufjärden, Kinnbäcksfjärden, Holmöarna, Örefjärden, Gaviksfjärden, Långvindsfjärden, Ängskärsklubb, Lagnö, Landsort, Kvädöfjärden, Byxelkrok, St.Karlsö, SE Gotland, Utlängan, Hanöbukten, Abbekås, Kullen, Fladen, Nidingen, Väderöarna, Fjällbacka, Tjärnö, Ålands hav, Bornholmsbassängen, Bottenviken, Bottenhavet, Egentliga Östersjön, Västkusten

**Keywords for subject (specify in Swedish)**
Miljögifter, tidstrender, spatiala trender, aggregerade bassängtrender, DDT, PCB, HCH, HCB, dioxiner, furaner, metaller, Pb, Cd, Cu, Zn, Cr, Ni, Ag, As, Se, PBDE, HBCDD, PFAS, PFOS, PAH, tennorganiska föreningar, biota, fisk, strömming, sill, abborre, tånglake, torsk, blåmussla, sillgrissla, strandskata, fisktärna, ägg

**Period in which underlying data were collected**
1968–2018

Summary
The report summarises the monitoring activities within the National Swedish Contaminant Programme in marine biota.
Few trends are found for the biological variables of the biota included in the programme (age, weight, length, and fulton´s condition factor). This is expected, as samples are selectively selected to avoid trends. However, all variables for Holmöarna in the Bothnian Bay and the fish age at the West Coast sites (Kullen, Fladen and Väderöarna) stands out for herring, cod and perch as having upward trends over the last 10 years. Since many of the contaminants presented in this report bioaccumulate, this is likely to affect the trends presented below.
For the aggregated herring data (on sub-basin scale) on chlorinated pesticides, PCBs, dioxins and furans, brominated flame retardants and perfluorinated substances (PFAS) a general downward trend for the last 10 years (2009-2018) is seen for all sub-basins (Southern and Northern Baltic Proper and Bothnian Sea and Bay) except the West Coast. On the West Coast, changes are small and often non-significant. The contaminant concentrations are in general lowest on the West Coast but the concentration difference between the West Coast and the other sub-basins has shrunk over the last 10 years as the concentrations in the other sub-basins are decreasing towards West Coast levels. The perfluorinated compound FOSA is an exception to this picture, with 2-4 times higher concentrations on the West Coast compared to the Baltic Sea. For the metals, no common patterns are seen for the aggregated herring data across the metals or for each specific metal between sub-basins.

# Contents

# 1 Summary

This report presents a summary of the results within the Swedish National Monitoring Programme for Contaminants in marine biota until 2018 with focus on the last ten years.

Few trends are found for the biological variables of the biota included in the programme (age, weight, length, and Fulton´s condition factor). This is expected, as samples are selectively selected to avoid trends. However, all variables for Holmöarna in the Bothnian Sea and the fish age at the West Coast sites (Kullen, Fladen and Väderöarna) stands out for herring, cod and perch as having upward trends over the last 10 years. Since many of the contaminants presented in this report bioaccumulate, this is likely to affect the trends presented below.

For the aggregated herring data (on basin scale) on chlorinated pesticides, PCBs, dioxins and furans, brominated flame retardants and perfluorinated substances (PFAS) a general downward trend for the last 10 years (2009-2018) is seen for all basins (Southern and Northern Baltic Proper and Bothnian Sea and Bay) except the West Coast. On the West Coast, changes are small and often non-significant. The contaminant concentrations are in general lowest on the West Coast but the concentration difference between the West Coast and the other basins has shrunken over the last 10 years as the concentrations in the other basins are decreasing towards West Coast levels. The perfluorinated compound FOSA is an exception to this picture, with 2-4 times higher concentrations on the West Coast compared to the Baltic Sea. For the metals, no common patterns are seen for the aggregated herring data across the metals or for each specific metal between basins.

When expanding to look across all species (fish, mussel, and bird eggs) and sites there are no clear trends for any of the metals during the last ten years. For example, Cd shows downward trends in mussels and bird eggs and for Pb, concentrations are decreasing in mussels and bird eggs and in fish from some sites. For the organic contaminants, the chlorinated pesticides and PCB are in general decreasing in all biota with exceptions for HCB, where upward trends are seen at some sites. While dioxins and furans are in general decreasing in all biota except herring on the West Coast, concentrations in Eurasian oystercatcher are increasing significantly across almost all compounds. For brominated flame retardants concentrations are generally decreasing, but herring from Holmöarna and Ängskärsklubb in the Bothnian Sea show upward trends and a few herring sites at the West Coast show increasing concentrations as well as blue mussels on the West Coast. PFAS generally shows downward- or no trends. At the same time there are indications of increasing concentrations of FOSA in herring on the West Coast. Also PFOA, PFNA and br-PFOS indicates increasing concentrations in herring in the Bothnian Sea and the Baltic Proper. No general pattern is seen for PAH. All analysed PAHs show upward trends in blue mussels at Fjällbacka from the West Coast during the most recent ten years, these trends are mainly due to elevated concentrations in 2016-2018. Organotin compounds are mostly below LOQ all years but concentrations in 2018 were higher for some of the organotin compounds compared to all previous years.

# 2 Sammanfattning

Denna rapport presenterar en sammanfattning av resultaten från det nationella övervakningsprogrammet för miljögifter i marin biota fram till år 2018 med focus på de senaste tio åren.

Få trender ses för de biologiska variablerna i biota som registreras inom övervakningsprogrammet (ålder, vikt, längd och konditionsfaktor). Detta förväntas, eftersom individer selektivt väljs för att vara så homogena som möjligt (mellan och inom år) för att undvika trender. Alla biologiska variabler för Holmöarna i Bottenhavet och ålder på fisk från västkusten (Kullen, Fladen och Väderöarna) visar dock uppåtgående trender under de senaste tio åren i sill/strömming, torsk och abborre. Eftersom många av de miljögifter som presenteras i denna rapport bioackumuleras, kan detta sannolikt påverka trenderna som presenteras nedan. För aggregerade sill-/strömmingdata (på bassängnivå) ses generellt nedåtgående trender under de senaste tio åren (2009-2018) för klorerade pesticider, PCB, dioxiner och furaner, bromerade flamskyddsmedel och perfluorerade ämnen (PFAS) för alla bassänger (Bottenviken, Bottenhavet, norra och södra Egentliga Östersjön) utom västkusten. På västkusten är förändringarna små och ofta icke-signifikanta. Koncentrationerna av miljögifter är i allmänhet lägst på västkusten men koncentrationsskillnaden mellan västkusten och de andra bassängerna har krympt under de senaste tio åren eftersom koncentrationerna i de andra bassängerna minskar i större utsträckning. Det perfluorerade ämnet FOSA är ett undantag från denna bild, där 2-4 gånger högre koncentrationer ses på västkusten jämfört med Östersjön. För metallerna ses inga generella mönster för de aggregerade sill/strömming, det ses heller inga mönster för varje specifik metall mellan bassänger.

Tittar man istället på alla arter (fisk, mussla och fågelägg) och stationer finns det inga tydliga trender för någon av metallerna under de senaste tio åren. Till exempel visar Cd nedåtgående trender för musslor och fågelägg och Pb-koncentrationen minskar i musslor och fågelägg samt i fisk från vissa platser. För de organiska miljögifterna minskar de klorerade pesticiderna och PCB i biota med undantag för HCB, där uppåtgående trender ses på vissa stationer. Dioxiner och furaner minskar generellt i biota med undantag för Holmöarna (Bottenhavet) och Väderöarna (västkusten), det ses även ökande koncentrationerna av i princip alla kongener i strandskata från västkusten. Bromerade flamskyddsmedel minskar generellt i biota, men strömming från Holmöarna och Ängskärsklubb (Bottenhavet) visar uppåtgående trender, även sill och blåmussla från västkusten visar ökande koncentrationer. PFAS visar generellt nedåtgående eller inga trender. Samtidigt finns det indikationer på ökande koncentrationer av FOSA i sill på västkusten. Ökande koncentrationer indikeras även för PFOA, PFNA och br-PFOS i strömming från Bottenhavet och Egentliga Östersjön. För PAH'er ses inga generella mönster. Däremot visar alla PAH'er i blåmussla från Fjällbacka på västkusten uppåtgående trender under de senaste tio åren, dessa trender beror främst på förhöjda koncentrationer 2016–2018. Tennorganiska föreningar har oftast legat under LOQ för många av de undersökta åren men koncentrationerna under 2018 var högre för flera av de tennorganiska föreningarna jämfört med alla tidigare år.

# 3  Introduction

This report presents a summary of the monitoring activities within the Swedish National Monitoring Programme for Contaminants in marine biota until 2018. An updated report is published each year and the text in the Sampling and Analytical methods section is for a large part reused from previous years.  The report is the result of joint efforts from several institutes and agencies. The *Department of Environmental Research and Monitoring* at the Swedish Museum of Natural History is responsible for coordinating the monitoring programme, which includes administration, sample collection, sample preparation, recording of biological variables, storage of frozen biological tissues in the Environmental Specimen Bank, data compilation and preparation, statistical evaluation and report writing. The *Department of Environmental Science and Analytical Chemistry (ACES)* at Stockholm University is responsible for the analyses of metals, organochlorines, brominated flame retardants and per- and polyfluoroalkyl substances; the *Department of Chemistry* at Umeå University for the analyses of dioxin-like polychlorinated biphenyls, polychlorinated dibenzo-*p*-dioxins and -furans; the *Swedish Environmental Research Institute (IVL)* for the analyses of polycyclic aromatic hydrocarbons and organotin compounds. The monitoring programme is financed by the *Swedish Environmental Protection Agency* (SEPA).

The contaminant concentrations in the biological samples presented in this report represent the bioavailable portion i.e. the portion that has effectively passed through biological membranes and may cause toxic effects. The objectives of the Swedish National Monitoring Programme for Contaminants 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 of contamination, along the Swedish coast.

- To describe the general contaminant load and supply reference values for regional and local monitoring programmes.

- To monitor long-term time trends and 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 character and to monitor a potential increase from secondary sources of banned contaminants.

   *quantified objective:* to detect an increase of 100 % (a doubling) from one year to the other, with a power of 80 % at a 5 % significance level.

- To describe spatial variability within the Baltic Sea.

   *quantified objective:* to detect differences of a factor of 2 between sites, with a power of 80 % at a 5 % significance level.

- To explore historical and regional differences in the composition and pattern of e.g. PCB, HCH, DDT, PCDD/F, PBDE, HBCDD, PAH, OTC and PFAS as well as the ratios between these contaminants.

As part of the monitoring programme whole individuals or sub-sampled material from all sites are frozen together with the large number of additional samples in the *Environmental Specimen Bank* for later use. This enables retrospective studies of compounds that are

currently unknown or for which it is currently not possible to analyse, or to redo analysis in cases of suspected analytical errors in old analysis.

In addition to the above-mentioned objectives, the monitoring programme is also a valuable resource for research, regulation processes and other marine monitoring activities as it:

- Provides high quality data that can be used as the basis for research studies (data analysis or model development and validation) that seek to explore the fate and distribution of contaminants in marine ecosystems in general, and in the Baltic and North Sea environment in particular.

- Monitors concentrations of contaminants in important fish species like herring and cod that are highly relevant for human consumption. The Swedish Museum of Natural History collaborates with the Swedish Food Administration and the Swedish Radiation Safety Authority on the analysis of radionuclides in fish and blue mussels.

- Records biological variables such as Fulton's condition factor (K), the liver somatic index (LSI) and the fat content at all sites over time. At a few sites, an integrated monitoring exists where, in addition to the contaminant concentrations, also the fish physiology and population abundance are monitored in cooperation with the University of Gothenburg, Department of Biological and Environmental Sciences, and the Swedish University of Agricultural Sciences, Department of Aquatic Resources (SLU AQUA).

- Offers a reference work for the design of regional and local monitoring programmes with more than 40 years of experience.

- Is an integrated part of the national monitoring activities in the marine environment, as well as of the international programmes within ICES, OSPAR, HELCOM and the EU.

- Provides a basis for two of Sweden's 16 environmental objectives:
    1. A non-toxic environment
    2. A balanced marine environment, flourishing coastal areas & archipelagos

The report focuses on time series of analysed contaminants in biota during the most recent ten-year period and summarises the results from statistical analyses. Comments are also given on spatial variation and on contaminant levels in relation to existing thresholds. It should be stressed that temporal and geographical differences may not only reflect anthropogenic influences, but can also be affected by factors such as productivity, temperature, salinity etc. On occasions, notes on 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.

A selection of figures visualising mean concentrations of the contaminants geographically, time trend figures for the whole monitoring period and tables containing statistical summaries of the time trends are presented in the report Appendix for each compound class separately.

# 4   Sampling and analytical methods

## 4.1   Sampling area

Within the Swedish National Monitoring Programme for Contaminants in marine biota, specimens are sampled along the Swedish coastline from the Bothnian Bay on the east coast to the North Sea on the west coast. The locations and names of the sampling sites are shown in Figure 1. The sampling sites are regarded as locally uncontaminated areas, i.e. as far as possible located away from and uninfluenced by major river outlets or ferry routes and not in a close vicinity to densely populated areas or other known local emission sources. The Swedish sampling sites in the Baltic Sea are included in the Convention on the Protection of the Marine Environment of the Baltic Sea Area (HELCOM), and the sampling sites in the North Sea are included in the Oslo and Paris Commissions' Joint Assessment and Monitoring Programme (OSPAR, JAMP).



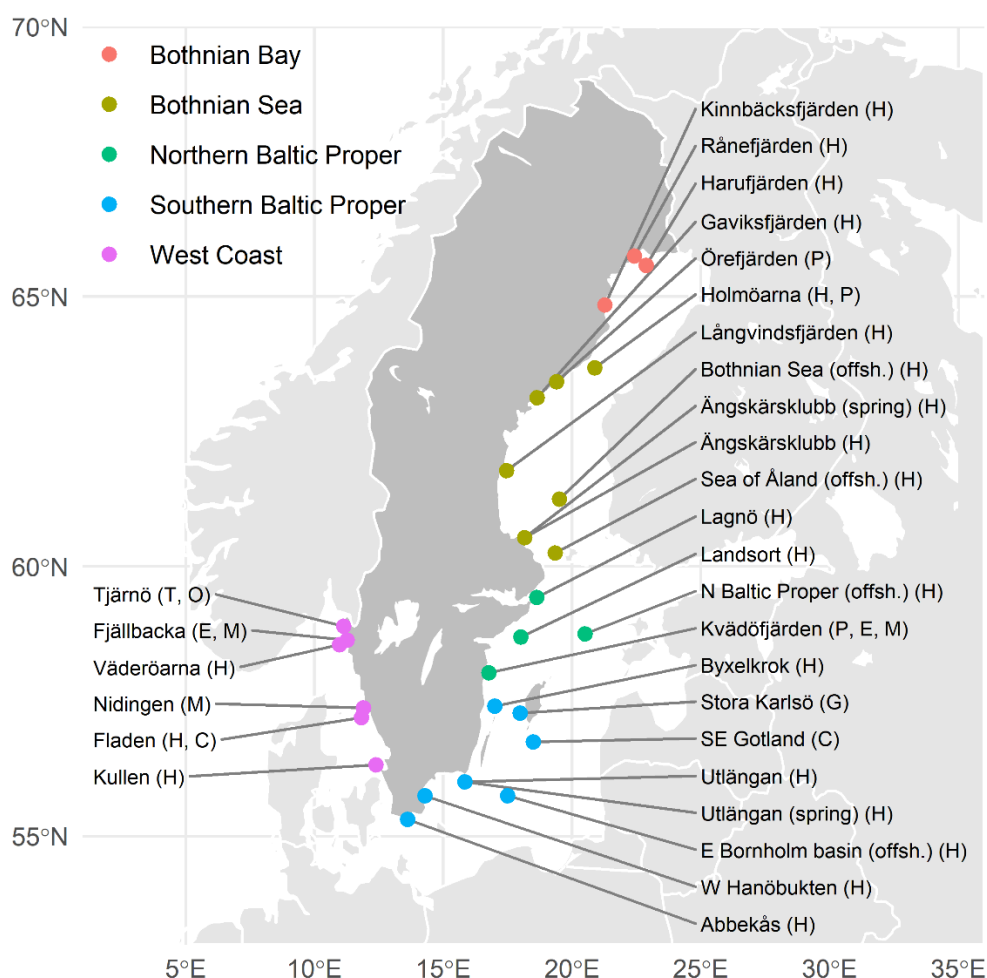**Figure 1.** Sampling sites within the Swedish National Monitoring Programme for Contaminants in marine biota (H=Herring, P=Perch, E=Eelpout, M=Blue mussel, G=Common guillemot, C=Cod, T=Common tern, O=Eurasian oystercatcher). The sites are divided into five larger areas, from here on referred to as basins. Data aggregated at a basin level is used for some of the statistical analysis (see section 5.2.2).

## 4.2   Collected specimen

Specimens analysed within the programme are fish muscle and/or liver samples of the species herring (*Clupea harengus*), cod (*Gadus morhua*), perch (*Perca fluviatilis*) and eelpout (*Zoarces viviparous*) as well as soft tissue samples of blue mussel (*Mytilus edulis*) and egg homogenate samples of the species common guillemot (*Uria aalge*), common tern (*Sterna hirundo*) and Eurasian oystercatcher (*Haematopus ostralegus*). Previously, dab (*Limanda limanda*) and flounder (*Platichtys flesus*) samples were analysed as well, however, these two species are currently only caught for storage in the Environmental Specimen Bank.

### 4.2.1   Herring (*Clupea harengus*)

Herring is a pelagic species that feeds mainly on zooplankton.  It becomes sexually mature at about 2–3 years of age in the Baltic Sea, and 3–4 years of age on the Swedish West Coast. It is an important prey for several predators in the marine environment and the most dominant commercial fish species in the Baltic. Due to its fat muscle tissue, this matrix is very suitable for analysis of fat-soluble contaminants e.g. hydrocarbons. Herring is the most commonly used indicator species for monitoring of contaminants in biota within the BMP (Baltic Monitoring Programme) in the HELCOM convention area, and is sampled by several countries: Finland, Estonia, Lithuania, Germany, Poland and Sweden.

### 4.2.2   Cod (*Gadus morhua*)

The Baltic cod lives below the halocline and feed on bottom organisms. In Swedish waters, it becomes sexually mature between 2–6 years old. Spawning takes place during May–August (occasional spawning specimens are 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 large fluctuations and decreased six-fold (by weight) between 1984 and 1993 [*Cardinale and Modin*, 1999]. Cod fishing for human consumption is economically important within the Baltic Sea and North Sea. Cod is among the 'first choice species' recommended within the JAMP (Joint Assessment and Monitoring Programme) and BMP. The cod liver is fat and therefore often store relatively high concentrations of organic contaminants. For that reason, it is a very suitable matrix for screening for 'new' contaminants.

### 4.2.3   Perch (*Perca fluviatilis*)

Perch is an omnivorous, opportunistic feeding predatory fish. Male perch become sexually mature between 2–4 years of age and females between 3–6 years of age. Spawning takes place during April–June when the water temperature reaches about 7–8 degrees Celsius. 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 SLU AQUA. Perch is also used as an indicator species for contaminant monitoring within the national monitoring programme of contaminants in freshwater biota.

### 4.2.4   Eelpout, viviparous blenny (*Zoarces viviparus*)

Eelpout is considered to be a relatively stationary species living close to the sea bottom, feeding on insect larvae, molluscs, crustaceans, worms, hard roe and small fish. It becomes sexually mature when 2 years of age and 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.*, 1986]. Integrated monitoring of fish

physiology and population development is carried out on eelpout in cooperation with the University of Gothenburg and the Department of Aquatic Resources at the Swedish University of Agricultural Sciences (SLU AQUA).

### 4.2.5   Blue mussel (*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.

### 4.2.6   Common Guillemot (*Uria aalge*)
Guillemots feed mainly on sprat (*Sprattus sprattus)* and herring (*Clupea harengus*). They breed for the first time at 4–5 years of age. Eggs hatch after about 32 days. The egg content is high in fat (11–13%), thus very appropriate for analysis of fat-soluble contaminants e.g. hydrocarbons. They are furthermore 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. Normally, the guillemot lay just one single egg but if this egg is lost, another may be laid. Replacement eggs, often laid later in the season, tend to contain significantly higher concentrations of organochlorines compared to the first laid eggs [*Bignert et al.*, 1995]. Ten guillemot eggs, collected between weeks 19–21, are analysed each year. In this report, only early laid eggs (assumed to be the first laid egg) are included.

### 4.2.7   Common Tern (*Sterna hirundo*)
Common tern feed mainly on small fish and is considered a top-predator in the marine food web [*Lemmetyinen*, 1973; *Reindl and Falkowska*, 2019]. It is a highly migratory seabird with a circumpolar distribution but northern breeding areas [*Austin*, 1953; *Becker and Ludwigs*, 2004]. It normally inhabits Sweden from April-May to September-October [*Becker and Ludwigs*, 2004]. The breeding period ranges from May to July [*Becker and Ludwigs*, 2004]. The common tern lays up to three eggs that hatch after 21–22 days.
Common tern is considered to be an income breeder, i.e. substances forming the eggs largely originate from nutrients incorporated by the female in the two weeks of courtship feeding by the male mate immediately before egg-laying [*Wendeln and Becker*, 1996]. In the breeding season, foraging of common terns takes place in comparatively small distances mostly within 10 km of the breeding colony [*Becker et al.*, 1993].

