



**The Swedish National Monitoring Programme for
Contaminants in marine biota (until 2017 year's data)
– Temporal trends and spatial variations**

Det svenska nationella miljöövervakningsprogrammet för
miljögifter i marin biota (fram till 2017 års data)
– Temporal- och spatial variation

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Överenskommelse: 2213-16-003

Report nr 2:2019

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2019-04-25

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Please cite as:

Danielsson, S., Ek, C., Faxneld, S., Winkens Pütz K. 2019. The Swedish National Monitoring Programme for Contaminants in marine biota (until 2017 year's data) – Temporal trends and spatial variations. 2:2019, Swedish Museum of Natural History, Stockholm, Sweden



SWEDISH ENVIRONMENTAL
PROTECTION AGENCY

NATIONAL
ENVIRONMENTAL
MONITORING
COMMISSIONED BY
THE SWEDISH EPA

FILE NO. NV-02950-16
CONTRACT NO. 2213-16-003
PROGRAMME AREA Miljögifter akvatiska
SUBPROGRAMME Metaller och
organiska
miljögifter

The Swedish National Monitoring Programme for Contaminants in marine biota (until 2017 year's data) – Temporal trends and spatial variations

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<p>Report title and subtitle Det svenska nationella miljöövervakningsprogrammet för miljögifter i marin biota (fram till 2017 års data) - Temporal- och spatial variation</p> <p>The Swedish National Monitoring Programme for Contaminants in marine biota (until 2017 year's data) – Temporal trends and spatial variations</p>	<p>Purchaser Swedish Environmental Protection Agency, Environmental Monitoring Unit SE-106 48 Stockholm, Sweden</p> <p>Funding National environmental monitoring</p>
<p>Keywords for location (specify in Swedish) Östersjön, Västkusten, Bottenviken, Bottenhavet, Egentliga Östersjön, Skagerrak, Kattegatt, Rånefjärden, Harufjärden, Kinnbäcksfjärden, Holmöarna, Örefjärden, Gaviksfjärden, Långvindsfjärden, Ångskärsklubb, Lagnö, Landsort, Kvädöfjärden, Byxelkrok, St.Karlsö, SE Gotland, Utlängan, Hanöbukten, Abbekås, Kullen, Fladen, Nidingen, Väderöarna, Fjällbacka, Tjärnö, Ålands hav, Bornholmsbassängen</p>	
<p>Keywords for subject (specify in Swedish) Miljögifter, tidstrender, spatiala trender, DDT, PCB, HCH, HCB, dioxiner, furaner, metaller, Pb, Cd, Cu, Zn, Cr, Ni, Ag, As, Se, PBDE, HBCDD, PFAS, PFOS, biota, PAH, tennorganiska föreningar, fisk, strömning, sill, abborre, tånglake, torsk, blåmussla, sillgrissla, strandskata, fisktärna, ägg</p>	
<p>Period in which underlying data were collected 1968–2017</p>	
<p>Summary The report summarises the monitoring activities within the National Swedish Contaminant Programme in marine biota. In this report, we present time series and maps of most contaminants in various species but we only comment results that shows large differences compared to the previous year. Results show that herring from Holmöarna had elevated concentrations during the last two years of the time series for approximately half of the analysed metals, most chlorinated pesticides, polychlorinated biphenyls (PCBs) and brominated flame retardants (BFRs). The last two years' Herring is on average one year older than the previous years', heavier and the last years' fat percentage is ca. 2 % higher compared to the year before. These differences in biological variables may have had an influence on the elevated contaminant concentrations. Also, herring from Väderöarna shows increasing concentrations for several chlorinated pesticides, PCBs, dioxins, and BFRs during the ten most recent years. A reason for this might be that the included fish were older during the seven most recent years, although their size was similar or smaller than previous years. Additionally, the fat percentage of the tissue has also decreased over time and might influence the results. This year's report is also the first to show PFAS concentrations in Cod, Perch and Eelpout where Perch has the highest concentrations of most PFASs within the Baltic Proper sampling sites. This report is also the first report within the Swedish National Monitoring Programme of Contaminants in marine biota presenting environmental contaminant data in eggs of Common tern and Eurasian oystercatcher. In general, many of the compounds could be detected in Common tern and Eurasian oystercatcher.</p>	

Concentrations of several of the contaminants are similar in the two species, but there are exceptions among each contaminant class and even opposing time trends for some contaminants. There are clear differences in contaminant concentrations between species for PCDD/Fs and BFRs; for most PCDD/Fs, concentrations are higher in Eurasian oystercatcher compared to Common tern, while for the BFRs concentrations are higher in Common tern, with the exception of HBCDD.