### 4.2.8   Eurasian Oystercatcher (*Haematopus ostralegus*)
Eurasian Oystercatcher is a wader primarily found at estuarine mudflats, saltmarshes and sandy and rocky shores. Most populations of this species are fully migratory and the Swedish population migrates between late August and mid-March to other parts of the North Sea region. Polychaetes and crustaceans are the main parts of the diet, however, molluscs are most important on rocky shores. Prey, such as earthworms and insect larvae may form an important part of the diet when foraging inland. In the breeding season, foraging of oystercatcher takes place in comparatively small distances mostly less than 5 km of the breeding colony [*Becker et al.*, 1993]. The species breeds from April to July, 2–4 eggs are laid. The oystercatcher is a capital breeder, producing its eggs from substances stored in the body over longer time periods. The species is a resident breeder over large parts of the North Sea area [*Koffijberg et al.*, 2006].

## 4.3 Sampling

### 4.3.1 Number, frequency, and quality of samples

Sampling is carried out once (for two stations twice) per year for all sites to ensure strong statistical and interpretational power to locate temporal changes.

The sampling design has changed over the time the monitoring programme has been running (oldest samples from 1969). From the start of the programme, individual analyses of contaminants were prioritised for the majority of the substances (typically 10-12 individuals were analysed for each species and station per year). More recently, in the first decade of the 2000s, the monitoring programme was expanded from 18 stations to include an additional 14 new herring stations. This was done to provide better geographical coverage. To compensate for the increased costs, due to this geographical expansion and an addition of more substances in the programme, pooled samples (usually 2 pools of 10 or 12 individuals, larger pools for blue mussels), are now predominantly analysed for each species at each site. However, individual samples are still being analysed for some selected species and stations. Consequences of analysing pooled samples instead of individual samples are discussed in Bignert et al. [2014].

Sampling of fish and blue mussels is carried out every autumn, outside of the spawning season. For two sites, Ängskärsklubb and Utlängan, herring is also sampled in the spring. The two springtime time series were started in 1972. In the beginning, only organochlorines where analysed but since 1996 analysis have also included metals. Sites with two yearly samplings provides the possibility for studies on seasonal differences. In addition, it also gives an opportunity to study possible changes in the frequencies of spring and autumn spawners. Guillemot eggs are collected in the first half of May. If a first egg is lost, a second egg is often laid. These second eggs should not be collected since they contain higher levels of contaminants than the first laid eggs and thus increase the variation within the samples [*Bignert et al.*, 1995]. To avoid this, only early laid eggs are sampled.

When possible, healthy looking and undamaged specimens are sampled. The collected specimens are placed individually in polyethylene plastic bags, and are frozen and transported to the laboratory pending sample preparation as soon as possible. For fish, adult individuals of a narrow length range (specific for each species) are chosen for analysis to minimize the within-year and between-year variation. The sampling recommendation also requests a narrow age range for sampled species.

## 4.4 Sample preparation and registration of biological variables

A short description of the sampling matrices and the various types of variables that are registered are given below. See TemaNord [*NMR*, 1995] for further details. The sampling and sample preparations are all performed according to the manual for collection, preparation and storage of fish [*SMNH*, 2012]. Data is stored in a database with other information on the specimen.

### 4.4.1 Fish

For each specimen, total body weight, total length, body length, sex, age, 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, 1 g for organotin compounds, and 0.5 g for analysis of perfluorinated substances.

### 4.4.2   Blue mussel

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 PAH are analysed in pools of approximately 20 specimens on the West Coast and in the Baltic Sea 50 and 75 specimens respectively for organochlorine/bromine and PAHs analyses.

### 4.4.3   Bird egg

Initially, the length, width and total weight of the egg is recorded, after which its contents are removed (blown out, the eggs are collected soon after they are laid and hence the embryos are small) and the total egg content homogenized. Weight of the empty, dried eggshell is then recorded and 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.

## 4.5   Contaminants

Table 1 contains information on the contaminants that are included in the monitoring programme for contaminants in biota and Table 2 presents information on which sites and species the contaminants are analysed.

**Table 1. Summary of source, ecotoxicology and regulations for the contaminants (substances or groups of substances) included in the monitoring program for contaminants in biota.**

| | Anthropogenic sources | Ecotoxicology | Regulations | Referencer |
|---|---|---|---|---|
| Silver (Ag) | Electronics, batteries, by-products in copper and lead smelting, coal combustion, cloud smelting, antiseptic, bacteriostatic agents, and antiodour agents. | Ionic Ag+ is the most toxic silver salt. Ag binds to the metal-binding protein metallothionein in liver and kidney. | | [*Eisler*, 1996; *IPCS*, 2002; *IVL*, 2007] |
| Copper (Cu) | Fungicide used as wood, leather and fabric preservative. Mining of copper ore, road run-off, and during water treatment. | | | [*Dorsey et al.*, 2004] |
| Mercury (Hg) | Coal fired power plants, small-scale goal mining, wastewater disposal, landfills, and cement and metal manufacturing. | Bioaccumulative and biomagnifying, neurotoxin. The organic form of Hg (methylmercury) accumulates in fish muscle. Embryos and very young animals are most affected by Hg damage due to its ability to interfere with cell division processes and neural development. | 1966: the use of alkyl-Hg in agriculture and paper pulp banned in Sweden. 1990s: Banned from use in thermometers and some electronic components and instruments. 2009: all use banned on the Swedish market. 2011: EU regulation bans export. Identified as a priority hazardous substance in WFD. Included in REACH. | [*Huber*, 1997; *Suzuki et al.*, 1991] |
| Nickel (Ni) | Used in alloy manufacture (e.g. stainless steel), battery industry, appliances and electroplating. Generated from combustion of fossil fuel, domestic and non-ferrous metal smelters, incineration of waste and wastewater effluents, smoking tobacco. | Human exposure primarily through ingestion of contaminated drinking water or food, and inhalation. A common cause of allergic contact dermatitis. Potentially carcinogenic in some animals and modes of human exposure. Can enhance lipid peroxidation in the liver, kidney, lung, bone marrow and serum. | Included in REACH. | [*Andrea*, 2005; *Cempel and Nikel*, 2006; *Denkhaus and Salnikow*, 2002; *Kasprzak et al.*, 2003; *WHO*, 1991] |
| Zink (Zn) | Mining, steel production, coal burning, used in steel and iron galvanization to prevent rust and corrosion. | Not believed to bioaccumulate, but changes found in biological tissues may still reflect changes in ambient water concentration. Dissolved Zn can increase water acidity. | | [*ATSDR*, 2005] |
| Lead (Pb) | Ammunition, leaded petrol, leaded pigments, cables, and batteries. | Bioaccumulates in soft tissues and to a greater extent the bone matrix. A neurotoxin that damages the nervous system. Can cross the blood-brain barrier and the placenta leading to highest risk for unborn and small children. Neurophysiological development affects are seen in children at low exposure. Lead shots are a common cause of poisoning for birds. | 1995: banned in gasoline in Sweden. Later, also banned in paint, toys, electrical equipment and jewelers. Not allowed in ammunition for hunting in wetland areas or during clay pigeon shooting. 2016: Banned in all goods that children can put in their mouth. Included in REACH. | [*Cook and Trainer*, 1966; *Gidlow*, 2004; *Klaassen and Rozman*, 1991] |
| Cadmium (Cd) | It is an impurity in phosphate rock and used in rechargeable batteries, colour pigment in paints and a stabiliser in plastics and for metal plating and alloys and fertilizer manufacturing. | Bioaccumulate and biomagnify. Chronic exposure results in accumulation of Cd in kidney and liver, leading to kidney damage and renal tube dysfunction. Cd can bind to specific metal-binding proteins in the body which prevents it from exerting its toxicity. Reproductive and carcinogenic effects reported. | 1982: Cd was banned for use as electrogalvanisation and as a thermal stabiliser in Sweden. 1987: a national fee on batteries was introduced. 1993: restriction to use cadmium in fertilizers. Cd identified as a priority hazardous substance in WFD. Cd included in REACH. | |
| Chromium (Cr) | Used in manufacturing of stainless steels, electroplating, leather tanning, pigments for inks | At high levels Cr can cause nausea, skin ulcerations and lung cancer depending on exposure | Chromium (IV) included in REACH. | |

| | | | | |
|---|---|---|---|---|
| | and paints. | pathway and amounts of uptake. | | |
| Arsenic (As) | Generated from industrial smelters and coal power plants and used in production of pesticides and herbicides. Dumped chemical ammunitions from the end of World War II might contribute to increased levels in the Baltic Sea, Skagerrak and Kattegat. | Bioaccumulative but does not biomagnify. Acute, subacute and chronic effects can involve the respiratory, gastrointestinal, cardiovascular, nervous, and haematopoietic systems. Disturbance of the liver function observed in both humans and animals after chronic exposure. Might affect the human heart. | 2006: use of As as a wood preservative is restricted within EU. As included in REACH. | [*Eisler*, 1994; *Garnaga et al.*, 2006; *HELCOM*, 2010; *OSPAR Commision*, 2005; *SGU*, 2005; *United Nations Environment Programme et al.*, 1981] |
| Selenium (Se) | Originates from impurities in metal sulphide ores and is often produced as a by-product during the electrolytic refinement of these. Commercially, Se is used for various applications e.g. in glass, in electronics, as a vulcanizing agent in the rubber industry, as semiconductors in photocells, frame-proofing in electric cables, and an antifungal agent in pharmaceuticals. | Biomagnifies. Both acute and sub-chronic effects in aquatic organisms; mortality as well as reproductive failure and reduced growth reported for a number of fish species. Reduced weight and reproductive failure seen in aquatic birds. Se can also reduce the toxicity for a number of metals e.g. Hg and Ag by forming inert selenide complexes. | Se is regulated in drinking water and the maximum level within EU is 0.07 mg/day (adults), 0.015 mg day/day (children) (EFSA). | [*Ikemoto et al.*, 2004; *Lemly*, 2004] |
| DDT | A persistent synthetic pesticide that primarily degrades to DDE and DDD. It was used as vector control during the second World War. It has since been used for control of agricultural pests, of vector diseases (e.g. Malaria), of ectoparasites in farm animals and insects in domestic and industrial premises. | Severe health effects on wildlife. For example, reduced reproductive success for fish-eating birds due to factors such as failure to return to nesting sites, egg shell thinning and inability to hatch and nestling brood size. Embryo mortality, thyroid malfunction, and immunosupression have been documented. | 1970: DDT was partially banned. 1975: completely banned in Sweden. Included in the Stockholm Convention. | [*Hamlin and Guillette Jr*, 2010; *Helander et al.*, 2008; *Li and Macdonald*, 2005; *Walker et al.*, 2001] |
| HCB | Previously used as a fungicide. Still reaches the environment as a by-product of chlorinating processes, for example pentachlorophenol and vinyl chloride monomer production. | Carcinogenic | 1980: totally banned in Sweden. Banned as fungicide in all Baltic countries. Included in the Stockholm Convention. Identified as a priority hazardous substance in WFD. | |
| HCH | Insecticides used for control of agricultural pests and parasites in farm animals. | Persistent, bioaccumulative and biomagnifying. Immunotoxic, reproductive and developmental effects in laboratory animals and aquatic organisms. Potentially carcinogenic effects in humans. | 1970s: use was restricted. 1978: use prohibited in agriculture. 1988: remaining usages banned. Included in the Stockholm Convention. Identified as a priority hazardous substance in WFD. Included in REACH. | [*Li and Macdonald*, 2005] |
| Dioxins | Not produced intentionally but formed as by-products in industrial and combustion processes. Found as minor impurities in several chlorinated chemical products (e.g. PCBs, chlorophenols, hexachlorophene etc.). Historically, pulp bleaching was an important source. | Can cause a variety of biological and toxicological effects in animals and humans. The most relevant toxic effects are developmental toxicity, carcinogenity and immunotoxicity. | Included in the Stockholm Convention and the Convention on Long Range Transboundary Air Pollution. EU legislations covers e.g. the plan for integrated pollution prevention and control and the directive on waste incineration. | |
| PCB | Synthetic chemicals used in manufacturing processes, especially as plasticizers, insulators and fire retardants. Widely distributed in the environment through e.g. inappropriate handling of waste material or leakage from large capacitors and hydraulic systems. | Fat and oil soluble. Bioaccumulate in biota to high concentrations. Can influence human health by affecting multiple organ systems. The toxicological effects on e.g. reproduction in mink is well documented. Degrade very slowly. | 1973: banned in open systems. 1978: all new use forbidden. Included in the Stockholm Convention. | [*ATSDR*, 2000; *Aulerich and Ringer*, 1977; *Bleavins et al.*, 1980; *Carpenter*, 1998; 2006; *Jensen et al.*, 1977; *Newman and Unger*, 2003] |
| PBDE and | PBDE and HBCDD are used as | Several PBDE congeners and | 2004: penta- and octaBDE | [*Darnerud*, 2008; |

| | | | |
|---|---|---|---|
| HBCDD | additive flame retardants in plastics and textiles. PBDEs leak into the environment during production, use, or disposal of such products. PBDE are mainly spread via diffuse distribution in the atmosphere and in rivers. | HBCDD cause neurotoxic effects in rats and mice. In mammals, effects on behavior, learning and hormonal functions have been reported. Animals exposed during sensitive stages of brain development show reduced memory and learning disabilities. In birds, reduced reproductive success is documented. PBDE are endocrine disruptors. | were banned in EU. 2008: DecaBDE was included in the RoHS directive - restrictions in electronical equipment. PBDE and HBCDD are included in the Stockholm Convention. PBDE included as prioritized substances within WFD. | *Eriksson et al.*, 2006a; *Eriksson et al.*, 2006b; *Fernie et al.*, 2009; *Legler*, 2008; *UNEP and WHO*, 2013; *Viberg*, 2004] |
| PFAS (e.g. PFOS,PFOA) | Persistent anthropogenic surfactants. Used industrially (e.g., production of fluoropolymers) and commercially (water and stain proofing agents and fire-fighting foams) since the 1950s. Emitted to air and water during production and application. Secondary emissions from consumer products and sewage treatment plant. PFAA also produced during degradation of semi-volatile precursor compounds. Human exposure is believed to be primarily through dietary intake. Contaminated fish from the Baltic Sea is also a source. | Accumulate in protein rich tissues (blood, liver and eggs). Toxic effects in laboratory experiments (mostly rodents) include weight loss, liver enlargement, immunotoxicity and developmental effects. The common carp experienced lowered condition factor and hepatosomatic index with increased exposure. In humans, concentrations of PFOA in maternal blood and PFOA and PFOS in cord blood during pregnancy is negatively associated with birth weight, ponderal index, head circumference[34], and birth length. | 2009: PFOS and its salts were included in the Stockholm Convention. 2019: PFOA was added to the Stockholm Convention. | [*Apelberg et al.*, 2007; *Berger et al.*, 2009; *Buck et al.*, 2011; *Fei et al.*, 2008; *Hagenaars et al.*, 2008; *Vestergren and Cousins*, 2009] |
| PAH | Produced naturally (e.g. in smoke from forest fires or in oil deposits) and anthropogenically (e.g. incomplete combustion of organic materials). The largest input to the environment comes from human activities, such as waste from industrialized and urbanized areas or petroleum production and transportation. | Found in nature as complex mixtures of many components with varying toxic potencies, many of which are considered carcinogens. | Identified as a priority hazardous substance in WFD. | [*Nisbet and Lagoy*, 1992; *Petry et al.*, 1996; *Soclo et al.*, 2000] |
| OTC (e.g. TBT, TPhT) | Antifouling agents in paints used to prevent attachment of barnacles and slime on ship hulls, docks, buoys, and fishnets. From the paint it slowly leaches into the water. Also used as a wood preservative in industry and agriculture and as a stabilizer in PVC plastics manufacturing. | Toxic at low doses. Bioaccumulates in gastropods. An endocrine disruptor that can induce imposex in gastropods. | 1989: use of TBT on small boats banned in Sweden. 1993: all use of TBT prohibited in Sweden. 1999: ban on small boats in EU. 2008: International ban on TBT and other OTC. Identified as a priority hazardous substance in the WFD. | [*Encinar et al.*, 2001; *Smith*, 1981; *Sternberg et al.*, 2010; *SWE-EPA*, 2008] |

**Table 2. Summary of the collected specimens from different sampling sites and analyses of different contaminant groups. Coordinates system is RT90 (X=North coordinate), Y=East coordinate), SIA, Stable isotope analysis; PCB, polychlorinated biphenyls; OCPs, Organo chlorinated pesticides; PBDE, polybrominated diphenyl ethers; PFAS, per- and polyfluorinated substances; PAH, polyaromatic hydrocarbons; OTC, organotin compounds.**

| Sampling site | Species | X | Y | PSU | Metals + SIA | PCB + OCPs | Dioxins and furans | PBDE + HBCDD | PFAS | PAHs | OTCs |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1. Rånefjärden | | 7310900 | 1802700 | <3 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | | | | | | | |
| 2. Harufjärden | | 7294000 | 1825900 | <3 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 3. Kinnbäcksfjärden | | 7204900 | 1759200 | - | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | | | | | | | |
| 4. Holmöarna | | 7073600 | 1750800 | 4 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | x | x | x | x | x | | x |
| 5. Örefjärden | | 7039900 | 1679300 | - | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | x | x | x | | x | | x |
| 6. Gaviksfjärden | | 7005100 | 1642800 | - | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | | | | | | | |
| 7. Långvindsfjärden | | 6852200 | 1587100 | - | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | | | | | | | |
| 8. Bothnian Sea (offsh.) | | 6798326 | 1698277 | - | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 9. Ängskärsklubb | | 6715100 | 1629400 | 6 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | | | | | | | |
| 9. Ängskärsklubb - spring | | 6715100 | 1629400 | 6 | | | | | | | |
| | Herrring | | | | x | x | x | x | | | |
| 10. Sea of Åland (offsh.) | | 6686566 | 1696248 | 6 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 11. Lagnö | | 6593400 | 1660100 | 6-7 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| | Perch | | | | | | | | | | |
| 12. N. Baltic Proper (offsh.) | | | | 6-7 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 13. Landsort | | 6510000 | 1627500 | 6-7 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 14. Kvädöfjärden | | 6434800 | 1556700 | 6-7 | | | | | | | |
| | Perch | | | | x | x | x | x | x | | x |
| | Eelpout | | | | x | x | | x | x | | |
| | Flounder | | | | | | | | | | |
| | Blue mussel | | | | x | x | | x | | x | |
| 15. Byxelkrok | | 6365800 | 1571500 | 7 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 16. Stora Karlsö | | 6352800 | 1631500 | 7 | | | | | | | |
| | Common guillemot | | | | x | x | x | x | x | | |
| 17. SE Gotland | | 6294700 | 1664600 | 7-8 | | | | | | | |
| | Cod | | | | x | x | | x | x | | |
| 18. Utlängan | | 6208830 | 1501600 | 8 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 18. Utlängan - spring | | 6208830 | 1501600 | 8 | | | | | | | |
| | Herring | | | | x | x | x | x | | | |
| 19. E Bornholm Bas. (offsh.) | | 6181395 | 1606416 | - | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 20. W Hanöbukten | | | | 8 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 21. Abbekås | | 6134000 | 1360700 | 8 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 22. Kullen | | 6249400 | 1288200 | 20-25 | | | | | | | |
| | Herring | | | | x | x | x | x | x | | |
| 23. Fladen | | 6348600 | 1258800 | 20-25 | | | | | | | |

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | Herring | | | | x | x | x | x | x |
| | Cod | | | | x | x | | x | x |
| | Dab | | | | | | | | |
| 24. Nidingen | | 6368600 | 1265100 | - | | | | | |
| | Blue mussel | | | | x | x | | x | | x |
| 25. Väderöarna | | 6502000 | 1218300 | 25-30 | | | | | |
| | Herring | | | | x | x | x | x | x |
| | Flounder | | | | | | | | |
| 26. Fjällbacka | | 6510100 | 1236200 | | | | | | |
| | Eelpout | | | | x | x | | x | x |
| | Blue mussel | | | | x | x | | x | | x |
| 27. Tjärnö | | 6539496 | 1230900 | 30 | | | | | |
| | Common tern | | | | x | x | x | x | x |
| | Eurasian oystercatcher | | | | x | x | x | x | x |

## 4.6 Analytical methods

Methods covering the entire time period of the monitoring programme are presented below. Results from 2009 to 2018 are presented in the main report below while results for the entire time period can be found in the Appendix. Names of the specific contaminants that belongs to the groups of compounds listed in the method section below and which are part of the monitoring programme is found in section 7 (Result section) and Appendix.