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1 Summary

Herring from Holmöarna shows elevated concentrations during the last two years of the time series for approximately half of the analysed metals, most chlorinated pesticides, polychlorinated biphenyls (PCBs) and brominated flame retardants (BFRs). The last two years' Herring is on average one year older than the previous years', heavier and the last years' fat percentage is ca. 2 % higher compared to the year before. These differences in biological variables may have had an influence on the elevated contaminant concentrations.

Herring from Väderöarna shows increasing concentrations for several chlorinated pesticides, PCBs, dioxins, and BFRs during the ten most recent years. A reason for this might be that the included fish were older during the seven most recent years, although their size was similar or smaller than previous years. Additionally, the fat percentage of the tissue has also decreased over time and might influence the results.

This is the first report within the Swedish National Monitoring Programme of Contaminants in marine biota presenting environmental contaminant data in eggs of Common tern and Eurasian oystercatcher. In general, many of the compounds could be detected in Common tern and Eurasian oystercatcher. Concentrations of several of the contaminants are similar in the two species, but there are exceptions among each contaminant class and even opposing time trends for some contaminants. There are clear differences in contaminant concentrations between species for PCDD/Fs and BFRs; for most PCDD/Fs, concentrations are higher in Eurasian oystercatcher compared to Common tern, while for the BFRs concentrations are higher in Common tern, with the exception of HBCDD.

This year's report is also the first to show PFAS concentrations in Cod, Perch and Eelpout. Among the detected PFASs, Perch has the highest concentrations within the Baltic Proper sampling sites compared to Cod and Eelpout, except for FOSA. FOSA concentrations are highest in Cod, i.e. double as high as in Eelpout and nearly 15 times higher than in Perch.

2 Introduction

This report briefly summarises the monitoring activities within the Swedish National Monitoring Programme for Contaminants in marine biota. It is the result of joint efforts from several departments, institutes and agencies: 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 *Department of Environmental Research and Monitoring* at the Swedish Museum of Natural History is responsible for the coordination of this 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 monitoring programme is financed by the *Swedish Environmental Protection Agency (SEPA)*.

The contaminant concentrations in the biological samples of 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 to supply reference values for regional and local monitoring programmes.
- To monitor long-term time trends and to estimate the rate of changes found.
quantified objective: to detect an annual change of 10 % within a 10 year time period, with a power of 80 % at a 5 % significance level.
- To estimate the response in marine biota of measures taken to reduce the discharge of various contaminants.
quantified objective: to detect a 50 % decrease within a 10 year time period, with a power of 80 % at a 5 % significance level.
- To detect incidents of regional or widely spreading character and to monitor the potential renewed usage of (regionally) 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 indicate large scale spatial differences.
quantified objective: to detect differences of a factor of 2 between sites, with a power of 80 % at a 5 % significance level.
- To explore historical and regional differences in the composition and pattern of e.g. PCBs, HCHs, DDTs, PCDD/Fs, PBDEs, HBCDD, PAHs, OTCs and PFASs as well as the ratios between various contaminants.

The storage of frozen individuals or sample materials from the annual systematic sampling together with the large number of additional samples in the *Environmental Specimen Bank* enables future retrospective studies of today unknown or impossible to analyse compounds and future control analyses after suspected analytical errors.

In addition to the above-mentioned objectives, the monitoring programme is also a valuable resource for marine monitoring as it:

- Provides high quality data; the material collected and analysed in a consistent manner is available for evaluation of hypotheses and as input data in models concerning the fate and distribution of various 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. A cooperation with the Swedish Food Administration is established as well as with the Swedish Radiation Safety Authority for analysing 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 from the time series of the national programme.
- 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.

The present report displays the time series of analysed contaminants in biota and summarises the results from statistical analyses. Generally, it does *not* give background information nor explanations for the reasons of significant changes over time. Increasing concentrations thus indicate the need for further studies. Short comments are given on temporal trends, on spatial variation and on some contaminants that differ in their geometric mean concentration between various species caught at the same site. It should be stressed that geographical differences may not always only reflect anthropogenic influences, but can also be affected by factors such as productivity, temperature, salinity etc. On occasions, notes of seasonal variation and differences in concentration between tissues in the same species are given. This information may indicate the relative appropriateness of the sampled matrix and be of help in designing future monitoring programmes.

This report is a shorter version compared to the annual previous reports of this programme (*Comments Concerning the National Swedish Contaminant Monitoring Programme in Marine Biota*) and focuses mainly on differences compared to the previous year and earlier time trends. Detailed results are given on contaminants in eggs from the Eurasian oystercatcher and the Common tern, as well as on PFASs in Perch, Cod and Eelpout as these have not previously been reported and/or are of high concern. More detailed background information can be found in the report of the previous year (Bignert et al. 2017). A selection

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

3 Sampling and analytical methods

3.1 Sampling area and collected specimen

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 resulting data are annually reported to the International Council for the Exploration of the Sea (ICES) as 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).

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 viviparus*) as well as soft tissue samples of Blue mussel (*Mytilus edulis*) and egg homogenate samples of the species Guillemot (*Uria aalge*), Common tern (*Sterna hirundo*) and Eurasian oystercatcher (*Haematopus ostralegus*). Previously, Dab (*Limanda limanda*) and Flounder (*Platichthys flesus*) samples were analysed as well, however, currently these two species are only caught and stored in the Environmental Specimen Bank.

Generally, only healthy looking and undamaged individuals and specimens are sampled. The collected specimens are placed individually in polyethylene plastic bags, and are as soon as possible frozen and transported to the laboratory pending sample preparation. For all fish, adult individuals of a narrow length range (specific for each species) are analysed to minimize the within-year and in between-year variation. For Herring in particular, the individuals selected for contaminant analysis have also a narrow age range. In the laboratory, biological parameters, such as total weight, length, size, sex, age etc. are recorded and stored in the database with other additional information on the specimen. Subsamples of fish muscle and liver tissues are prepared together with homogenates of bird egg and blue mussel samples and thereafter sent to the different laboratories for chemical analysis. For further detailed information on the species, sampling and specimen treatment see the previous more detailed report (Bignert et al. 2017).