### 4.6.1 Trace metals

The analyses of metals concentrations in fish liver and fish muscle (mercury), blue mussel soft body and bird eggs, are carried out at the Department of Environmental Science and Analytical Chemistry (ACES) at Stockholm University. Analytical methods for metals in liver are performed according to the Swedish standards SS-EN 13805:2009 (Foodstuffs – Determination of trace elements – Pressure digestion) and SS-EN ISO 17294-2:2005 (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 is accredited by SWEDAC for determination of dry matter (by freeze drying), Ag, Al, As, Cd, Cr, Cu, Ni, Pb, Zn and Hg in biota, and participates in the periodic QUASIMEME intercalibration rounds.

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

### 4.6.2 Organochlorines and brominated flame retardants

The analyses of organochlorines and brominated flame retardants are carried out at the Department of Environmental Science and Analytical Chemistry (ACES) at Stockholm University. Specific analytical methods applied are described in the respective chapters where applicable. Before 1988, organochlorines were analyzed 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 and co-workers [*Jensen et al.*, 1983] where wet tissues

are extracted with a mixture of polar and non-polar solvents. The organochlorines are analyzed on a gas chromatograph (GC) equipped with a μ-electron capture detector [*Eriksson et al.*, 1994]. The brominated flame retardants BFRs are analyzed 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 CB-28, -52, -101, -118, -153, -138, -180, HCB, p,p'-DDE, p,p'-DDD, p,p'-DDT as well as α-, β- and γ-HCH in biological tissues. So far the BFRs are not accredited but the analysis of BDE-28, -47, -99, -100, -153, -154 and HBCDD 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 origin of all standards is well documented with known purity and certified concentration with uncertainty for the solutions.

## Selectivity

To have the possibility to control impurities in solvents, equipment and glassware, one blank sample is extracted together with each batch of environmental samples.

Co-elution 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 co-elution, two 60 m columns are used in parallel, the commonly used 5 % diphenyl - 95% dimethylpolysiloxane phase and the more polar 14 % cyanopropylphenyl – 86% dimethylpolysiloxane phase. The only remaining known co-elution is for CB-138, which co-elutes with CB-163 [*Larsen and Riego*, 1990]. Therefore CB-138 is reported as CB138+163. PBDE and HBCDD are analyzed on a 30 m column, 5 % diphenyl - 95% dimethylarylenesiloxane phase (TG-5SilMS), monitoring m/z 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 / new sampling locations are also examined for suitable surrogate standards.

## Reference Material

Three 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 and pike 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.

## 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 deviations of the assigned value. In 2000, the laboratory participated in the first interlaboratory study ever performed for PBDEs and HBCDD, contaminants that since 2001 are incorporated in the QUASIMEME proficiency testing scheme. Around 85% 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 8000 PCB- and pesticide values during a period of nearly 20 years and around 2000 BDE- and HBCDD values during nearly 15 years. The bias is estimated from proficiency testing of more than 8 samples during at least 4 years. The bias for PBDE is used also for HBCDD 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 3). The reproducibility for the PCBs and pesticides follows the theory stated by Horwitz where the relative standard deviation increase when the concentration level decrease [*Horwitz and Albert*, 2006]. The reproducibility for PBDE and HBCDD follows a function where the relative standard deviations increase first at the very lowest concentration.

**Table 3. Expanded uncertainty (%) at different concentrations**

|  | CB-28, -101, -118, -153, -138, -180, HCB | CB-52 | HCH $\alpha$, $\beta$, $\gamma$ | DDE & DDD | DDT |  | PBDEs | HBCDD |
|---|---|---|---|---|---|---|---|---|
| ng/g lw | % | % | % | % |  | ng/g lw | % | % |
| 2–50 | 36 | 49 | 40 | 43 |  | 0.2–1 | 73 |  |
| 4–50 |  |  |  |  | 52 | > 2 | 58 |  |
| > 50 | 29 | 30 | 34 | 31 | 38 | 2–25 |  | 103 |
|  |  |  |  |  |  | > 25 |  | 64 |

The quantification limit is estimated to approximately 2 ng/g fat weight for all analysed PCBs, $\alpha$, $\beta$, $\gamma$-HCH, HCB, DDE and DDD and 4 ng/g fat weight for DDT. For all analysed PBDEs the quantification limit is estimated to approximately 0.2 ng/g fat weight and for HBCDD 2 ng/g.

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

### 4.6.4   Perfluoroalkyl substances
The analyses of perfluoroalkyl substances were carried out at the Department of Environmental Sciences and Analytical Chemistry (ACES), Stockholm University.

**Sample preparation and instrumental analysis**
A sample aliquot of approximately 1.0 g homogenized tissue in a polypropylene (PP)-centrifuge tube was spiked with 1.0 ng each of a suite of mass-labelled internal standards ([18]O- or ^{13}C-labelled perfluoroalkyl 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 and volumetric standards M8PFOA and M8PFOS were added. Precipitation during overnight freezing occurred and the extract was centrifuged before the clear supernatant was transferred to an autoinjector vial for instrumental analysis. Aliquots of the final extracts were injected automatically on an ultra performance liquid chromatography (UPLC) system (Acquity, Waters) coupled to a tandem mass spectrometer (MS-MS; Xevo TQS, Waters). Compound separation was achieved on a BEH C18 UPLC column (1.7 µm particles, $50 \times 2.1$ mm, Waters) with a binary gradient of ammonium acetate buffered acetonitrile and water. The mass spectrometer was operated in negative electrospray ionisation mode. Quantification was performed in selected reaction monitoring chromatograms using the internal standard method.

**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 et al.*, 2005]. Recoveries during this study were determined by spiked in-house fish control samples. The recoveries ranged between 69 and 125% for all analytes except PFTeDA with an average recovery of 53.9%. Method quantification limits (MQLs) for all analytes were determined either on the basis of blank extraction experiments or if no blank contamination was detectable on the lowest calibration point and ranged between 0.012 and 0.16 ng/g wet weight for the different compounds.

### 4.6.5   Polycyclic Aromatic Hydrocarbons
The analysis of PAH is carried out at IVL, the Swedish Environmental Research Institute. The extraction and analysis of the samples were performed according to IVLs accredited method for PAH.

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 Agilent 1290, with a 3 µm $C_{18}$-column (Pursuit PAH 100* 3 mm, Agilent). A linear gradient elution program was used, starting with acetonitrile/water 50:50 and ending with 100% acetonitrile (Rathburne HPLC-grade). A fluorescence detector (Agilent 1260) with a wavelength program optimised for each PAH was used for

quantification. The peak heights were registered with a chromatographic system from Thermo (Chromeleon 7.0). The concentrations of 16 different PAH compounds were calculated by comparison to a certified standard, NIST, SRM 1647f (Priority Pollutant Polycyclic Aromatic Hydrocarbons in Acetonitrile).

All of the standards used (both internal standard and quantification standards) are certified with known purity and precision. Certified reference material (CRM) was run in parallel to the mussel samples and used to check the method performance (NIST, SRM 2974a, Organics in freeze-dried mussel tissue (*Mytilus edulis*).

### 4.6.6   Organotin compounds
Analysis of organotin compounds is carried out at IVL Swedish Environmental Research Institute.

To freeze dry samples, internal standards (monoheptyltin, tripropyltin, tripropyltin och tetrapropyltin) and 10 ml hydrobromic acid (konc) were added. The mixture was extracted twice with 20 ml dichloromethane on a shaker. The organic extract was reduced in volume by evaporation under a stream of nitrogen and the solvent changed to 2 ml hexane: methanol 1:1. 40 µL 25% sodium tetraethylborate in tetrahydrofuran was added and was allowed to react for 2 h at room temperature. After cooling, water was added and the hexane phase (together with an additional hexane extract) was reduced in volume and was cleaned up on an alumina column with hexane as solvent.

A six point calibration curve was prepared by diluting Organotin mix 8 stock solution (LGC Promochem) to which fresh solutions of monophenyltin trichloride and diphenyltin dichloride in methanol had been added. After ethylation using sodium tetraethylborate water was added and the ethylates extracted with hexane.

Instrumental analysis was carried out using a 7890A gas chromatograph connected to a 7010 triple quadrupole mass spectrometer (Agilent) used in electron ionization and multiple reaction monitoring (MRM) mode. For most compounds two MRMs were recorded, one as a quantifier and the other as a qualifier to increase specificity. Certified reference materials are used to check the performance of the method.

## 4.7   Methods for biological data analysis

### 4.7.1   Fat percentage
In the present investigation, the sample's fat content is determined after the extraction with acetone and hexane with 10 % ether without heating [*Jensen et al.*, 1983]. Results of the fat determination may vary considerably depending on the applied extraction method. In the present report, two laboratories determine the fat content during their compound analysis. For consistency, the fat content determined by the laboratories at the Department of Environmental Science and Analytical Chemistry (ACES) is taken for the calculation, because their fat analysis goes back longest in time.

During the sample preparation of herring muscle tissue, the subcutaneous fat layer was removed. This was done before the analyses of the fat content. It has been shown that samples including skin and subcutaneous fat have at least a 1.5-times higher fat content than samples without skin [*Bignert et al.*, 2005].

The fat content is determined in samples that are analysed for contaminants with high affinity to lipids, such as organochlorines (chlorinated pesticides, PCB, PCDD and PCDF) and brominated flame retardants (PBDE and HBCDD), i.e. in herring, perch and eelpout muscles,

cod liver, blue mussel soft tissue as well as in guillemot, Eurasian oystercatcher and common tern eggs. In general, an extremely low fat content, e.g. due to starvation, may cause elevated concentrations of organochlorines expressed on a fat weight basis. A strong negative correlation between the organochlorine concentrations (expressed on a fat weight basis) and the fat content in spring-caught herring has been shown earlier [*Bignert et al.*, 1993], as well as between the concentration of various metals and the fat content in cod liver [*Grimås et al.*, 1985]. The analysed concentrations of these contaminants were therefore adjusted for varying fat content.

### 4.7.2 Condition

The condition of the fish is a common measure for the well-being of an individual or a population. It can be estimated based on the relation of the weight and length of an individual.

In this report, the commonly used Fulton's condition factor, K [*Vibert and Lagler*, 1961] is used for determining the condition of fish:

$$K = \frac{100 \times weight\ (g)}{[length\ (cm)]^3}$$

## 4.8 Methods for stable carbon and nitrogen isotope analysis

Material for stable isotope analysis was subsampled from the samples prepared for mercury analysis (fish muscle) or metal and mercury analyses (mussel and egg). The sample preparation was carried out at the Department of Environmental Sciences and Analytical Chemistry (ACES), Stockholm University. The analyses of stable isotopes were carried out at the Stable Isotope Facility, UC Davis, California, USA.

### 4.8.1 Preparation of samples

Frozen samples were delivered from the Swedish Museum of Natural History to ACES in acid-cleaned and pre-weighed plastic vials. The samples were freeze-dried for about one week and weighed before and after drying to check that the lyophilisation was successful and to enable calculation of the dry matter (%). After material had been secured for metal or Hg analysis, freeze-dried and pulverized tissue samples were transferred into 5x8 mm tin capsules (Säntis Analytical AG) and weighed. The target weight for dual-isotope analysis of biological samples was 1±0.2 mg. The filled tin capsules were formed into small spheres using clean metal tools and placed into 96-well microplates that were stored dry before being sent for analysis. Sample forms with sample type, weight and position in the microplate were sent electronically to the analysis lab. At least one blank sample (empty tin capsule) and one control sample (perch tissue) were included in each shipment of samples.

### 4.8.2 Analyses of C and N stable isotopes

Total carbon, total nitrogen and the isotope ratios $^{13}C/^{12}C$ and $^{15}N/^{14}N$ were determined by EA-IRMS (Elemental Analysis – Isotope Ratio Mass Spectometry) using a PDZ Europa ANCA-GSL elemental analyser in tandem with a PDZ Europa 20-20 isotope ratio mass spectrometer. Laboratory standards that are calibrated to certified reference materials and of similar composition as the samples, as well as blanks, are repeatedly analysed within each sample run.

Delta values, $\delta^{13}C$ and $\delta^{15}N$, are reported in ‰ relative to international standards V-PDB (Vienna PeeDee Belemnite) for carbon and Air for nitrogen. Laboratory long-term standard deviation is 0.2 ‰ for $\delta^{13}C$ and 0.3 ‰ for $\delta^{15}N$.

## 4.9 Data registration

Data prior to 2018 are stored in a flat ASCII file in a hierarchical fashion, where each individual specimen represents one level. The primary data files are processed through a quality control programme. Questionable values are checked and, if necessary, corrected. Data are retrieved from the primary file into a table format suitable for import to database or statistical software.

# 5  Statistical treatment and graphical presentation

One of the main objectives of the monitoring programme is to detect statistical significant time trends. In this annual report we focus on *recent* rather than *long term* trends. Hence, we only include the past ten years of data (2009-2018) in our statistical analysis, even when longer series exists. While this will lead to a loss of power in detecting trends that remain constant over a longer period, it better reflects the current movements in the levels of contaminants. A wider range of statistics, also applied to longer series of data, can be found in the Appendix.

Note that, due to the large amounts of regression analyses that underlies the results in this report, no formal checks of the validity of statistical model assumptions are done. We refer the reader to the Appendix for alternate models and visualisations of raw data.

## 5.1  Data manipulation

We aim to analyse data in raw form as reported by the laboratories. The only exception to this rule is concentrations reported as being below the Level Of Quantification (LOQ). Such values are included in the analysis as if they were true observations with a value of $LOQ/\sqrt{2}$. Due to the arbitrariness of this procedure, any results based on series with a high rate of values below LOQ should be interpreted with caution (see Appendix).

## 5.2  Computation

Statistical analysis is done in R version 3.6.1 [*R Core Team*, 2019]. For pre-processing and summary statistics, a purpose-built R package, MoCiS [*Lampa*, 2019] is used. The package source code and relevant scripts for reproducing the main results is publicly available at https://github.com/NRM-MOC.

### 5.2.1  Trends at sites

For each contaminant, matrix and sampling site, a yearly index is computed by taking the geometric mean of available measurements. These indices are then log-transformed and a simple linear regression is fitted with calendar year as explanatory variable. If we denote by $\beta$ the slope of the fitted line on the log-scale, an estimate of the trend in yearly percentages is given by $100 \times \left(e^{\beta} - 1\right)$. These percentages are visualised for each group of contaminants in heatmaps, truncated to a maximum absolute value of 15% (see heatmaps in chapter 7). Truncation is performed in order to avoid the colour-scale being dominated by outliers, for numerical values see the summary tables in the Appendix.

The statistical significance of trends can be assessed under model assumptions. For simple linear regression, the classical assumptions involve independence, homoscedasticity (equal variances) and normal distribution of residuals. Since time-series data often exhibit serial correlation, results based on the classical assumptions may be misleading. Hence, when assessing significance, MoCiS use an approach by Zeilies [2004] that is robust against deviations from assumptions of independence and equal variances. In the heatmaps, statistical significance is denoted by * (p < 0.05), ** (p < 0.01) and *** (p < 0.001).

### 5.2.2 Trends at an aggregate spatial level

In addition to the trend analysis performed at each sampling site, a common trend, combining herring sites at a basin level, is also estimated for a set of selected contaminants in Herring, see Figure 1. The data collected during spring at Ängkärsklubb and Utlängan is not included in this analysis. Here, a multiple linear regression model $y = \alpha_{site} + \beta \times t + \epsilon$ is fitted to the combined data for all sites in the basin, where $y$ denotes log-transformed yearly index, $\alpha_{site}$ a site specific intercept, $\beta$ a trend common to the sites in the basin, $t$ calendar year and $\epsilon$ a random variation independent in space and time and with a constant variance. In this way the level of a contaminant may differ within a basin, but the yearly trend is the same for all sites. The assumption of equal trends for sites within a basin, i.e. $\beta = \beta_{site}$, is formally tested with an ANOVA $F$-test comparing the fitted model with one, where the trends are allowed to differ between sites. Note that at this aggregate level, tests and confidence levels are based on the classical assumptions of a linear model, i.e. no correction for autocorrelation or heteroscedasticity is performed.

The results are presented for selected contaminants in figures throughout the Results (chapter 7) and in a table (section 7.11). In the figures, the fitted contaminant level, $e^{\bar{\alpha}+\beta \times t}$, is plotted as a function of calendar year $t$. Here, $\bar{\alpha}$ denotes a weighted average of the site intercepts, where the weights are proportional to estimated standard errors of the components. In case the assumption of equal trends is rejected, the level is drawn with a dashed line. While the trend may still be interpreted as a basin average trend in these cases, measures of uncertainty depend more crucially on the assumption of equal slopes. Hence, confidence bands in the figures and confidence intervals in the table are not displayed where the assumption is rejected.

Since we are mainly interested in *trends*, the confidence bands in the figures only reflect the uncertainty in the trend given a fixed level at the midpoint ($t = 2013{,}5$). Hence, the bands cannot be used to assess uncertainty in *contaminant level*, since uncertainty in the intercept is not taken into account.

# 6 Conventions, regulations, directives and legislations concerning chemicals and pollutants and set target levels

## 6.1 The Stockholm Convention on Persistent Organic Pollutants

The Stockholm Convention on Persistent Organic Pollutants (POPs) is an international agreement that takes measures to protect the environment and humans against adverse effects caused by contaminant. It put into place measures for reducing or preventing the use, release, production and trade of dangerous substances. The Stockholm Convention was adopted in 2001 and came into force in May 2004. The convention deals with organic compounds that are persistent (i.e. remain in the environment for a long time), have a long-range transport potential, bioaccumulate in fatty tissues of organisms, and have adverse effects on humans 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, nine additional chemicals were listed as POPs (hexa-/heptabromdiphenylether, tetra-/pentabrom-odiphenylether, chlordecone, hexabromobiphenyl, lindane, α- and β-hexachlorocyklohexane, pentachlorobenzene and perfluorooctane sulfonic acid (PFOS) its salts and perfluorooctane sulfonyl fluoride (POSF)).

In 2011, technical endosulfan and its related isomers were added to the list with specific exemptions. In 2013, hexabromocyclododecane (HBCDD) was also included and in 2015 Hexachlorobutadiene, Pentachlorophenol with its salts and esters, Polychlorinated naphtalenes were included and in 2017, Decabromodiphenyl ether (commercial mixture, "c-decaBDE") and Short-chained chlorinated paraffins (SCCP)) were included. In 2019 Dicofol and Perfluorooctanoic acid (PFOA) its salts and PFOA-related compounds and was added to the convention. Three more substances have been proposed to be included on the list and are currently reviewed by the Persistent Organic Pollutants Review Committee: Perflurohexane sulfonic acid (PFHxS) its salts and their related compounds, Dechlorane Plus, and Methoxychlor [*SC*, 2019].

## 6.2 The Helsinki Convention

The Helsinki Convention is the Convention on the Protection of the Marine Environment of the Baltic Sea Area. It was signed in 1992 by all states bordering the Baltic Sea, entered into force in 2000, and is governed by the Helsinki Commission (HELCOM). The main focus of the convention is to protect the marine environment of the Baltic Sea from all sources of pollution, even coming from inland waters. The future vision is to have a healthy and well-functioning 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 2021. The joint monitoring of pollutants in the Baltic Sea is an important effort to evaluate the status of the Baltic Sea. Data from the Swedish National Monitoring Programme for Contaminants in marine biota is reported to HELCOM every year via the International Council for the Exploration of the Sea (ICES) (HELCOM).

### 6.3 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 the meeting of The Oslo and The Paris Commissions, and entered into force in 1998. Within OSPAR, six different working areas have been identified as 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 the Strategy for the Joint Assessment and Monitoring Programme. The OSPAR Hazardous Substances Strategy works towards the pollution prevention in the marine environment. The hazardous substances work is implemented by OSPAR's Hazardous Substances Committee with the ultimate aim to achieve levels near background concentrations for naturally occurring substances, and levels close to zero for synthesised substances. Within OSPAR, hazardous substances are defined as persistent, bioaccumulative and toxic (PBT) substances. Due to the improvement on knowledge, OSPAR continuously updates their chemical lists on chemicals of priority concern, and on chemicals of possible concern. Data from the Swedish National Monitoring Programme for Contaminants in marine biota is reported to OSPAR every year through ICES (OSPAR).

### 6.4 The Convention on Long-range Transboundary Air Pollution

The Convention on Long-range Transboundary Air Pollution (CLRTAP) was initiated in 1972 at a United Nations Conference on the Human Environment in Stockholm. After the scientific finding that acidification in Swedish lakes was caused by sulphur emissions from continental Europe, the necessity for international measures to reduce air emissions and protect environments far from emission source, was addressed. In 1979, the convention was signed in Geneva, and came into force in 1983. Initially, the convention focused on sulphuric compounds only, but eight additional protocols were added since for other groups of substances e.g., nitrogen oxides, volatile organic compounds (VOCs) and persistent organic pollutants (POPs). The CLRTAP does not state binding commitments, but rather encourages signing countries "to limit and, as far as possible, gradually reduce and prevent air pollution".