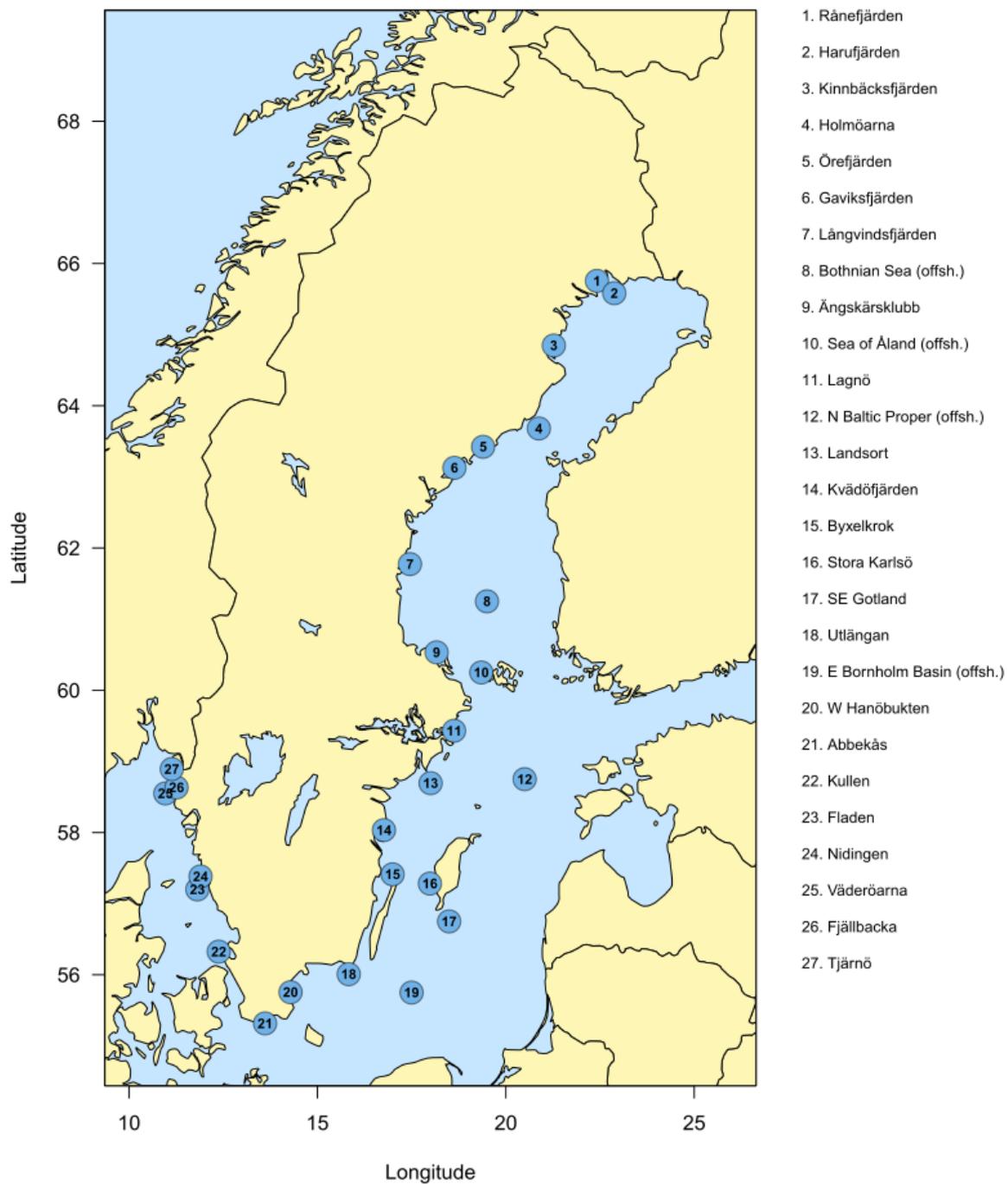


Figure 1: Sampling sites within the Swedish National Monitoring Programme for Contaminants in marine biota (N=North, E=East, W=West, S=South; offsh.=offshore);

3.2 Analytical methods

The methods are described in detail in the previous report (Bignert et al. 2017). In the following section, changes or adaptations in comparison to the previous protocols are briefly listed by compound class.

For organochlorines, including both the chlorinated pesticides and the PCBs, the reference for the analysis should read Eriksson et al. (1997) instead of Eriksson et al. (1994).

For PFASs, the homogenised sample aliquots of bird eggs were also spiked with 1.0 ng each of a suite of mass-labelled internal standards (¹⁸O- or ¹³C-labelled perfluoroalkyl sulfonates and carboxylic acids), as all the other samples. Thereafter the method proceeds as previously described. The entire quality control section should read as follows: 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 74 and 110 % for all analytes. 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 0.02 and 0.3 ng/g wet weight for the different compounds.

Determination of PAHs was carried out using a high performance liquid chromatograph (HPLC, type Varian Prostar 240, M410) with a 3 µm C18-column (Pursuit PAH 100* 3 mm, Agilent). A linear gradient elution programme was used, starting with acetonitrile/water 50:50 and ending with 100 % acetonitrile (Rathburne HPLC-grade). Additionally, the information on the certified reference material was updated to: 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). Certified reference material (CRM) was used to check the performance of the method (NIST, SRM 2974a, Organics in freeze-dried mussel tissue (*Mytilus edulis*)).

For the sample preparation for organotin compound analysis, internal standards (monoheptyltin, tripropyltin) and 5 ml hydrobromic acid (konc) were added to the freeze-dried samples. The mixture was extracted twice with 20 ml dichloromethane on a shaker. Thereafter, the procedure continued as previously pending the instrument analysis, for which the gas chromatograph (GC) was coupled to a new mass spectrometer, a 7010 triple quadrupole mass spectrometer (Agilent).

There were no changes or adaptations to the previous analytical methods for PCDD/Fs, metals and stable isotopes.

4 Statistical treatment and graphical presentation

4.1 Data treatment

In general, raw data is used in the statistical analyses; however, due to a large variation in fat content for specific matrices the raw data for some compounds has been adjusted for fat content prior to the statistical analyses to reduce a high variation and hence improve the statistical power. Adjustments are done for cadmium, lead and DDE analysed in cod liver as well as for DDE analysed in muscle from Herring sampled at Ängkärsklubb and Utlängen during the spring.

The time trends for Σ PCB are based on calculated values; up to 1988 the Σ PCB is based on a relationship between PCB-10 and total sum of PCBs, whereas after 1988 the Σ PCB is instead based on the relationship between both PCB-138+PCB-163 and PCB-10 and the total sum of PCBs.

4.2 Trend detection

One of the main objectives of the monitoring programme is to detect statistical significant time trends. The detection of trends is carried out in four steps as follows below.

4.2.1 Log-linear regression analyses

The log-linear regression analyses are performed for data based on the *entire investigated time period* as well as for data from the *most recent ten years*. The rationale for additionally analysing data exclusively from the last ten years is to provide a more recent trend of the contaminant concentrations, because earlier decades' concentrations might reflect a different trend, strongly affecting the trend of the entire time period.

The slope of the regression line describes the annual percentage change. A slope of 5 % implies that the initial concentration is halved after 14 years, whereas a 10 % slope corresponds to a concentration reduction by half after 7 years (Table 1).

The log-linear model assumes that the residuals are independent of each other and evenly distributed, which means that the residuals for each year should be evenly (i.e. without any systematic trend) spread around zero. If this assumption is violated, statistical inferences can be erroneous. Since observations in time series data often exhibit autocorrelation (i.e. correlating residuals), a robust estimate of the variance matrix is being used which is consistent under autocorrelation and heteroscedasticity (i.e. non-constant variance of the residuals) (Zeileis 2004).

Table 1: The approximate number of years required to double or halve the initial concentration, assuming a continuous annual change of 1, 2, 3, 4, 5, 7, 10, 15 or 20 %.

Annual slope (%)	1%	2%	3%	4%	5%	7%	10%	12%	15%	20%
Years required to double the concentration (years)	70	35	24	18	14	10	7	6	5	4
halve the concentration (years)	69	35	23	17	14	10	7	6	4	3

4.2.2 Non-parametric trend test

The regression analysis presumes (among other things) that the regression line gives a good description of the trend. The 'leverage effect' of points lying at one end of the regression line is well-known. An exaggerated slope caused by a single or a few points lying towards one end of the line increases the risk of detecting a false significant trend in cases where no real trend exists. A non-parametric alternative to the regression analysis is the Mann-Kendall trend test (Gilbert 1987, Helsel and Hirsch 1992, Swertz 1995). This test generally has a lower power than the regression analysis and does not take differences in the magnitude of concentrations into account. The Mann-Kendall trend test only counts the number of consecutive years during which the concentration increases or decreases compared to the year/s before. The eventual case that the regression analysis yields a significant result, but the Mann-Kendall test does not, could be explained by the lower power of the Mann-Kendall test, or alternatively a disproportionately large influence of points on the slope. Hence, the Kendall's ' τ ', and the corresponding p-value are reported in the statistics tables in the Appendix. The Kendall's ' τ ' ranges from 0 to 1, like the traditional correlation coefficient, Pearson's ' r ', but will generally be lower. Comparably strong linear correlations of 0.9 or above, correspond to a τ -value of about 0.7 or above (Helsel D.R. and Hirsch R.M. 1992). The Mann-Kendall test was recommended by the US EPA as a complementary trend test in water quality monitoring programmes when evaluating annual samples (Loftis J.C. et al. 1989).

4.2.3 Non-linear trend components

As an alternative to the regression line, smoothed lines can be used to describe the development of data over time. The smoother applied here is a locally estimated scatterplot smoother (LOESS). For each point, the LOESS fits a weighted linear model using a subset of points located close to the measured data point. The Generalized Cross Validation determines the value range that is considered as being close to the measured data point. With an analysis of variance (ANOVA) test, the significance of the trend lines is tested to compare the variance explained by the smoother against the variance explained by the linear model (Cleveland and Devlin 1988). This procedure is used in assessments at ICES and is described by Cleveland and Devlin (1988).

4.2.4 Change point detection

For contaminant monitoring programmes such as this one, an important objective is to observe and evaluate if bans or restrictions of chemicals led to the desired goal of reducing the concentrations of contaminants in the environment. Change point detection is one method to estimate at which time point upward trends change direction and become downward trends, possibly induced by regulatory actions and restrictions.

The algorithm for change point detection is described in Sturludottir et al. (2017). Briefly, two models are fitted to the data and are compared with each other. The first model allows for different slopes in two subsets of the data, whereas the second model does not allow for a change point. The change point is iteratively sought for by splitting the data in two groups, one year at a time and with a minimum of four years in each group. In each step, a likelihood-ratio test is performed and the maximum value of the test statistics is compared to critical values. This method is presented by Sturludottir et al. (2017). The null hypothesis is that no change-point exists.

4.3 Outliers and values below the limit of quantification

Of special concern are observations further away from the regression line than expected based on the residual variance around the line. These deviations may be caused by an atypical occurrence in the physical environment, a change in the pollution load, or errors in the

sampling or analytical procedure. The procedure to detect suspected **outliers** in this context is described by Hoaglin and Welsch (1978). It makes use of the *leverage coefficients* and the *standardised residuals*. The standardised residuals are tested against a $t_{0.05}$ distribution with $n-2$ degrees of freedom. When calculating the i^{th} standardised residual, the current observation is left out, implying that the i^{th} observation does not influence the slope or the variance around the regression line. The suspected outliers are merely indicated in the figures and are still included in the statistical calculations.