### 6.5 The Minamata Convention on Mercury

The Minamata Convention on Mercury is a global treaty that recognises the metal as of global concern due to its long-range transport and its induced toxic effects. The convention's purpose is to control and to minimise the release and emissions of mercury induced by human activities, such as mining and the application of mercury in certain products and processes. The final goal is to reduce adverse toxic effects of mercury on human health and in the environment. During two meetings in 2013, the Minamata Convention was agreed on in Geneva, Switzerland, and adapted in Kumamoto, Japan. In 2017, the Minamata Convention on Mercury came into force and has already more than 128 signatories and 116 ratifications in 2020. The convention's name is in remembrance of the thousands of people that died or became severely disabled or sick after a long-term exposure (from the 1930s until the late 1960s) to methylmercury from industrial wastewater that was discharged into the Minamata Bay in Japan.

### 6.6 EU regulations and directives

#### 6.6.1 REACH regulation
Registration, Evaluation, Authorization and Restriction of Chemical Substances (REACH) is the EU chemicals regulation that entered into force on the 1st of June 2007 [*OJEC No. L396 30.12.2006*, 2006]. The responsibility is placed on industry side, on the actors in the

production and trade chain to gather information about their chemicals. Any chemical produced within the EU or entering the EU market are/were registered gradually tonnage depended. The registered dossiers on the chemicals are the basis for their evaluation by the European Chemicals Agency (ECHA), based in Helsinki, and the substance evaluation by member states. Some of the compounds can stick out with their properties and might be of potential concern. They for instance can be listed as substance of very high concern (SVHC), which requires their authorisation before application by ECHA and the provision of information to downstream users on the article content, if it contains more than 0.1% by weight of the SVHC. Further, some of the substances can be restricted to certain conditions during production, placing on the market or use.

### 6.6.2   RoHS directive

The Directive on the Restriction of Hazardous Substances (RoHS) was adopted by the EU in February 2003. The RoHs directive requires to reduce the use of six chemical substances in electrical and electronic products that were released on the market after July 2006: mercury, cadmium, lead, chromium VI, polybrominated biphenyls (PBBs) and polybrominated diphenyl ethers (PBDEs). The maximum allowed weight percent of these substances in electrical and electronic products is set to 0.01% for cadmium and 0.1% for the other substances [*OJEC No. L 174 1.7.2011*, 2011].

### 6.6.3   Water Framework Directive

The Water Framework Directive (WFD) aims to achieve both, a good ecological and a good chemical status of all surface waters and ground water bodies in the EU by 2015. The WFD was adopted in October 2000 and deals with fresh water, coastal-zone and estuary waters. Within the WFD, a list of initially 33 priority substances has been established [*OJEC No. L348 24.12.2008*, 2008], to which further substances were added later. To evaluate if a "good chemical status" has been achieved, threshold values, or Environmental Quality Standards (EQS), have been established for the listed substances (see chapter 6.8). It is the responsibility of each member state to assess and report if the goal has been fulfilled [*The European Commission*, 2000].

### 6.6.4   Marine Strategy Framework Directive

The Marine Strategy Framework Directive (MSFD) was adopted in 2008 with the aim of achieving a good environmental status in all European marine waters by 2020. Two of the eleven identified descriptors for a good environmental status deal with contaminants: "contaminants and pollution effects" and "contaminants in fish and other sea food" (OJEC No. L164 25.6.2008 2008). The implementation of the Swedish waterbodies into the MSFD will be based on the regional international conventions of HELCOM and OSPAR.

## 6.7   Swedish chemical legislation

Sweden has formulated 16 environmental quality objectives, of which one is "A non-toxic environment". This entails that concentrations of non-naturally occurring substances should be close to zero, and naturally occurring substances close to background concentrations. Their impact on human health and ecosystems should further be negligible [*KEMI*, 2014]. The agency responsible for coordinating this work is the Swedish Chemicals Agency (KEMI). The Swedish chemical legislation follows any EU legislation. Many existing national legislations concerning chemicals were replaced by REACH after June 2007 [*KEMI*, 2014].

## 6.8  Target levels for chemical status assessment

Good Environmental Status, in accordance with the Marine Strategy Framework Directive 2008/56/EC (MSFD), is for contaminants defined as "concentrations of contaminants at levels not giving rise to pollution effects" [*OJEC No. L164 25.6.2008*, 2008]. The Good environmental status (GES) is reached if the target level based quality assessment reveals that the threshold is not exceeded. Target levels have been developed within several groups or conventions to protect sensitive organisms from the harmful effects of hazardous substances. Examples for these are the Environmental Quality Standards (EQS) within the European Commission to evaluate the GES, and the Environmental Assessment Criteria (EAC) within OSPAR. In addition to EQSs and EACs, the concentrations of chemicals should also be assessed in animals considering consumption, as wild animals often serve as food sources for humans. Maximum levels for certain contaminants in foodstuffs are set in the Commission Regulation No 1881/2006 [*OJEC No. L364 20.12.2006*, 2006] and are referred to as "food regulation" in Table 4.

The Environmental Quality Standards Directive [*OJEC No. L226 24.8.2013*, 2013] lists Environmental Quality Standards (EQSs) for priority substances and certain other pollutants, as defined in Article 16 (strategies against pollution of water) of the Water Framework Directive [*OJEC No. L226 24.8.2013*, 2013], with the aim to achieve a good chemical status of waters in order to protect pelagic and benthic freshwater and marine ecosystems, as well as humans from adverse effects of chemical contaminants. The EQS values are mostly defined for the annual average (arithmetic mean) concentration of a chemical in water (AA-EQS), which should protect biota against long-term effects in the case of chronical exposure. For some bioaccumulative substances (11 out of 45 substances), EQS thresholds have been set for concentrations of chemicals in biota ($EQS_{biota}$) with the aim to protect a) top predators from risks of secondary poisoning due to bioaccumulation of the chemicals along the food chain ($QS_{biota, sec pois}$) and, b) humans and their health, which might be at risk after the consumption of contaminated food such as fish or mussels ($QS_{biota, hh food}$). In this report, only $EQS_{biota}$ are considered and not the AA-EQS. The final $EQS_{biota}$ threshold is based on the lowest QS in order to protect both groups at risk, i.e. humans and top predators. Substance EQS Data Sheets (SDS) contain background information on the derivation of EQS, available at the CIRCABC webpage [*CIRCABC*].

Within the OSPAR convention, Environmental Assessment Criteria (EAC) have been developed for the interpretation of chemical monitoring data in sediments and biota [*OSPAR Commision*, 2014]. Concentrations below the EACs are considered to present no significant risk to the environment and have a similar purpose like the EQSs. In this report, primarily internationally applied target levels such as EQS, EAC or European Commission recommendations for foodstuffs are used (Table 4). If reliable national target levels have been reported with specific regard to environmental conditions in Sweden, these are considered (e.g. HCH reported by the Swedish Environmental Research Institute, IVL, see Table 4). Only one type of target level (e.g. EACs or EQSs) is applied within each substance group and the one chosen is indicated in bold in Table 4. Concentrations of substances lacking international target levels consequently are presented without any evaluation against target levels (e.g. chromium, copper, zinc, arsenic, silver, aluminum, tin and bismuth).

**Table 4. Target levels for various environmental pollutants; the chosen target levels in the report are bold.**

| Group of substance | | Target levels | | | |
|---|---|---|---|---|---|
| | | Fish (µg/kg ww) | Mussels (µg/kg dw) | | Reference |
| Metals | Cadmium | **160**[1,2] | | QS$_{biota, sec pois}$ | [*EU*, 2005b] |
| | Lead | **300**[1] | 1500 ww | food regulation | [*OJEC No. L364 20.12.2006*, 2006] |
| | Mercury | **20**[1] | | EQS$_{biota}$ (QS$_{biota, sec pois}$) | [*EU*, 2005c] |
| PAHs | Fluoranthene | | **110** / 30 ww | EAC / EQS$_{biota}$ (QS$_{biota, hh food}$) | [*OSPAR*, 2020] / [*Fluoranthene EQS dossier*, 2011] |
| | Anthracene | | **290** | EAC | [*OSPAR*, 2020] |
| | Naphtalene | | **340** | EAC | [*OSPAR*, 2020] |
| | Phenantrene | | **1700** | EAC | [*OSPAR*, 2020] |
| | Pyrene | | **100** | EAC | [*OSPAR*, 2020] |
| | Benzo(a)anthracene | | 80 | EAC | [*OSPAR*, 2020] |
| | Benzo(a)pyrene | | **600** / 5 ww | EAC / EQS$_{biota}$ (QS$_{biota, hh food}$) | [*OSPAR*, 2020] / [*PAH EQS dossier*, 2011] |
| | Benzo(ghi)perylene | | **110** | EAC | [*OSPAR*, 2020] |
| Pesticides | DDE (p,p') | **5** | | EAC | [*OSPAR Commision*, 1998] |
| | ∑HCH (incl. lindane)[4] | **2.6 (marine)** 26 (limnic) | | IVL | [*Lilja et al.*, 2010; *OSPAR Commision*, 1998] |
| | HCB | **10** | | EQS$_{biota}$ (QS$_{biota, hh food}$) | [*EU*, 2005a] |
| PCBs / Other compounds | PCB 118 | **24** lw | | EAC | [*OSPAR*, 2020] |
| | PCB 153 | **1600** lw | | EAC | [*OSPAR*, 2020] |
| | ∑PBDE (BDE-28, -47, -99, -100, -153, and -154)[5] | **0.0085** | | EQS$_{biota}$ (QS$_{biota, hh food}$) | [*BDE EQS dossier*, 2011] |
| | HBCDD | **167** | | EQS$_{biota}$ (QS$_{biota, sec pois}$) | [*HBCDD EQS dossier*, 2011] |
| | ∑PCDD+PCDF (TEQ) | **0.0035** 0.0006[3] | | food regulation QS$_{biota, sec pois}$ | [*OJEC No. L364 20.12.2006*, 2006] [*Doioxin EQS dossier*, 2011] |
| | PFOS | **9.1**[1] | | EQS$_{biota}$ (QS$_{biota, hh food}$) | [*PFOS EQS dossier*, 2011] |

[1] The threshold is set in a different tissue than which is analysed within the monitoring programme. The threshold used for evaluation in the report and appendix has thus been recalculated to represent the corresponding concentration of the threshold in the monitored tissue. Which for lead in liver is: 0.085 µg/g wet weight, for Cd in liver: 6.65 µg/g wet weight and for PFOS in liver: 155 ng/g wet weight. For further information, see *Bignert et al.* [2017].

[2] Target level presented for Cadmium is based on QS$_{biota}$, sec pois, however secondary poisoning is not considered as the most sensitive protection objective for these substances and will thus not provide the highest protection level.

[3] Half the value of QS$_{biota}$ (0.0012 µg/kg ww), sec pois for ∑PCDD+PCDF+dl-PCB (TEQ2005) has been applied as a threshold for ∑PCDD+PCDF (TEQ) since dl-PCB are not included.

[4] The threshold for the time series is used separately for α-HCH, β-HCH, and lindane. This is done to avoid doing sums including data below LOQ, which have been frequent over the last 10 years.

[5] Data on BDE-28 is lacking for most years, therefore it is not possible to compare the sum of PBDEs against the threshold. However, in all areas, the concentration of BDE-47 congener is by itself above the threshold for the sum of BDE-28, -47, -99, -100, -153, and -154.

# 7 Results

## 7.1 Biological data: Length, weight, age, condition, and fat content

The monitoring programme is designed to minimise the influence of the biological variables between years by selectively using fish of the same size and age for contaminant analysis. In line with this strategy, there are in general no systematic temporal changes during the last ten years for the biological variables. A deviation from this is the age of the analysed fish that are increasing at several stations due to difficulty in catching fish in the correct age interval. In addition, Holmöarna stands out with increases in most biological variables for the two fish species caught here: herring and perch. These observed differences in biological variables over the past 10 years are important to consider when evaluating changes in contaminant concentration as several of the monitored contaminants within the national contaminant monitoring programme are bioaccumulative and therefore likely to increase in concentration with both age and size (Table 1).
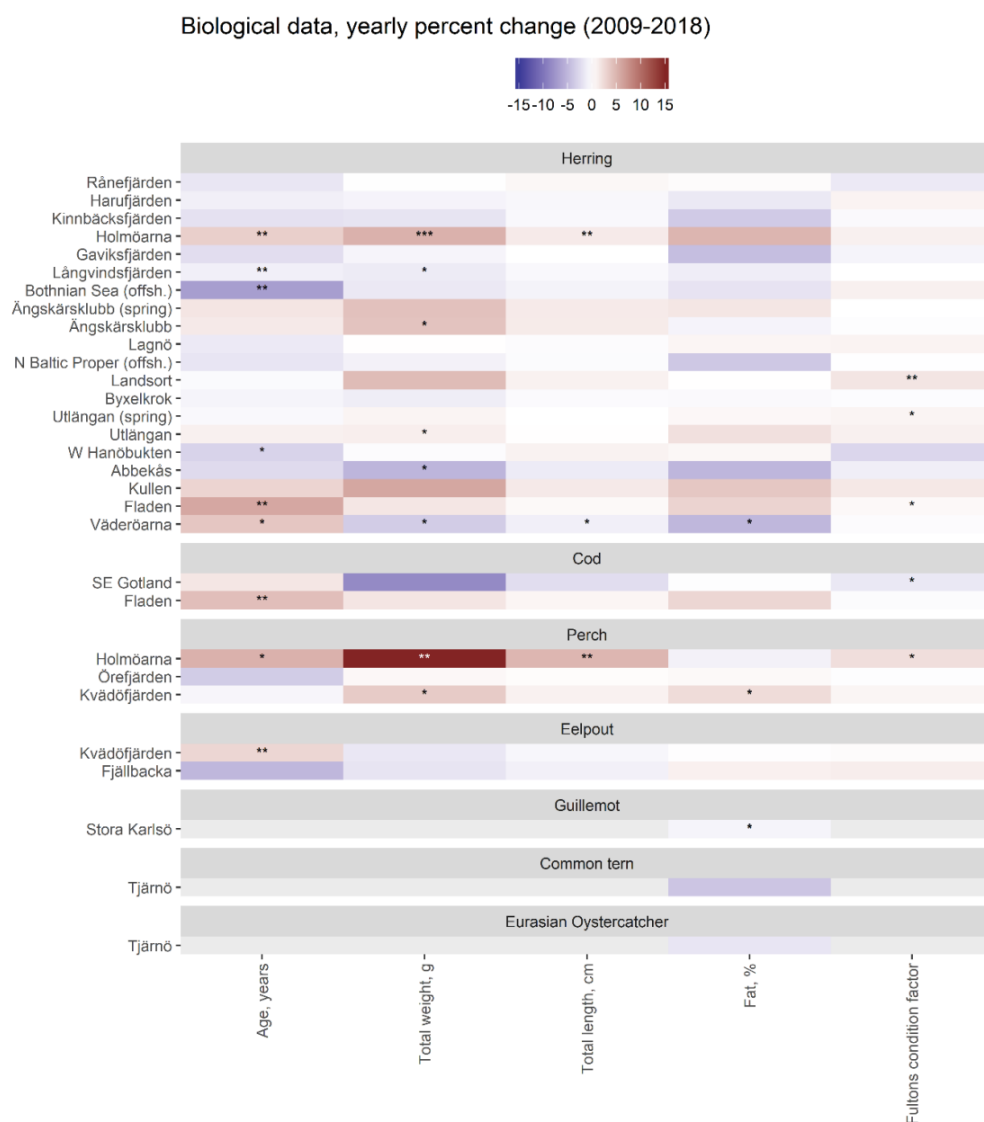


**Figure 2.** The heatmap illustrates the trends of the biological data for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). * p<0.05, ** p<0.01, *** p<0.001.

## 7.2 Stabile isotopes

Two correlation plots between $\delta^{15}N$ and $\delta^{13}C$ shows separation in the isotope signatures between species (Figure 3) and geographical locations (Figure 4). Figure 4, focussing on herring, shows how the basins split into three groups with Bothnian Bay having the lowest values, the Bothnian Sea, Northern Baltic Proper and Southern Baltic Proper having intermediate values and the West Coast having the highest values for both $\delta^{15}N$ and $\delta^{13}C$. The spread within each basin is found to be large, causing considerable overlap between the isotopic signatures. The spread between basins are most obvious for herring, for which there is six years of data from all sites as well as measurements were done at more sites than for the other species, but the difference between the Baltic Proper and the West Coast can also be seen for blue mussel, eelpout and cod (Figure 3).

In aquatic environments, $\delta^{13}C$ can be useful for distinguishing between terrestrial sources (lower $\delta^{13}C$) and pelagic sources (higher $\delta^{13}C$) as well as between benthic production (higher $\delta^{13}C$) and pelagic (lower $\delta^{13}C$) production [*France*, 1995]. Furthermore, $\delta^{13}C$ normally only increase by 0.5-1‰ per trophic level [*Post*, 2002], resulting in the $\delta^{13}C$ of an organism reflecting the $\delta^{13}C$ at the base of the food web. $\delta^{15}N$ are on the other hand enriched by 3-4‰ for a species relative to its diet, which results in higher $\delta^{15}N$ levels at the top of long food webs [*Peterson and Fry*, 1987]. The baseline $\delta^{15}N$ level differs between systems and seasons based on the N sources that feed the system [*Voss et al.*, 2005] and $\delta^{15}N$ values can therefore not be compared across systems without considering the variability in $\delta^{15}N$ at the base of the food web. The blue mussel is a primary consumer and considered as a baseline organism. A baseline organism provides information on the general isotope profile in the ecosystem. For $\delta^{13}C$ this means that the level in the mussels should be reflected in its predators. For $\delta^{15}N$ this means that the trophic position of predators higher in the food web can be calculated based on the relation between the predator's $\delta^{15}N$ and the $\delta^{15}N$ of the baseline species.

The impact of terrestrial sources on the $\delta^{13}C$ is clear when focusing on the change in herring $\delta^{13}C$ across the five basins (Figure 3). Here the lowest $\delta^{13}C$ values in the Bothnian Sea, heavily influenced by terrestrial inputs are seen. The Bothnian Sea and Baltic Proper is influenced less by terrestrial sources and the mix of coastal and offshore stations results in the large spread seen within basins in the $\delta^{13}C$ measurements. This shift towards higher $\delta^{13}C$ between the Baltic Proper and the West Coast is also seen for mussels, cod and eelpout. The difference between the benthic feeders (eelpout and perch; higher $\delta^{13}C$) and pelagic feeders (herring and cod; lower $\delta^{13}C$) could be explained by their different feeding sources. However, the benthic dwelling mussels do not fit into this pattern and generally have lower $\delta^{13}C$ than eelpout and perch.

Separation between basins is also seen for $\delta^{15}N$ where especially samples from the West Coast stand out as being higher than the Baltic Sea. The $\delta^{15}N$ for blue mussel (baseline species) indicates that this difference is 1-2.0‰, which suggests that the $\delta^{15}N$ baseline is higher on the West Coast.