Reported values below the limit of quantification (LOQ) are substituted by the value of the reported LOQ divided by the square root of 2. Historically, in cases where the information on the value of the LOQ might have been missing, the minimum value of the substance concentration during that year was taken instead of the LOQ in this calculation.

4.4 Plot legends

Analytical results for each of the investigated compounds are displayed graphically. A selection of sites and species are presented as figures; none of the presented time series is shorter than five years.

Each figure has a header with the species name and sampling sites. In the figure, the geometric mean concentration of each year (big circular points) is displayed together with the individual analyses/observations (small dots) and the 95 % confidence interval of the geometric means (bars around the mean). A cross inside a big circular point indicates that one or several suspected outliers are included in the geometric mean value and hence the statistical analysis. Bar charts (in grey) indicate that all values during that particular year are below the limit of quantification (LOQ), the heights of the bar chart is equal to the geometric mean concentration of the reported LOQ. The LOQ can differ for the observations within the same sampling year e.g. based on different matrix effects due to varying fat content. Sample concentrations below the LOQ are included into the statistics as the LOQ value divided by the square root of two.

4.4.1 Trend components

The trend for the entire time period and the last ten years is presented by a red regression line if $p < 0.05$ and by a blue regression line if $0.05 < p < 0.10$. The residual variance around the line (i.e. the coefficient of variation, CV) compared to the residual variance for the entire time period indicates if the sensitivity has increased, e.g. as a result of improved sampling technique or improved chemical analysis.

A smoother is applied to test for non-linear trend components (see section 4.2.3). The smoothed line is plotted if $p < 0.05$, indicated by a red line, and if $0.05 < p < 0.10$, indicated by a blue line.

4.4.2 Figure text

Threshold, lw% or dw%. If the **threshold** (see chapter 6.8) is expressed in a different unit than the concentration given in the figure, the threshold is recalculated to be comparable with the presented data. The recalculation is based on the mean value (over the entire time series) for e.g. **lw%** lipid weight, or **dw%** dry weight in percent for each site.

slope reports the slope, expressed as the annual change in percent together with its 95 % confidence interval;

R² reports the coefficient of determination (R^2) together with a probability value (p) for a two-sided test (H_0 : slope = 0), i.e. a significant value is interpreted as a true change, given that the assumptions of the regression analysis are fulfilled;

y(YYYY) reports the concentration estimated from the smoother line for the last year together with a 95 % confidence interval e.g., $y(2016)=2.55(2.17, 3.01)$ is the estimated concentration (2.55) of the year 2016, where the residual variance around the regression line is used to calculate the confidence interval (between 2.17 and 3.01) in the unit for the concentration given on the y-axis.

4.5 Spatial visualisation of the geometric mean concentrations

The size of the circles represents the geometric mean for the time period indicated at the top of the figure (mostly last 3 years). The geometric mean concentration is given in two significant digits and is symbolised by the size and filled colour of the circles to the right of the figure. The listed concentration range is given for the 5th, 25th, 50th, 75th and 95th percentiles for each compound with more than five monitoring sites. In cases where the number of sites is five or less than five, the unique values are shown. A red border around the circle indicates that the geometric mean concentration value at the according site exceeds the threshold value. If the threshold is expressed in a different unit than the concentration given in the figure, the threshold is recalculated to be comparable with the presented data. The recalculation is based on the mean value (mostly last 3 years) for e.g. lipid weight, or dry weight in percent for each site.

5 The power of the programme

For the interpretation of time series data, it is essential to know with which statistical power temporal changes can be detected, i.e. how likely it is to reveal true changes in concentrations over time. It is crucial to know if the absence of a trend implies a stable situation in the environment or if the monitoring programme is designed too poorly to detect potential changes in concentrations over time. There are two different approaches to test this: 1) by estimating the power based on a 'random' between-year variation, and 2) by estimating the lowest detectable trend at a fixed power as a measure of the time series' sensitivity. In this monitoring programme, both approaches are used (see also objectives in chapter 0).

To estimate the power based on 1), a 'random' between-year variation, we use the residual distance from the log-linear regression line as this line often fits current observations and can be considered an acceptable and 'neutral' representation of the true time series trend. However, in some cases for which a significant 'non-linear' trend has been detected, the log-linear regression line may not be an appropriate fit.

The power of the programme is largely dependent on the length of the time series. For most compounds the sampling and analysis are already ongoing for decades, which makes for a high power of the monitoring programme. However, the length of the time series varies depending on the site and investigated compound. In addition, one or several outliers in a time series can influence the power by increasing the estimated between-year variation. In the presented monitoring programme, suspected outliers are included, which means that the power and sensitivity might be underestimated. The underestimation is from a monitoring perspective more favourable than an overestimation of the power of the programme. It prevents us from interpreting changes in data as a true increase/decrease, which in fact are not statistically significant.

For more detailed information on the power and the sensitivity of specific time series, see the statistics tables presented in Appendix. In the tables, the power to detect an annual change of 10 % based on the number of monitored years of the entire time series, as well as the power estimated as if the slope were 10 % and the number of years were only 10, is presented based on the residual variation for the whole time series. The sensitivity of the time series is reported as the lowest detectable trend (LDT, given in percent per year) for the entire monitoring period as well as for a ten-year period, with the actual between-year variation (i.e. the coefficient of variation, CV) of the monitoring data (whole time period or 10 last years) and at a power of 80 %. Further, the years required (YRQ) to detect an annual change of 10 % with a power of 80 % is given in the appendix table as well.

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 to protect the environment and humans of contaminant and that requires 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-/heptabromodiphenylether, tetra-/pentabrom-odiphenylether, chlordecone, hexabromobiphenyl, lindane, α - and β -hexachlorocyclohexane, pentachlorobenzen and PFOS).

In May 2011, technical endosulfan and its related isomers were added to the list with specific exemptions. Since November 2014, hexabromocyclododecane (HBCDD) was also included in the Stockholm Convention. In May 2015 and May 2017, five more substances (Hexachlorobutadiene, Pentachlorophenol with its salts and esters, Polychlorinated naphthalenes, Decabromodiphenyl ether (commercial mixture, “c-decaBDE”) and Short-chained chlorinated paraffins (SCCPs)) were included in the Stockholm Convention. Three more substances have been proposed to be included on the list and are currently reviewed by the Persistent Organic Pollutants Review Committee: Dicofol, Perfluorooctanoic acid (PFOA) and Perfluorohexane sulfonic acid (PFHxS) as well as the salts of both Perfluoroalkyl acids and their related compounds (SC 2017).

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 aim is to achieve levels near background concentrations for naturally occurring substances, and levels close to zero for synthesised substances. The hazardous substances work is implemented by OSPAR's Hazardous Substances Committee, which is trying to achieve this goal by 2020. 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 125 signatories and 105 ratifications in 2019. 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.Σ

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. (EC 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 the Oslo Paris Convention (OSPAR, i.e. the Convention for the Protection of the Marine Environment of the North-East Atlantic). 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 2.

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 chronic 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 Commission 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 2). 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 2). 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 2. 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 2: Target levels for various environmental pollutants; the chosen target levels in the report are bold.

Group of substance		Target levels		
		Fish ($\mu\text{g}/\text{kg ww}$)	Mussels ($\mu\text{g}/\text{kg dw}$)	Reference
Metals	Cadmium	160 ^{1,2}		QS _{biota, sec pois}
	Lead	300 ¹	1500 ww	food regulation
	Mercury	20		EQS _{biota} (QS _{biota, sec pois})
PAHs	Fluoranthene		110 30 ww	EAC EQS _{biota} (QS _{biota, hh food})
	Anthracene		290	EAC
	Naphtalene		340	EAC
	Phenantrene		1700	EAC
	Pyrene		100	EAC
	Benzo(a)anthracene		80	EAC
	Benzo(a)pyrene		600 5 ww	EAC EQS _{biota} (QS _{biota, hh food})
	Benzo(ghi)perylene		110	EAC
Pesticides	DDE (p,p')	5		EAC
	HCH (incl. lindane)	2.6 (marine) 26 (limnic)		IVL
PCBs	PCB 28	64 lw		EAC
	PCB 52	108 lw		EAC
	PCB 101	120 lw		EAC
	PCB 118	24 lw		EAC
	PCB 138	316 lw		EAC
	PCB 153	1600 lw		EAC
	PCB 180	480 lw		EAC
Other compounds	Σ PBDEs (BDE-28, -47, -99, -100, -153, and -154)	0.0085		EQS _{biota} (QS _{biota, hh food})
	HBCDD	167		EQS _{biota} (QS _{biota, sec pois})
	Σ PCDDs+PCDFs (TEQ)	0.0035		food regulation
	Σ PCDDs+PCDFs+dI-PCBs (TEQ ₂₀₀₅)	0.0065 0.0012		EQS _{biota hh} based on the food regulation QS _{biota, sec pois}
	HCB	10		EQS _{biota} (QS _{biota, hh food})
	PFOS	9.1 ¹		EQS _{biota} (QS _{biota, hh food})