Comparing the difference of $\delta^{15}N$ between basins within the same species furthermore suggests an increase at the West Coast compared to the Baltic Proper with increased trophic position (according to food source). This increase goes from ~2.0‰ in blue mussels to ~3.5‰ in cod. This could indicate a slightly longer food web in the more species rich West Coast ecosystem compared to the Baltic Proper. However, stable isotopes for baseline species are currently only measured at three stations, all coastal and all in the Baltic Proper or on the West Coast. Not all of these stations measure all fish species. This spatial limitation hampers our ability to determine the $\delta^{15}N$ distance between the baseline specie (mussel) and the predatory species and therefore the determination of trophic level of the predatory species in

Baltic Proper offshore waters and in the Bothnian Bay and Bothnian Sea. In addition to a better spatial cover of baseline species stable isotopic levels, there is also a need for more years of data in order to accurately track temporal trends.
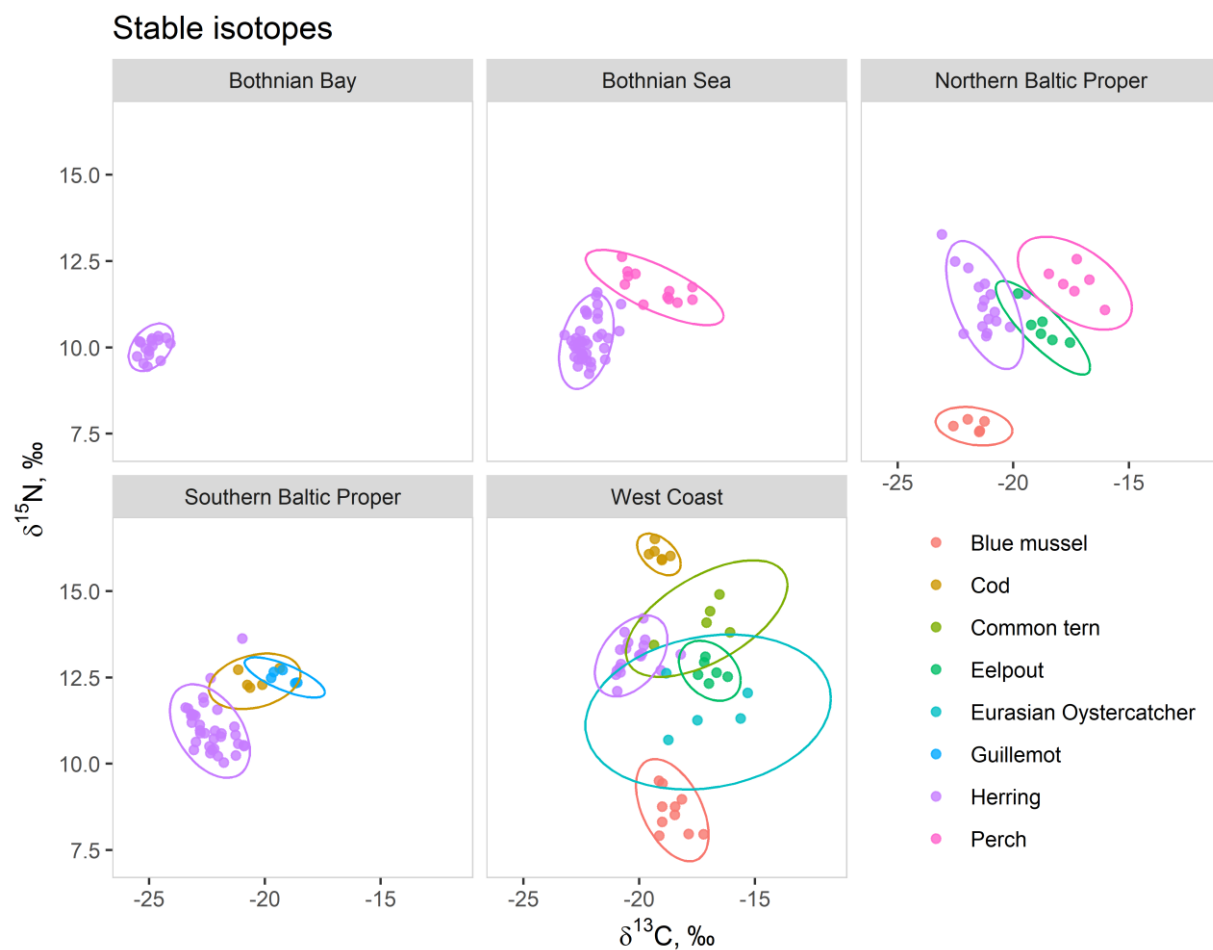


**Figure 3. $\delta^{15}$N vs. lipid normalised $\delta^{13}$C for selected species included in the contaminant monitoring. Data are yearly means at the sampled stations and covers the period 2013-2018.**
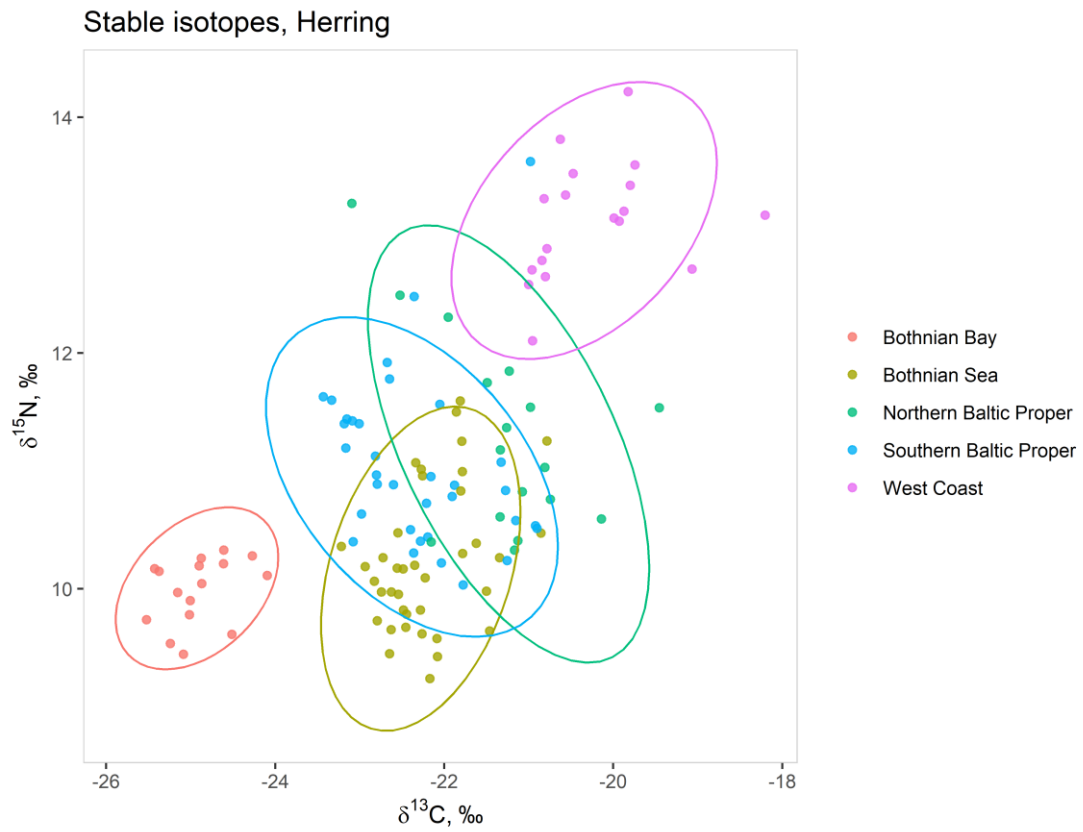
**Figure 4. δ¹⁵N vs. lipid normalised δ¹³C in herring divided into the five regions. Data are yearly means at the sampled stations and covers the period 2013-2018.**

## 7.3 Metals (As, Cd, Cr, Cu, Pb, Hg, Ni, Se, Ag and Zn)

There is no clear trend for any of the metals during the most recent ten-year period when considering all fish, mussel and bird egg time series (Figure 5).

For As, Se and Zn the trends, even when significant, are very small across the last ten years indicating few changes in any of the various biota (Figure 5).

For Ag there is no general trend among the various biota, however, among the significant trends, upward trends dominate as seen for herring from Holmöarna, perch from Kvädöfjärden and cod from SE Gotland (Figure 5).

For Ni there is no general trend but an indication of decreasing concentrations especially for bird's egg and eelpout (Figure 5).
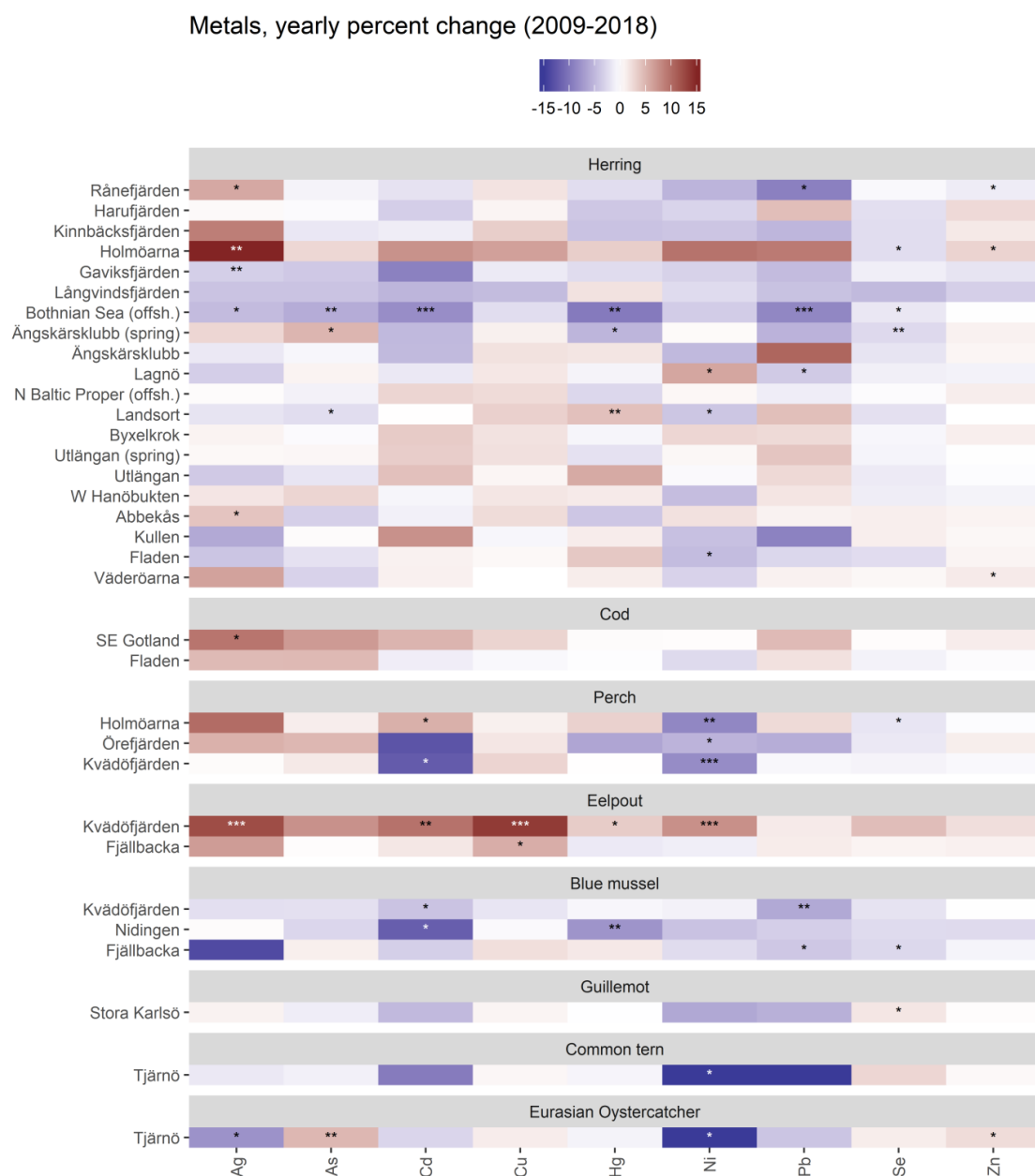


**Figure 5:** The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). * p<0.05, ** p<0.01, *** p<0.001.

While there is no general trend for Cd in the fish, a consistently negative trend is found for Cd in mussels and bird's eggs over the last ten years (Figure 5). Looking at the aggregated basin data for herring (Figure 6) it is seen that there is an indication of decreasing levels in the northern parts of the Baltic Sea (with the exception of Holmöarna where concentrations in perch increase significantly) and an indication of a small increase in the Baltic Proper and on the West Coast. Concentrations are generally higher in the Baltic Proper than the northern Baltic and West Coast.

No change to small increases in Cu concentration are seen in biota for the last ten-year period, the only exception being the faster increase in eelpout (Figure 5). This is also true when looking at the aggregated herring data (Figure 6). Concentrations are similar across all basins.

There are few significant trends for Hg for the last 10 years across the various biota (Figure 5). However, focusing on the aggregated data in Figure 6 a downward trend is seen for the Bothnian Bay and a tendency to an upward trend at the West Coast. Concentrations are highest in the north and decrease towards the south. The levels of Hg in fish are above the threshold at most stations (see Table 2 and Appendix).

While there is no clear trend for Pb, all significant changes for fish over the last ten years are decreasing and for mussels and bird's eggs only decreasing trends are observed (Figure 5). Looking at the aggregated herring data there is an indication of increasing concentrations in the Southern Baltic Proper, and a decrease on the West Coast but the trends are not very strong (Figure 6). Concentrations are generally higher in the Baltic Proper than the Northern Baltic and West Coast.
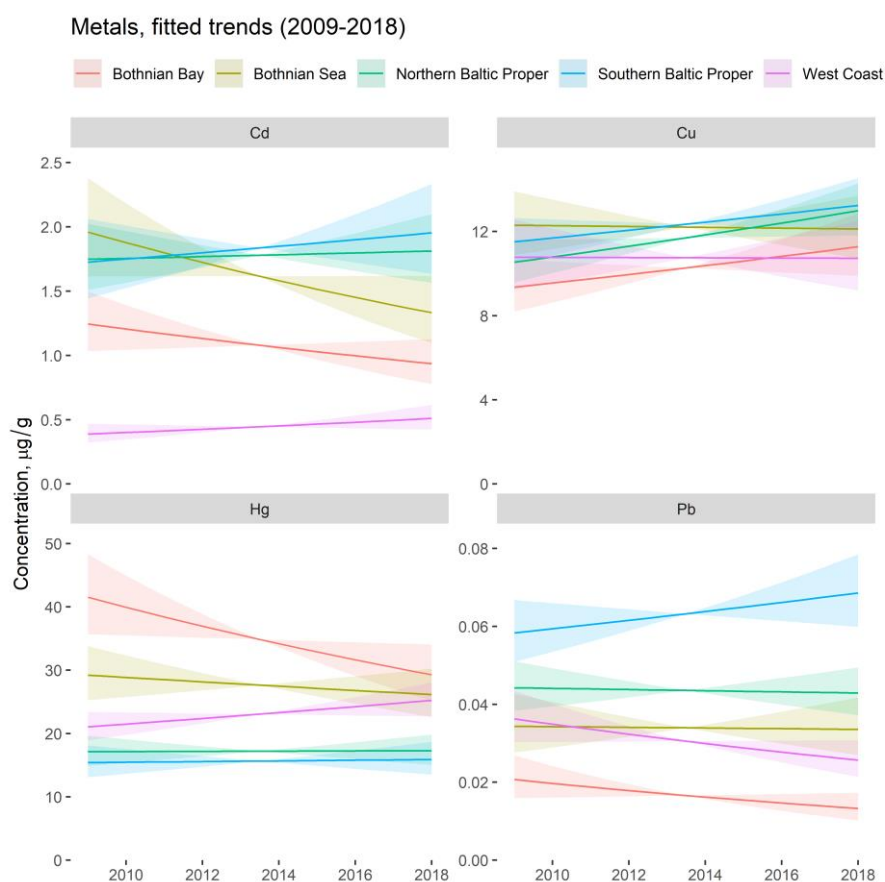


**Figure 6**. Trends at basin level for various metals concentrations in herring. A table with detailed results is found in section 7.11. Basins where there is a significantly different trend between sites are drawn with dashed lines. See section 5.2.2 for interpretation of the 95% confidence bands.

## 7.4   Chlorinated Pesticides (DDT, HCH, HCB)

In general downward trends are seen for DDT, DDD, and DDE in all biota during the last 10 years. A few exceptions being upward trends in herring from Holmöarna, Ängskärsklubb and Väderöarna and blue mussel from Nidingen and Fjällbacka (Figure 7). The upward trends in the herring sites could potentially be driven by the increase in herring age from these sites over the last 10 years (see section 7.1). When looking at the aggregated DDE data for herring it is seen that concentrations are decreasing in all basins except on the West Coast and that concentrations are higher in the Baltic Proper than in the Northern Baltic and West Coast (Figure 8). Concentrations of DDE are below the threshold value in most fish, however, herring from Ängskärsklubb spring, Utlängan spring, Hanöbukten, and cod from SE Gotland and Fladen, have concentrations above the threshold (see Table 2 and Appendix).
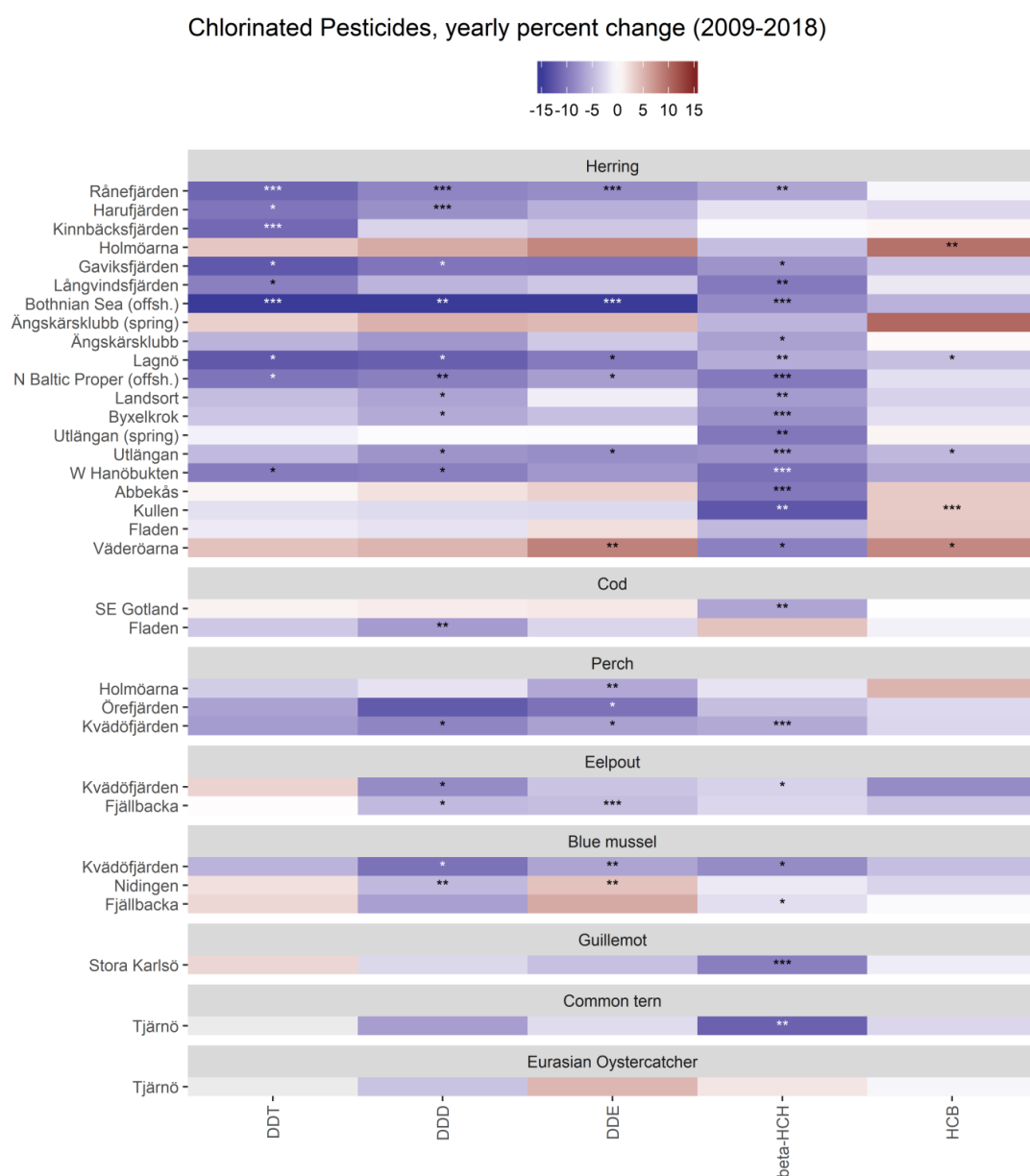


**Figure 7.** The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). * p<0.05, ** p<0.01, *** p<0.001.

β-HCH also shows a consistent decrease at almost all sites and for all types of biota during the last ten years (Figure 7). Concentrations of HCHs are below their respective threshold in all fish (see Table 2 and Appendix).

For HCB there is no general pattern among the biota. Trends are small and non-significant for blue mussels, bird's eggs and fish species, except for a few herring stations where Holmöarna, Kullen and Väderöarna show upward trends and Lagnö and Utlängan downward trends (Figure 7). For herring the aggregated trends indicate downward trends in the Bothnian Bay, and the Northern and Southern Baltic Proper, while an upward trend is seen on the West Coast (Figure 8), driven by increasing concentrations at Kullen and Väderöarna. Similar to DDE, HCB concentrations are higher in the Baltic Proper than in the Northern Baltic and West Coast (Figure 8) and below the threshold (see Table 2 and Appendix).
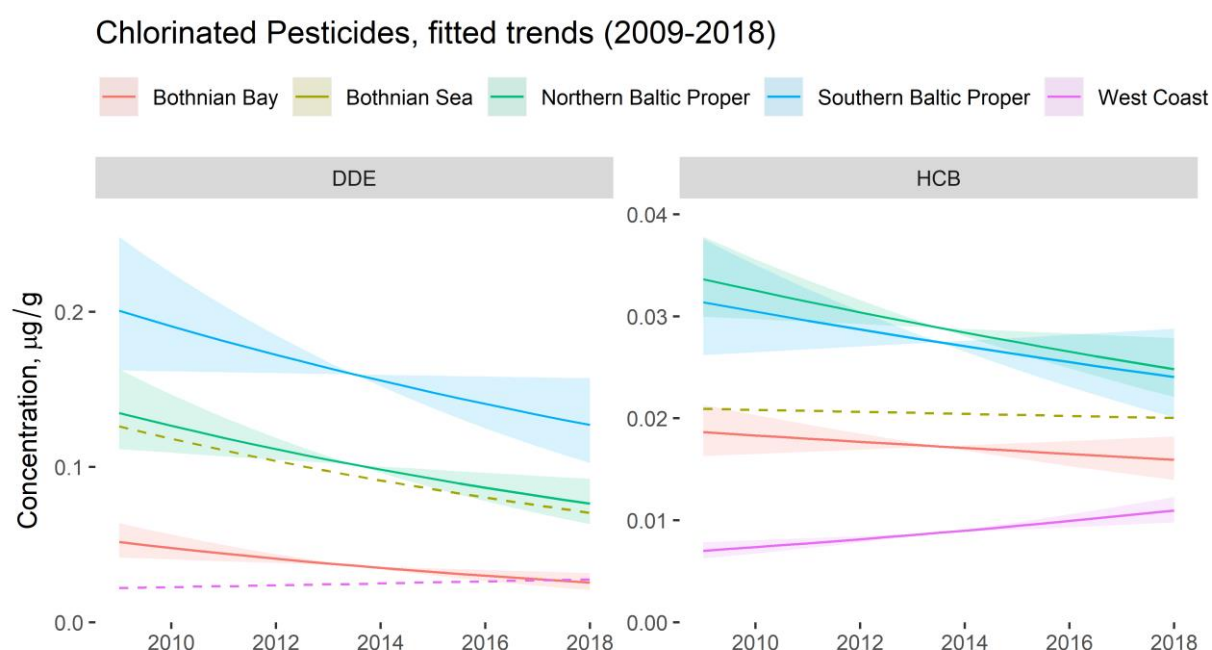


**Figure 8.** Trends at basin level for various chlorinated pesticides concentrations in herring. A table with detailed results is found in section 7.11. Basins where there is a significantly different trend between sites are drawn with dashed lines. See section 5.2.2 for interpretation of the 95% confidence bands.

## 7.5 Polychlorinated Biphenyls (PCB)

The PCB concentrations show a general downward trend across all basins for the last 10 years except the West Coast sites where no general trend is seen but an upward trend in herring from Väderöarna (Figure 9). It is also seen that Holmöarna in the Bothnian Sea show a different trend than the rest of the northern Baltic Sea, which could be due to the influence of the higher age of herring caught at this site during the last years (see section 7.1).
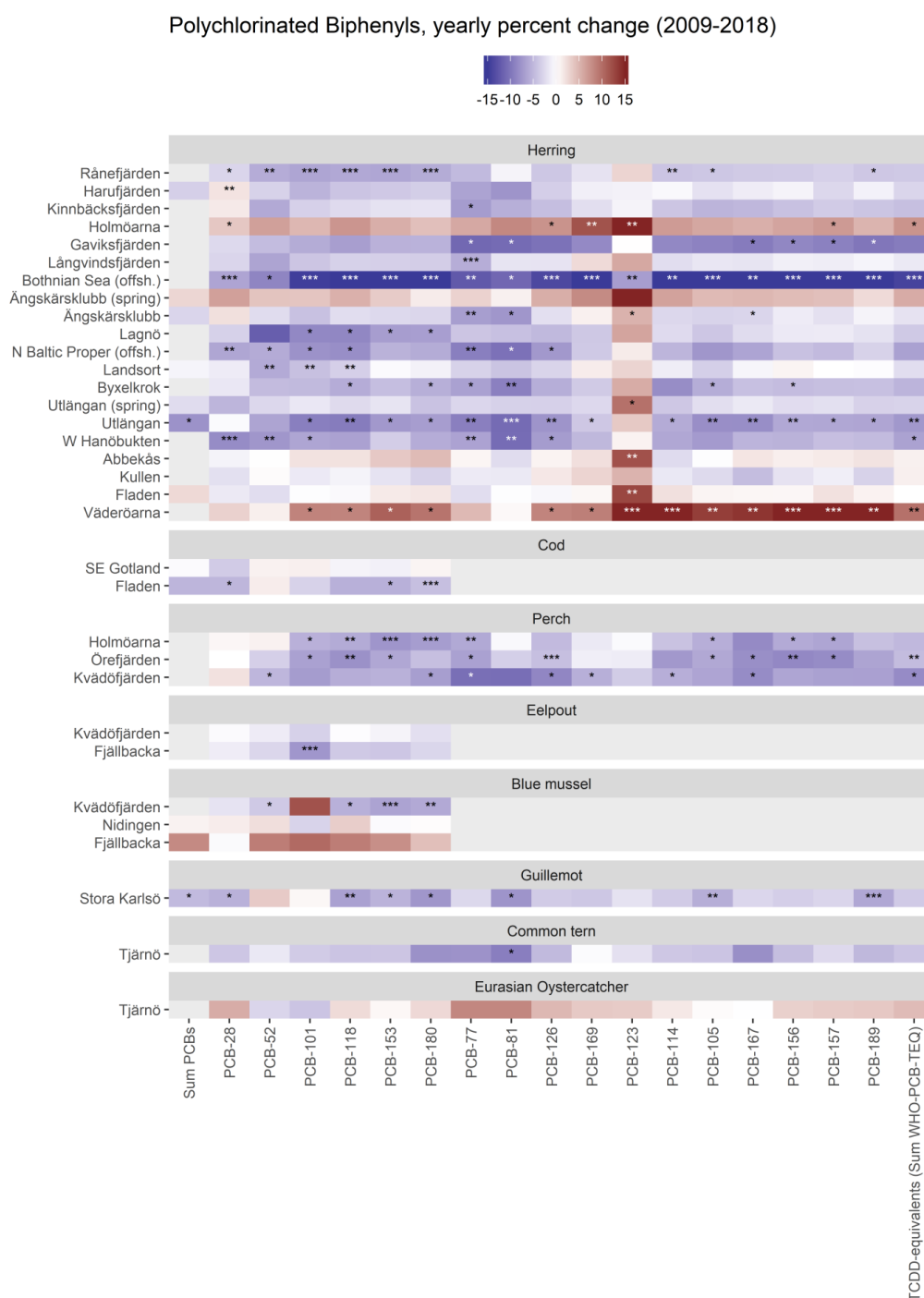


**Figure 9.** The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). * $p<0.05$, ** $p<0.01$, *** $p<0.001$.

Concentrations of PCB-118 and PCB-153 are higher in the Baltic Proper than in the Bothnian Bay and West Coast but in recent years the concentration difference between basins is decreasing (Figure 10). The concentrations of PCB-118 are below the threshold value in most fish, but in herring from Ängskärsklubb spring and in eelpout from Fjällbacka, concentrations are above the threshold (see Table 2 and Appendix). Concentrations of PCB-153 are below the threshold for all fish.
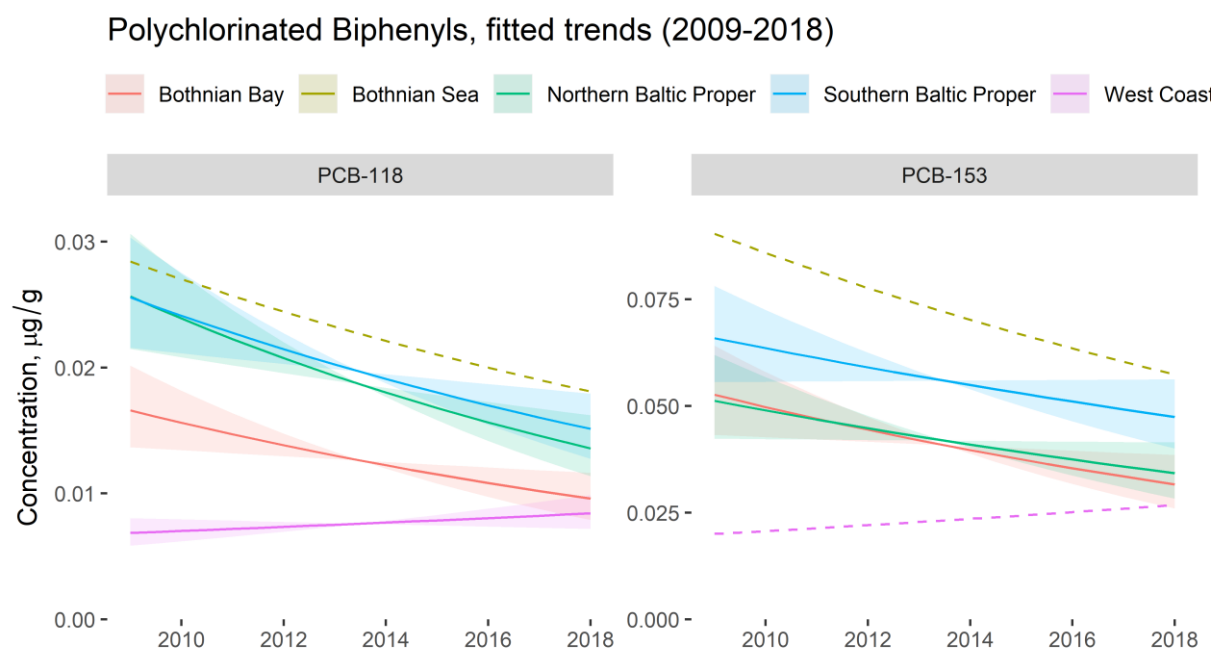


**Figure 10.** Trends at basin level for various PCBs concentrations in herring. A table with detailed results is found in section 7.11. Basins where there is a significantly different trend between sites are drawn with dashed lines. See section 5.2.2 for interpretation of the 95% confidence bands.

## 7.6 Polychlorinated Dibenzodioxins /-furans (PCDDF)

In general, concentrations of dioxins and furans show a clear decrease over the last 10 years except for the West Coast sites where a few sites show increasing trends (Figure 11). The only compounds for which this general trend is less clear is 1,2,3,7,8,9-HxCDF, 1,2,3,4,7,8,9-HpCDF, 1,2,3,7,8,9-HxCDD, but can most likely be explained by difficulties with the analysis and low values. In the Bothnian Sea, Holmöarna and Ängskärsklubb (spring) show trends (increasing) that goes in the opposite direction of the other sites in this area for almost all compounds, a similar increase is also seen for herring from Väderöarna. This pattern could potentially be driven by the increase in herring age from these sites over the last 10 years (see section 7.1).
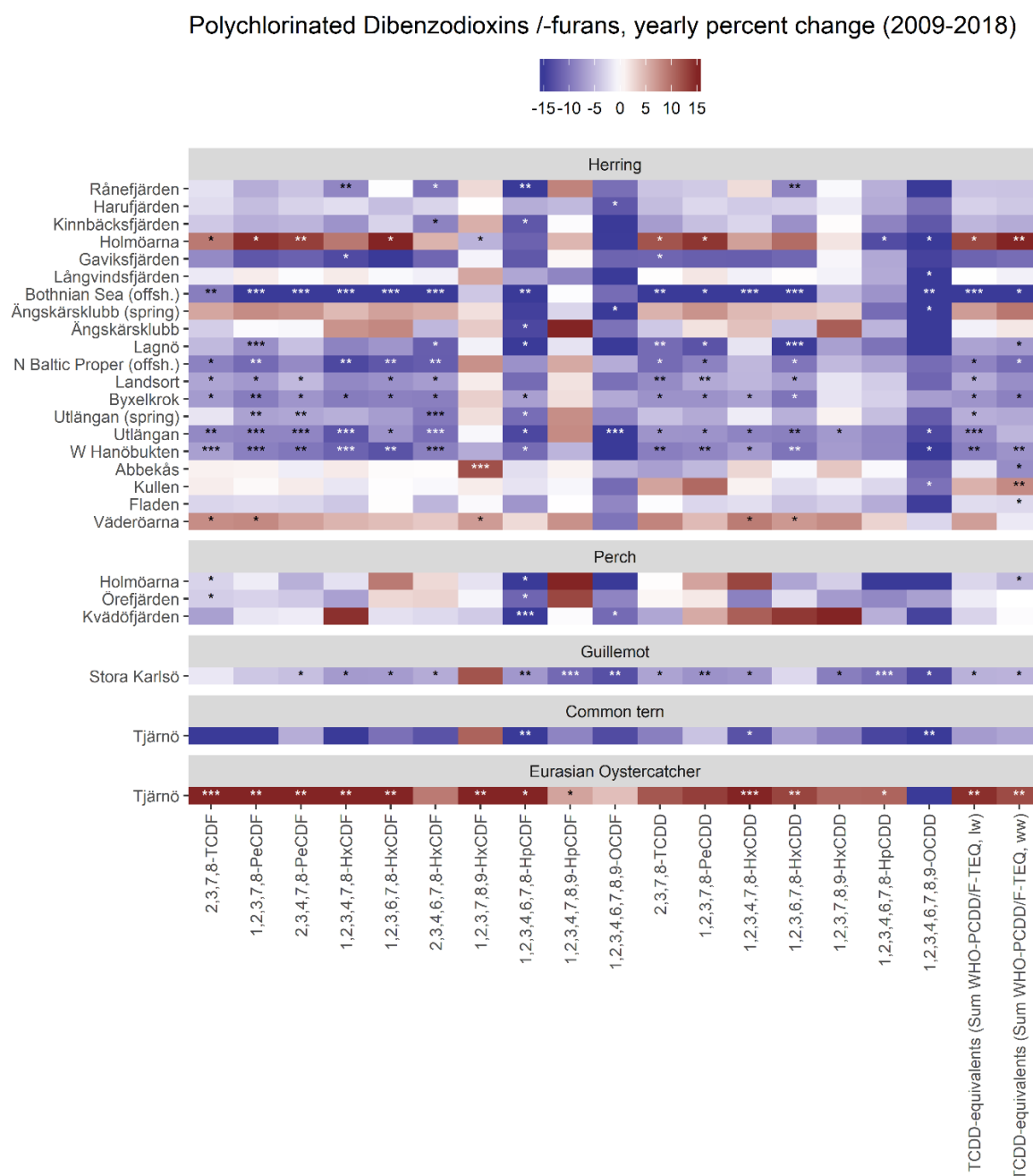


**Figure 11.** The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). * p<0.05, ** p<0.01, *** p<0.001.

The change of dioxin in eggs from the Oystercatcher and Common Tern from the same site (Tjärnö) goes in the opposite directions the last 10 years (Figure 11). The levels in the eggs of Common Tern are decreasing while the levels in Oystercatcher increases. These two species have different diet and their migration pattern differs during winter, which suggest that such differences could be driven by exposure to the contaminants prior to arrival at Tjärnö. Thus, the differences in the trends could potentially be explained by different behaviours of the two species.

In general, the levels in herring are higher in the Bothnian Sea while the lowest concentrations are found on the West Coast (Figure 12). Concentrations are below the threshold value based on food legislation for all fish but above the threshold value set to protect from secondary poisoning in ca 50% of the herring sites (see Table 2 and Appendix).
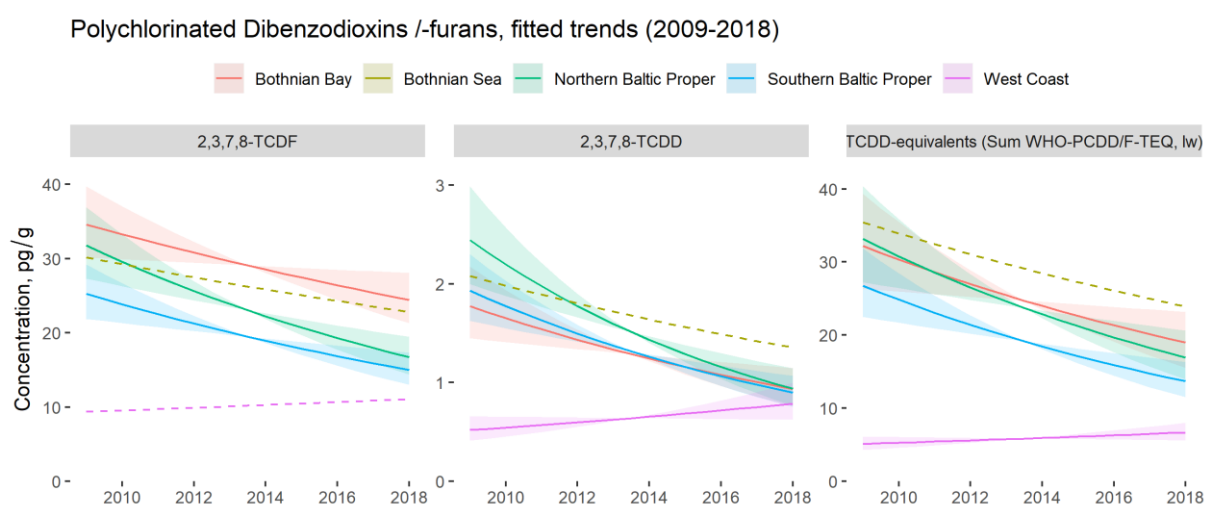


**Figure 12.** Trends at basin level for various dioxins and furans concentrations in herring. A table with detailed results is found in section 7.11. Basins where there is a significantly different trend between sites are drawn with dashed lines. See section 5.2.2 for interpretation of the 95% confidence bands.

## 7.7 Brominated Flame Retardants (BFR)

In general, concentrations of brominated flame retardants decrease among biota, with a few exceptions (Figure 13). In the Bothnian Sea, Holmöarna and Ängskärsklubb (spring) has trends (increasing) that goes in the opposite direction of the other sites in this area for almost all BFRs. This pattern could potentially be driven by the increase in herring age from these sites over the last 10 years (see section 7.1). For the aggregated herring data decreasing trends are seen for all basins for BDE-47 and HBCDD except for BDE-47 on the West Coast where no significant trend is observed (Figure 14). Concentrations of BDE-47 and HBCDD are higher in the Baltic Proper and northern Baltic Sea than on the West Coast but in recent years the difference in concentrations are going down (Figure 14).
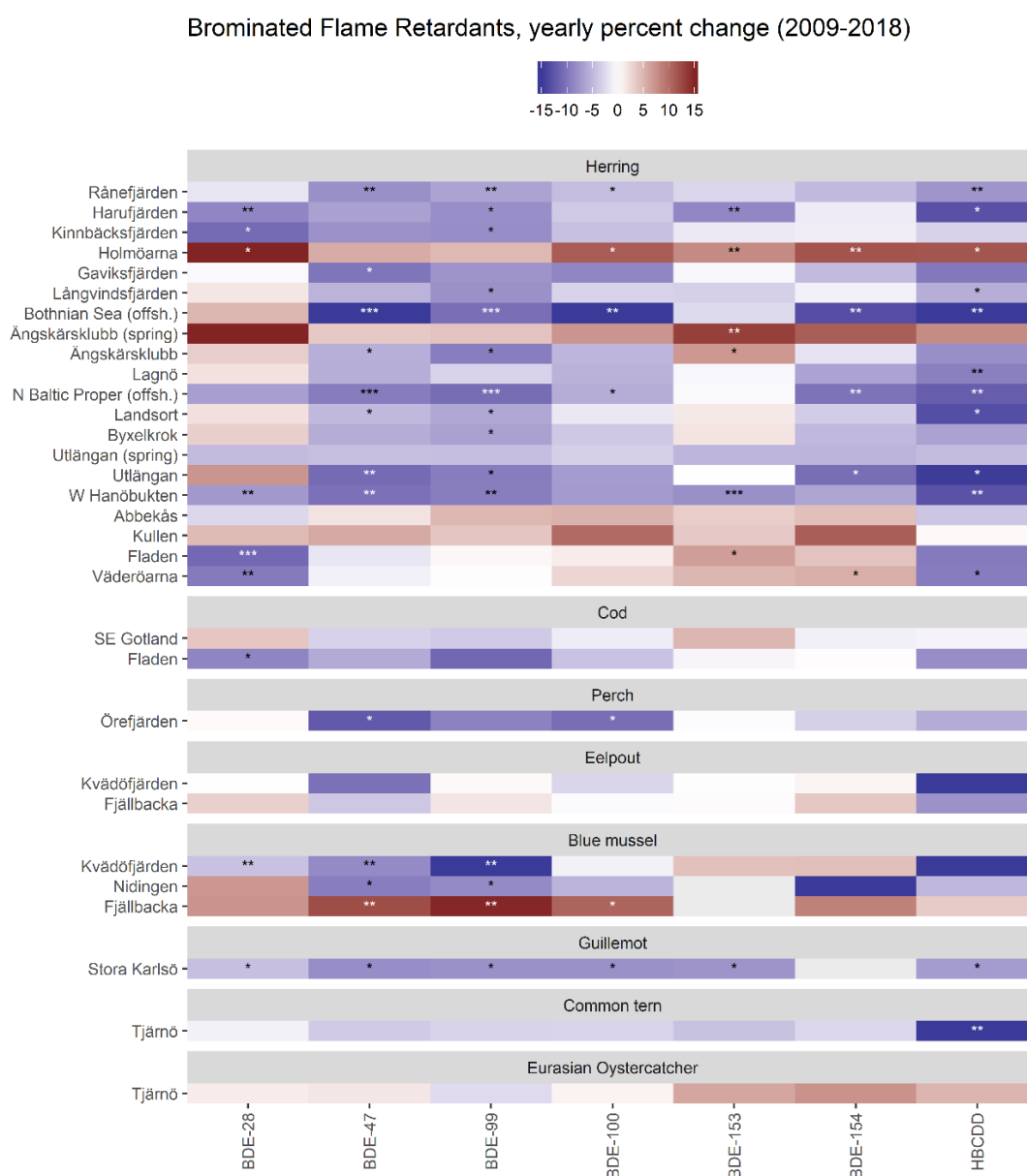


**Figure 13.** The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). * p<0.05, ** p<0.01, *** p<0.001.

BDE-28 and BDE-153 are below LOQ in most sites and for most species and therefore these results must be interpreted with caution (Appendix). Concentration of BDE-47 is above the threshold value at all herring end cod sites, but in perch and eelpout concentrations are just on, or slightly below the threshold (Table 2 and Appendix). Concentration of HBCDD is below the threshold value in fish at all sites (Table 2 and Appendix).
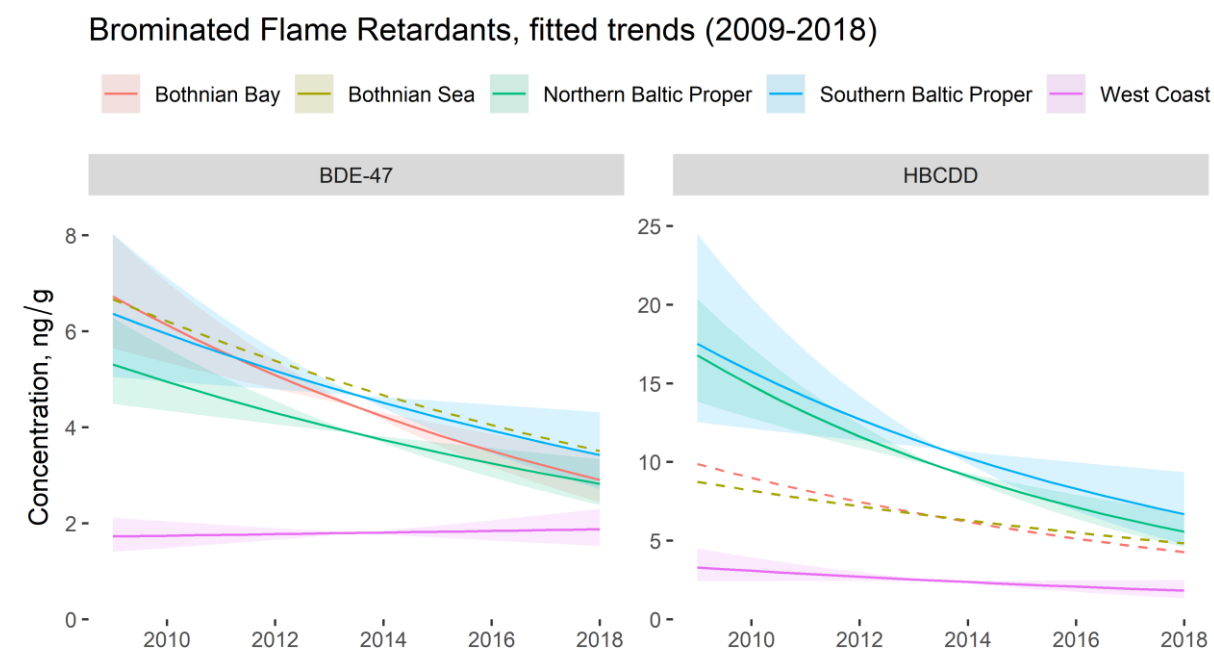


**Figure 14.** Trends at basin level for various brominated flame retardants concentrations in herring. A table with detailed results is found in section 7.11. Basins where there is a significantly different trend between sites are drawn with dashed lines. See section 5.2.2 for interpretation of the 95% confidence bands.

## 7.8  Perfluoroalkyl Substances (PFAS)

In general, perfluoroalkyl substances (PFAS) shows no trends or significantly decreasing trends among all biota during the last ten years (2009-2018). Although, there might be an indication of increasing concentrations in PFOA and PFNA in herring from Baltic Proper and Bothnian Sea and in branched PFOS in the Baltic proper as well as in FOSA on the West Coast (Figure 15). For the aggregated herring data we see that concentrations of PFUnDA and PFOS are lower at the West Coast than the rest of the Baltic Sea. For FOSA on the other hand, concentrations are higher on the West Coast (Figure 16). Concentrations of PFOS in fish are below the threshold value (see Table 2 and Appendix).
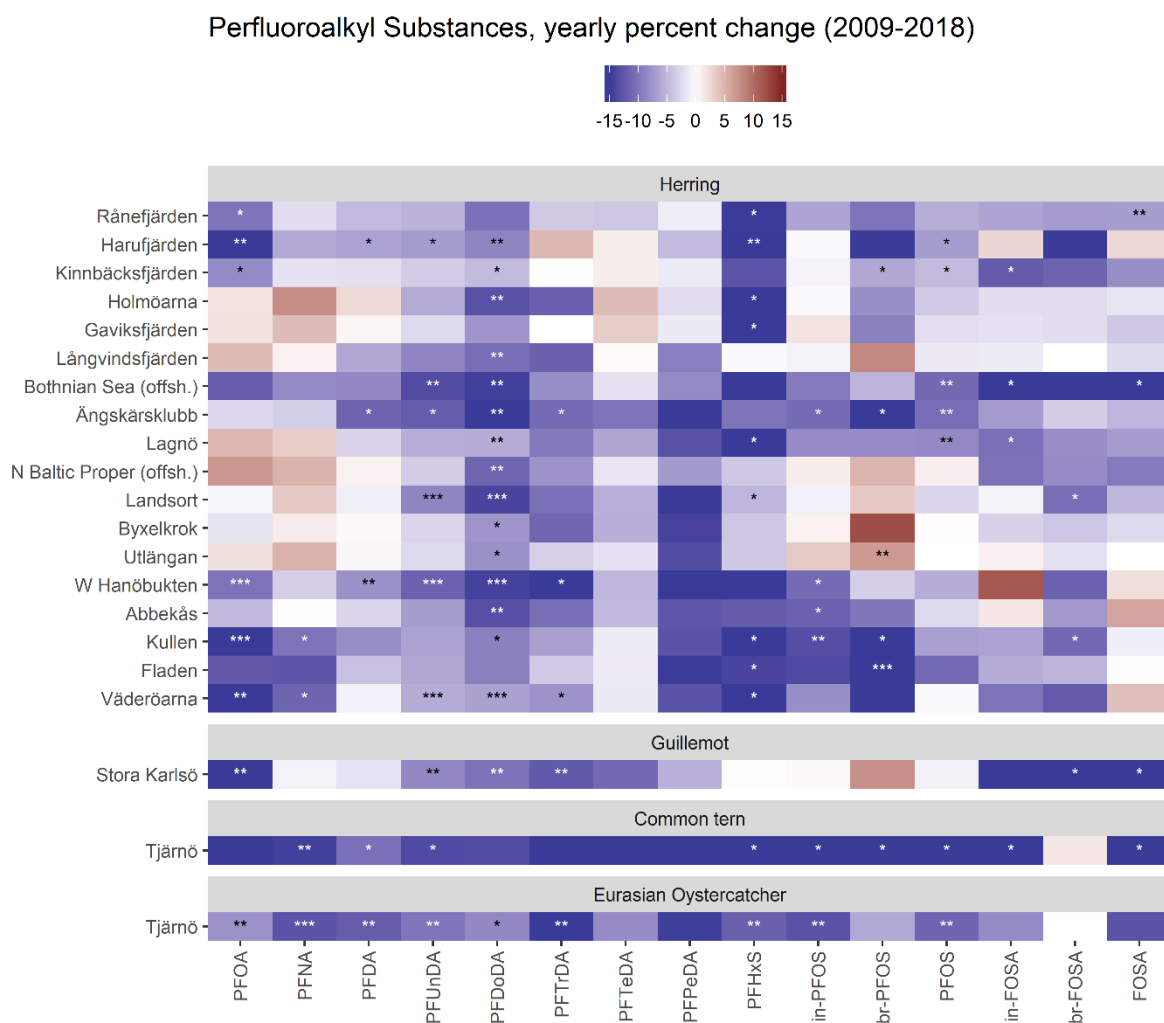


**Figure 15.** The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea), and also divided into species group. * p<0.05, ** p<0.01, *** p<0.001.

Perfluoroalkyl Substances, fitted trends (2009-2018)

Bothnian Bay — Bothnian Sea — Northern Baltic Proper — Southern Baltic Proper — West Coast
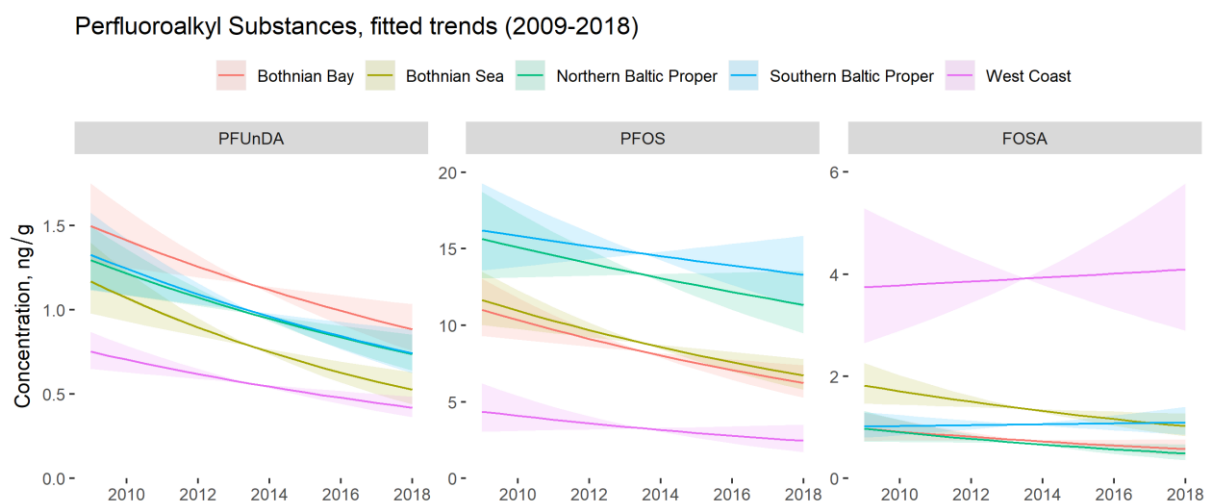


**Figure 16.** Trends at basin level for various PFAS concentrations in herring. A table with detailed results is found in section 7.11. Basins where there is a significantly different trend between sites are drawn with dashed lines. See section 5.2.2 for interpretation of the 95% confidence bands.

## 7.9  Polycyclic Aromatic Hydrocarbons (PAH)

Only blue mussel is analysed for PAH. There is no general pattern for PAH trends among the three sites (Figure 17). At Fjällbacka, on the West Coast, strong upward trends are seen for most of PAH. This increase is mainly caused by high concentrations between 2015 and 2018. At Nidingen, also on the West Coast, concentrations show no significant trends except for Naphtalene (decreasing). Kvädöfjärden, in the Baltic Sea generally shows downward trends for the PAHs. Concentrations of PAH compounds are below their respective threshold value at all sites (see Table 2 and Appendix).

Polycyclic Aromatic Hydrocarbons, yearly percent change (2009-2018)
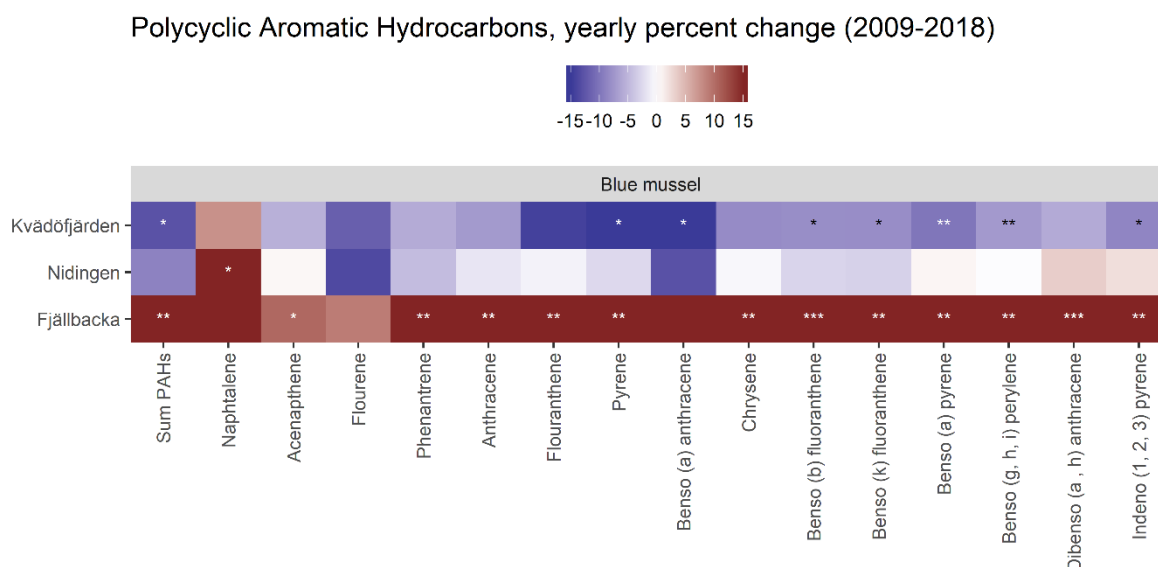


**Figure 17.** The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). * p<0.05, ** p<0.01, *** p<0.001.

## 7.10 Organotin Compounds (OTC)

Most of the organotin compounds are below LOQ during all or almost all years, therefore no heatmap is presented. However, in 2018 mono-, di- and tributyltin and monooctyltin were higher compared to the other 9 years (see Appendix).

## 7.11 Summary table for spatial aggregation

**Table 5. Slopes with 95% c.i. for the herring sites that have been aggregated across basins. Confidence intervals have been omitted for basins where there is a significantly different trend between sites.**

| Contaminant | Southern Baltic Proper | Northern Baltic Proper | Bothnian Sea | West Coast | Bothnian Bay |
|---|---|---|---|---|---|
| Cd | 1.39 (-2.54, 5.47) | 0.4 (-2.82, 3.73) | -4.2 (-8.24, 0.03) | 3.06 (-1.13, 7.44) | -3.11 (-7.02, 0.97) |
| Cu | 1.56 (-0.53, 3.7) | 2.35 (0.2, 4.54) | -0.17 (-2.86, 2.6) | -0.05 (-3.42, 3.43) | 2.1 (-0.84, 5.13) |
| Hg | 0.34 (-3.21, 4.03) | 0.1 (-2.92, 3.23) | -1.23 (-4.37, 2.02) | 2.03 (-0.35, 4.47) | -3.81 (-6.99, -0.52) |
| Pb | 1.82 (-1.2, 4.92) | -0.34 (-3.45, 2.86) | -0.26 (-4.99, 4.71) | -3.76 (-7.57, 0.2) | -4.81 (-10.23, 0.94) |
| 2,3,7,8-TCDD | -8.16 (-11.63, -4.56) | -10.1 (-14.02, -5.99) | -4.63 | 4.71 (-0.6, 10.3) | -6.86 (-10.96, -2.56) |
| TCDD-eqv (ΣWHO-PCDD/F-TEQ, lw) | -7.17 (-10.7, -3.5) | -7.18 (-11.13, -3.05) | -4.28 | 3.01 (-0.95, 7.13) | -5.7 (-9.8, -1.41) |
| 2,3,7,8-TCDF | -5.61 (-8.59, -2.54) | -6.87 (-9.94, -3.7) | -3.04 | 1.88 | -3.78 (-6.69, -0.79) |
| PCB-118 | -5.66 (-9.16, -2.02) | -6.81 (-10.41, -3.07) | -4.89 | 2.3 (-1.26, 5.99) | -5.92 (-9.88, -1.78) |
| PCB-153 | -3.58 (-7.16, 0.13) | -4.36 (-8.32, -0.23) | -4.91 | 3.28 | -5.5 (-9.54, -1.28) |
| DDE | -4.95 (-9.34, -0.35) | -6.11 (-9.98, -2.08) | -6.25 | 2.54 | -7.5 (-11.78, -3.01) |
| HCB | -2.91 (-6.72, 1.05) | -3.33 (-5.79, -0.81) | -0.49 | 5.1 (2.48, 7.78) | -1.72 (-4.6, 1.24) |
| BDE-47 | -6.66 (-11.34, -1.72) | -6.78 (-10.19, -3.24) | -6.88 | 0.92 (-3.54, 5.59) | -8.9 (-12.37, -5.29) |
| HBCDD | -10.14 (-16.59, -3.19) | -11.54 (-15.25, -7.67) | -6.39 | -6.38 (-12.62, 0.31) | -8.91 |
| FOSA | 0.81 (-4.52, 6.43) | -7.35 (-13.44, -0.84) | -6.2 (-10.59, -1.59) | 0.98 (-6.47, 9.03) | -5.73 (-11.44, 0.34) |
| PFOS | -2.15 (-5.86, 1.71) | -3.51 (-7.28, 0.42) | -5.92 (-8.98, -2.76) | -6.18 (-13.28, 1.5) | -6.13 (-9.6, -2.52) |
| PFUnDA | -6.26 (-9.77, -2.6) | -6.05 (-9.01, -3) | -8.51 (-12.06, -4.81) | -6.26 (-9.2, -3.22) | -5.68 (-8.89, -2.36) |

# 8 References

Andrea, C. (2005), Breaking tolerance to nickel, *Toxicology*, *209*(2), 119-121.

Apelberg, B. J., F. R. Witter, J. B. Herbstman, A. M. Calafat, R. U. Halden, L. L. Needham, and L. R. Goldman (2007), Cord serum concentrations of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in relation to weight and size at birth, *Environmental Health Perspectives*, *115*(11), 1670-1676.

ATSDR (2000), Toxicological Profile for Polychlorinated Biphenyls (PCBs), Agency for Toxic Substances and Disease Registry.

ATSDR (2005), Toxicological Profile for Zinc, 352 pp, Agency for Toxic Substances and Disease Registry.

Aulerich, R. J., and R. K. Ringer (1977), Current Status of PCB Toxicity to Mink, and Effect on Their Reproduction, *Archives of Environmental Contamination and Toxicology*, *6*(2-3), 279-292.

Austin, O. L. (1953), The migration of the Common Tern (Sterna hirundo) in the Western Hemisphere, *Bird-Banding*, 39-55.

BDE EQS dossier (2011), POLYBROMINATED DIPHENYL ETHERS (BDES) EQS dossiers: The European Commission, Available at: https://circabc.europa.eu/sd/d/d07ed9f5-0760-4561-b642-04bc1e4a580e/PBDE%20EQS%20dossier%202011.pdf.

Becker, P., and J. Ludwigs (2004), Common tern Sterna hirundo, *BWP Update*, *6*(1-2), 91-137.

Becker, P., D. Frank, and S. Sudmann (1993), Temporal and Spatial Pattern of Common Tern (Sterna-Hirundo) Foraging in the Wadden Sea, *Oecologia*, *93*(3), 389-393.

Berger, U., A. Glynn, K. E. Holmström, M. Berglund, E. H. Ankarberg, and A. Törnkvist (2009), Fish consumption as a source of human exposure to perfluorinated alkyl substances in Sweden - Analysis of edible fish from Lake Vattern and the Baltic Sea, *Chemosphere*, *76*(6), 799-804.

Bignert, A., E. Greyerz, E. Nyberg, K. Sundqvist, and K. Wiberg (2005), Geografisk variation i koncentrationer av dioxiner och PCB i strömming från Bottniska viken och norra egentliga Östersjön, Länsstyrelsen.

Bignert, A., U. Eriksson, E. Nyberg, A. Miller, and S. Danielsson (2014), Consequences of using pooled versus individual samples for designing environmental monitoring sampling strategies, *Chemosphere*, *94*, 177-182.

Bignert, A., S. Danielsson, C. Ek, S. Faxneld, and E. Nyberg (2017), Comments Concerning the National Swedish Contaminant Monitoring Programme in Marine Biota, 2017 (2016 years data), 341 pp, Swedish Museum of Natural History, Stockholm, Sweden.

Bignert, A., K. Litzen, T. Odsjö, M. Olsson, W. Persson, and L. Reutergardh (1995), Time-Related Factors Influence the Concentrations of Sddt, Pcbs and Shell Parameters in Eggs of Baltic Guillemot (Uria-Aalge), 1861-1989, *Environmental Pollution*, *89*(1), 27-36.

Bignert, A., A. Göthberg, S. Jensen, K. Litzén, T. Odsjö, M. Olsson, and L. Reutergårdh (1993), The Need for Adequate Biological Sampling in Ecotoxicological Investigations - a Retrospective Study of 20 Years Pollution Monitoring, *Science of the Total Environment*, *128*(2-3), 121-139.

Bleavins, M. R., R. J. Aulerich, and R. K. Ringer (1980), Polychlorinated biphenyls (Aroclors 1016 and 1242): effects on survival and reproduction in mink and ferrets, *Arch Environ Contam Toxicol*, *9*(5), 627-635.

Buck, R. C., J. Franklin, U. Berger, J. M. Conder, I. T. Cousins, P. de Voogt, A. A. Jensen, K. Kannan, S. A. Mabury, and S. P. van Leeuwen (2011), Perfluoroalkyl and polyfluoroalkyl substances in the environment: terminology, classification, and origins, *Integr Environ Assess Manag*, *7*(4), 513-541.

Cardinale, M., and J. Modin (1999), Changes in size-at-maturity of Baltic cod (Gadus morhua) during a period of large variations in stock size and environmental conditions, *Fisheries research*, *41*(3), 285-295.

Carpenter, D. O. (1998), Polychlorinated biphenyls and human health, *Int J Occup Med Environ Health*, *11*(4), 291-303.

Carpenter, D. O. (2006), Polychlorinated biphenyls (PCBs): routes of exposure and effects on human health, *Rev Environ Health*, *21*(1), 1-23.

Cempel, M., and G. Nikel (2006), Nickel: a review of its sources and environmental toxicology, *Polish J. of Environ. Stud*, *15*(3), 375-382.

CIRCABC CIRCABC webpage, edited, The European Commision.

Cook, R. S., and D. O. Trainer (1966), Experimental lead poisoning of Canada Geese, *Journal of Wildlife Management*, *30*(1), 1-&.

Danielsson, C., K. Wiberg, P. Korytar, S. Bergek, U. A. T. Brinkman, and P. Haglund (2005), Trace analysis of polychlorinated dibenzo-p-dioxins, dibenzofurans and WHO polychlorinated biphenyls in food using comprehensive two-dimensional gas chromatography with electron-capture detection, *Journal of Chromatography A*, *1086*(1-2), 61-70.

Darnerud, P. O. (2008), Brominated flame retardants as possible endocrine disrupters, *International Journal of Andrology*, *31*(2), 152-160.

Denkhaus, E., and K. Salnikow (2002), Nickel essentiality, toxicity, and carcinogenicity, *Critical Reviews in Oncology/Hematology*, *42*(1), 35-56.

Doioxin EQS dossier (2011), POLYCHLORINATED DIBENZO-P-DIOXINS (PCDDS), POLYCHLORINATED DIBENZOFURANS (PCDFS), AND DIOXIN-LIKE POLYCHLORINATED BIPHENYLS (DL-PCBS) EQS dossiers: The European Commission, Available at: https://circabc.europa.eu/d/a/workspace/SpacesStore/f0d90906-c361-4af1-82b1-d2e52f826c14/Dioxins%20%26%20PCB-DL%20EQS%20dossier%202011.pdf.

Dorsey, A., L. Ingerman, and S. Swarts (2004), Toxicological profile for copper, edited, p. 314 pp, U.S. Dept of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry.

Eisler, R. (1994), A review of arsenic hazards to plants and animals with emphasis on fishery and wildlife resources, in *In: Arsenic in the environment, Part 2: Human Health and Ecosystem Effects*, edited by e. Nriagu JO, pp. 184-261, John Wiley & Sons, Inc.

Eisler, R. (1996), Silver hazards to fish, wildlife and invertebrates: A synoptic review. Contaminant Hazards Review Report 32 edited, p. 53, National Biological Service, U.S. Department of the Interior.

Encinar, J. R., M. I. M. Villar, V. G. Santamaria, J. I. G. Alonso, and A. Sanz-Medel (2001), Simultaneous determination of mono-, di-, and tributyltin in sediments by isotope dilution analysis using gas chromatograpby-ICPMS, *Analytical Chemistry*, *73*(13), 3174-3180.

Eriksson, P., C. Fischer, and A. Fredriksson (2006a), Polybrominated diphenyl ethers, a group of brominated flame retardants, can interact with polychlorinated biphenyls in enhancing developmental neurobehavioral defects, *Toxicological Sciences*, *94*(2), 302-309.

Eriksson, P., C. Fischer, M. Wallin, E. Jakobsson, and A. Fredriksson (2006b), Impaired behaviour, learning and memory, in adult mice neonatally exposed to hexabromocyclododecane (HBCDD), *Environmental Toxicology and Pharmacology*, *21*(3), 317-322.

Eriksson, U., A. Johansson, K. Litzén, L. Häggberg, A. Winberg, and S. Zakrisson (1994), Analysmetod för bestämning av klorerade organiska miljögifter i biologiskt material. ITM rapport 18.

EU (2005a), Substance Data Sheet: Hexachlorbenzenes (incl. Limdane), Priority Substance No. 18, CAS-No. 608-73-1 and 58-89-9. Environmental Cuality Standards (EQS). Common Implementation Strategy for the Water Framework Directive. Final version 15 July 2005, Brussels, 22 pp.

EU (2005b), Substance Data Sheet: Cadmium and its compounds, Priority Substance No. 6, CAS-No. 74440-43-9. Environmental Cuality Standards (EQS). Common Implementation Strategy for the Water Framework Directive. Final version 31 July 2005, Brussels, 37 pp.

EU (2005c), Substance Data Sheet: Mercury and its compounds, Priority Substance No. 21, CAS-No. 74439-97-6. Environmental Cuality Standards (EQS). Common Implementation Strategy for the Water Framework Directive. Final version 15 January 2005. Available at: https://circabc.europa.eu/sd/a/ff8e163c-71f6-4fc0-98ef-875a20add4c8/21_Mercury_EQSdatasheet_150105.pdf, Brussels, 19 pp.

Fei, C. Y., J. K. McLaughlin, L. Lipworth, and J. Olsen (2008), Prenatal exposure to perfluorooctanoate (PFOA) and perfluorooctanesulfonate (PFOS) and maternally reported developmental milestones in infancy, *Environmental Health Perspectives*, *116*(10), 1391-1395.

Fernie, K. J., J. L. Shutt, R. J. Letcher, I. J. Ritchie, and D. M. Bird (2009), Environmentally relevant concentrations of DE-71 and HBCD alter eggshell thickness and reproductive success of American kestrels, *Environmental Science & Technology*, *43*(6), 2124-2130.

Fluoranthene EQS dossier (2011), EQS dossiers: The European Commission, Available at: https://circabc.europa.eu/sd/d/4336e1e5-ba0c-4545-abee-7743d2085bc3/Fluoranthene%20EQS%20dossier%202011.pdf.

France, R. L. (1995), C-13 Enrichment in Benthic Compared to Planktonic Algae - Foodweb Implications, *Marine Ecology Progress Series*, *124*(1-3), 307-312.

Garnaga, G., E. Wyse, S. Azemard, A. Stankevicius, and S. de Mora (2006), Arsenic in sediments from the southeastern Baltic Sea, *Environ Pollut*, *144*(3), 855-861.

Gidlow, D. A. (2004), Lead toxicity, *Occup Med (Lond)*, *54*(2), 76-81.

Grimås, U., A. Göthberg, M. Notter, M. Olsson, and L. Reutergårdh (1985), Fat Amount - a Factor to Consider in Monitoring Studies of Heavy-Metals in Cod Liver, *Ambio*, *14*(3), 175-178.

Hagenaars, A., D. Knapen, I. J. Meyer, K. van der Ven, P. Hoff, and W. De Coen (2008), Toxicity evaluation of perfluorooctane sulfonate (PFOS) in the liver of common carp (Cyprinus carpio), *Aquatic Toxicology*, *88*(3), 155-163.

Hamlin, H. J., and L. J. Guillette Jr (2010), Wildlife as sentinels of environmental impacts on reproductive health and fertility, in *Environmental impacts on reproductive health and fertility*, edited by Woodruff TJ, Janssen S, Guillette LJ and Giudice LC, pp. 103-110, Cambridge University Press, New York, USA.

HBCDD EQS dossier (2011), HEXABROMOCYCLODODECANE EQS dossier: The European Commission, Available at: https://circabc.europa.eu/sd/a/086ffe7c-8e63-4893-baac-994f3ff0eb34/HBCDD%20EQS%20dossier%202011.pdf.

Helander, B., A. Bignert, and L. Asplund (2008), Using raptors as environmental sentinels: Monitoring the white-tailed sea eagle Haliaeetus albicilla in Sweden, *Ambio*, *37*(6), 425-431.

HELCOM (2010), Hazardous substances in the Baltic Sea – An integrated thematic assessment of hazardous substances in the Baltic Sea. , in *Balt. Sea Environ. Proc. No. 120B.*, edited, p. 119, Baltic Marine Environment Protection Commission, Helsinki, Finland.

Horwitz, W., and R. Albert (2006), The Horwitz ratio (HorRat): A useful index of method performance with respect to precision, *Journal of Aoac International*, *89*(4), 1095-1109.

Huber, K. (1997), Wisconsin Mercury SourceBook, Section 1, edited, Wisconsin Department of Natural Resources, Madison, Wisconsin, USA.

Ikemoto, T., T. Kunito, H. Tanaka, N. Baba, N. Miyazaki, and S. Tanabe (2004), Detoxification mechanism of heavy metals in marine mammals and seabirds: interaction of selenium with mercury, silver, copper, zinc, and cadmium in liver, *Archives of Environmental Contamination and Toxicology*, *47*(3), 402-413.

IPCS (2002), Silver and silver compounds: environmental aspects - Concise International Chemical Assesment Document 44, International Panel on Chemical Safety, Geneva, Switzerland.

IVL (2007), Results from the Swedish National Screening Programme 2007. Subreport 5: Silver, report B1826, 54 pp, Swedish Environmental Research Institute Ltd.

Jacobsson, A., E. Neuman, and G. Thoresson (1986), The Viviparous Blenny as an Indicator of Environmental-Effects of Harmful Substances, *Ambio*, *15*(4), 236-238.

Jensen, S., L. Renberg, and L. Reutergårdh (1977), Residue Analysis of Sediment and Sewage Sludge for Organochlorines in Presence of Elemental Sulfur, *Analytical Chemistry*, *49*(2), 316-318.

Jensen, S., L. Reutergårdh, and B. Jansson (1983), Analytical methods for measuring organochlorines and methyl mercury by gas chromatography, edited, pp. 21-33, FAO Fisheries Technical Paper No 212.

Kasprzak, K. S., F. W. Sunderman, and K. Salnikow (2003), Nickel carcinogenesis, *Mutation Research/Fundamental and Molecular Mechanisms of Mutagenesis*, *533*(1), 67-97.

KEMI (2014), Giftfri miljö, edited, Swedish Chemicals Agency.

Klaassen, C. D., and K. Rozman (1991), Absorption, Distribution and Exretion of Toxicants, in *Casarett and Doull´s Toxicology - The Basic Science of Poisons 4th edition*, edited by Amdur MO., Doull J. and Klaassen CD., pp. 50-87, Pergamon Press, New York, USA.

Koffijberg, K., L. Dijksen, B. Hälterlein, K. Laursen, P. Potel, and P. Südbeck (2006), Breeding Birds in the Wadden Sea in 2001 - Results of the total survey in 2001 and trends in numbers between 1991-2001. Wadden Sea Ecosystem No. 22., 136 pp, Wilhelmshaven, Germany.

Lampa, E. (2019), MoCiS: Monitoring of Contaminants in Sweden. R package version 1.0.

Larsen, B., and J. Riego (1990), Interference from 2,3,5,6-3',4'-Hexachlorobiphenyl (CB-163) in the Determination of 2,3,4-2',4',5'-Hexachlorobiphenyl (CB-138) in Environmental and Technical Samples, *International Journal of Environmental Analytical Chemistry*, *40*(1-4), 59-68.

Legler, J. (2008), New insights into the endocrine disrupting effects of brominated flame retardants, *Chemosphere*, *73*(2), 216-222.

Lemly, A. D. (2004), Aquatic selenium pollution is a global environmental safety issue, *Ecotoxicology and Environmental safety*, *59*(1), 44-56.

Lemmetyinen, R. (1973), Feeding ecology of Sterna paradisaea Pontopp. and S. hirundo L. in the archipelago of southwestern Finland, paper presented at Annales Zoologici Fennici, JSTOR.

Li, Y. F., and R. W. Macdonald (2005), Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review, *Science of the Total Environment*, *342*(1-3), 87-106.

Lilja, K., H. Andersson, A. Woldegiorgis, A. Jönsson, A. Palm-Cousins, K. Hansson, and E. Brorstöm-Lunden (2010), Bedömning av miljögiftspåverkan i vattenmiljö – samordnad metodutveckling (IVL Rapport B1891), IVL, Sweden. Available at: https://www.ivl.se/download/18.343dc99d14e8bb0f58b75a8/1445517424282/B1891.pdf.

Liljelind, P., G. Söderström, B. Hedman, S. Karlsson, L. Lundin, and S. Marklund (2003), Method for multiresidue determination of halogenated aromatics and PAHs in combustion-related samples, *Environmental Science & Technology*, *37*(16), 3680-3686.

Newman, M., and M. Unger (2003), *Fundamentals of ecotoxicology.*, 458 pp., Lewis publishers, CRC press LCC, USA.

Nisbet, I. C. T., and P. K. Lagoy (1992), Toxic Equivalency Factors (Tefs) for Polycyclic Aromatic-Hydrocarbons (Pahs), *Regulatory Toxicology and Pharmacology*, *16*(3), 290-300.

Nissling, A. (1995), Salinity and oxygen requirements for successful spawning of Baltic cod, Gadus morhua, Diss (sammanfattning) Stockholm Univ thesis, 4 , 24 s. pp, Univ.,, Stockholm.

NMR (1995), *Nordic Enviromental Specimen Banking - Methods in Use in ESB: Manual for the Nordic Countries (TemaNord 1995:543)*, The Nordic Council of Ministers, Copenhagen, Denmark.

OJEC No. L164 25.6.2008 (2008), DIRECTIVE 2008/56/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 17 June 2008 establishing a framework for community action in the field of marine environmental policy (Marine Strategy Framework Directive), edited, Official Journal of the European Union.

OJEC No. L226 24.8.2013 (2013), DIRECTIVE 2013/39/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy, edited, Official Journal of the European Union.

OJEC No. L348 24.12.2008 (2008), DIRECTIVE 2008/105/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC of the European parliament and of the Council, edited, Official Journal of the European Union.

OJEC No. L364 20.12.2006 (2006), COMMISSION REGULATION (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs, edited, Official Journal of the European Union.

OJEC No. L396 30.12.2006 (2006), REGULATION (EC) No 1907/2006 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), establishing a European Chemicals Agency, amending Directive 1999/45/EC and repealing Council Regulation (EEC) No 793/93 and Commission Regulation (EC) No 1488/94 as well as Council Directive 76/769/EEC and Commission Directives 91/155/EEC, 93/67/EEC, 93/105/EC and 2000/21/EC, edited, p. 849, Official Journal of the European Union.

OJEC No. L 174 1.7.2011 (2011), DIRECTIVE 2011/65/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 8 June 2011 on the restriction of the use of certain hazardous substances in electrical and electronic equipment, edited, p. 23, Official Journal of the European Union.

OSPAR (2020), OSPAR 2020 assessment (Draft) based on data extracted on 10th October 2019 (4th November for fish disease), edited, Available at: https://ocean.ices.dk/oat/?assessmentperiod=2020 (accessed January 2020 - web platform).

OSPAR Commision (1998), Report of the Third OSPAR Workshop on Ecotoxicological Assessment Criteria, The Hague: 25-29 November 1996, Hazardous Substances Series, Available at: https://www.ospar.org/documents?d=6878.

OSPAR Commision (2005), Overview of Past Dumping at Sea of Chemical Weapons and Munitions in the OSPAR Maritime Area, 13 pp, The Convention for the Protection of the marine Environment of the North-East Atlantic.

OSPAR Commision (2014), Levels and trends in marine contaminants and their biological effects – CEMP Assessment Report 2013, edited.

PAH EQS dossier (2011), 5-6 RINGS POLYAROMATIC HYDROCARBONS (PAH). EQS dossiers: The European Commission, Available at: https://circabc.europa.eu/sd/d/4e13a4c4-07b9-4e55-a43d-823e7cd4ce82/PAH%2520EQS%2520dossier%25202011.pdf.

Peterson, B. J., and B. Fry (1987), Stable Isotopes in Ecosystem Studies, *Annual Review of Ecology and Systematics*, *18*, 293-320.

Petry, T., P. Schmid, and C. Schlatter (1996), The use of toxic equivalency factors in assessing occupational and environmental health risk associated with exposure to airborne mixtures of polycyclic aromatic hydrocarbons (PAHs), *Chemosphere*, *32*(4), 639-648.

PFOS EQS dossier (2011), PERFLUOROOCTANE SULPHONATE (PFOS) EQS dossiers: The European Commission, Available at: https://circabc.europa.eu/sd/a/027ff47c-038b-4929-a84c-da3359acecee/PFOS%20EQS%20dossier%202011.pdf.

Post, D. M. (2002), Using stable isotopes to estimate trophic position: Models, methods, and assumptions, *Ecology*, *83*(3), 703-718.

Powley, C. R., S. W. George, T. W. Ryan, and R. C. Buck (2005), Matrix effect-free analytical methods for determination of perfluorinated carboxylic acids in environmental matrixes, *Analytical Chemistry*, *77*(19), 6353-6358.

R Core Team (2019), *R: A language and environment for statistical computing. R Foundation for Statistical Computing*, Vienna, Austria (https://www.R-project.org/).

Reindl, A. R., and L. Falkowska (2019), Food source as a factor determining birds' exposure to hazardous organic pollutants and egg contamination, *Marine and Freshwater Research*.

SC (2019), The Stockholm Convention Webpage, edited, The Secretariat of the Stockholm Convention, Switzerland.

Sellström, U., A. Kierkegaard, C. de Wit, and B. Jansson (1998), Polybrominated diphenyl ethers and hexabromocyclododecane in sediment and fish from a Swedish river, *Environmental Toxicology and Chemistry*, *17*(6), 1065-1072.

SGU (2005), Mineralmarknaden. Tema: Arsenik., Geological Survey of Sweden, Uppsala.

Smith, B. S. (1981), Tributyltin compounds induce male characteristics on female mud snails Nassarius obsoletus = Ilyanassa obsoleta, *J Appl Toxicol*, *1*(3), 141-144.

SMNH (2012), Manual for collection, preparation and storage of fish., edited, Swedish Museum of Natural History.

Soclo, H. H., P. Garrigues, and M. Ewald (2000), Origin of polycyclic aromatic hydrocarbons (PAHs) in coastal marine sediments: Case studies in Cotonou (Benin) and Aquitaine (France) areas, *Marine Pollution Bulletin*, *40*(5), 387-396.

Sternberg, R. M., M. P. Gooding, A. K. Hotchkiss, and G. A. LeBlanc (2010), Environmental-endocrine control of reproductive maturation in gastropods: implications for the mechanism of tributyltin-induced imposex in prosobranchs, *Ecotoxicology*, *19*(1), 4-23.

Suzuki, T., N. Imura, and T. W. Clarkson (1991), Overview, in *Advances in mercury toxicology*, edited by Suzuki T., Imura N. and Clarkson TW., p. 32, Plenum Press, New York, USA.

SWE-EPA (2008), Övervakning av prioriterade miljöfarliga ämnen listade i Ramdirektivet för vatten. Rapport 5801, Swedish Environmental Protection Agency.

The European Commission (2000), The EU Water Framework Directive - integrated river basin management for Europe, edited, pp. 1-73, The European Commission.

UNEP, and WHO (2013), *State of the science of endocrine disrupting chemicals - 2012*, World Health Organization / United Nations Environment Programme, Geneva, Switzerland.

United Nations Environment Programme, International Labour Organization, World Health Organization, and International Program on Chemical Safety (1981), *Arsenic*, 174 p. pp., World Health Organization,, Geneva, Switzerland & Albany, NY.,USA.

Vestergren, R., and I. T. Cousins (2009), Tracking the Pathways of Human Exposure to Perfluorocarboxylates, *Environmental Science & Technology*, *43*(15), 5565-5575.

Viberg, H. (2004), *Neonatal developmental neurotoxicity of brominated flame retardants, the polybrominated diphenyl ethers (PBDEs)*, 62 s. pp., Acta Universitatis Upsaliensis : Univ.-bibl. [distributör], Uppsala.

Vibert, R., and K. F. Lagler (1961), *Peches continentales, biologie et amenagement*, 1 ed., 720 pp., Dunod, Paris, France.

Voss, M., K. C. Emeis, S. Hille, T. Neumann, and J. Dippner (2005), Nitrogen cycle of the Baltic Sea from an isotopic perspective, *Global biogeochemical cycles*, *19*(3).

Walker, C. H., S. P. Hopkin, R. M. Sibly, and D. B. Peakall (2001), *Principles of ecotoxicology*, 309 pp., Taylor & Francis Inc., New York , NY, USA.

Wendeln, H., and P. Becker (1996), Body mass change in breeding Common Terns Sterna hirundo, *Bird Study*, *43*(1), 85-95.

WHO (1991), Environmental health criteria 108. Nickel, Geneva.

Wiberg, K., M. Oehme, P. Haglund, H. Karlsson, M. Olsson, and C. Rappe (1998), Enantioselective analysis of organochlorine pesticides in herring and seal from the Swedish marine environment, *Marine Pollution Bulletin*, *36*(5), 345-353.

Zeileis, A. (2004), Econometric computing with HC and HAC covariance matrix estimators.