¹ The threshold is expressed in a different tissue than which is analysed within the monitoring programme. The threshold used for evaluation in the report has thus been recalculated to represent the corresponding concentration of the threshold in the monitored tissue. Which for lead in liver is: 0.085 $\mu\text{g}/\text{g}$ wet weight, for Cd in liver: 6.65 $\mu\text{g}/\text{g}$ wet weight and for PFOS in liver: 155 ng/g wet weight. For further information, see Bignert et al. (2017).

²Target 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.

7 Biological data: Length, weight, age, condition and fat content

Concentrations of contaminants in biota give a good picture of the bioavailable contaminant load in the environment. However, the contaminant concentration measured in an individual may also be affected by other factors e.g. size, age, condition and lipid content. It is thus important to along with contaminant analysis also record and follow these type of biological variables. The monitoring programme is designed to minimise the influence of the biological variables by sampling a narrow length range (specific for each species) within the same sampling period of four weeks each year.

7.1 Age

Age is determined and recorded for fish in connection with sample preparation. For Herring in particular, more individuals than selected for contaminant analysis are age determined, so a narrow age range can be selected.

Despite the effort to minimise the age range, differences still occur in age between years within the same time series. In addition, an increased age over time can be seen for Herring sampled from Ängskärsklubb in spring, Utlängan in both spring and autumn and from Väderöarna.

The age of Eelpout during the most recent eight years is lower compared to the whole time series.

7.2 Weight and length

The contaminant concentration can be influenced by the size of the fish. To avoid high variability in contaminant concentration within a year and between years, only fish within a specified length range (species specific) is collected, and sampled within the monitoring programme.

Due to the limited length range, similar sizes of the sampled fish are seen over time for many of the monitored sites. However, for some sites, changes in size over time of the sampled fish still occur. The length and weight are tightly correlated and follow similar patterns. Length and weight of sampled Herring have increased over time at Holmöarna, Ängskärsklubb (caught in spring) and Byxelkrok, while a decrease in weight can be seen in Herring from Harufjärden. A decrease in both the total length and weight in Herring from Landsort, Utlängan (caught in spring and only for weight also in autumn caught Herring) and Abbekås. Cod from Fladen has increased in length and weight over time, as well as Perch from Kvädöfjärden. The size of Perch sampled at Holmöarna was rather big during the first three years and the last two years of the time series.

The length and weight of sampled Eelpout from Fjällbacka are currently lower compared to the beginning of the time series and has decreased during the most recent ten years.

7.3 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, PCBs, PCDDs and PCDFs) and brominated flame retardants (PBDEs 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.

In Eelpout muscle tissue, the calculated fat content in 2017 based on the time series lies between 0.49 and 0.64 % (Fjällbacka, Holmöarna and Kvädöfjärden) and in Perch between 0.67 and 0.68 % (Holmöarna, Örefjärden and Kvädöfjärden). The fat percent for Cod liver tissue was calculated to be 17 % at Fladen (West Coast) and 50% at South East Gotland (Baltic Proper). The fat content in the Cod liver samples might be influenced due to a switch from individual samples to pooled samples in 2015. The Herring fat content in Harufjärden and Utlängan (both autumn and spring caught Herring) have decreased over time.

7.4 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:

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

For most of the sampling sites (except Ängskärsklubb spring), where Herring has been monitored since the late 1970s, there has been a decrease in the condition of Herring from the start of the series until the mid-1990s or early 2000s, followed by an improved or stable condition of Herring thereafter. The condition of Cod follows a different pattern. The condition of Cod increased during the 1980s until mid-1990s, followed by a decreased condition in the case of Cod from South East Gotland. The condition of Eelpout from Holmöarna has decreased over time, whereas it increased in Eelpout from Kvädöfjärden.

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

In general, **metal** concentration trends seem to go down over time in the northern part of the Baltic Sea, although most of the trends are not statistically verified (Figure 2). In the Baltic Proper, the metal trends appear to be stable or going slightly upwards, although for most sites these trends were not statistically significant.

Noteworthy is that for **different species** sampled at the same monitoring site, metal time trends can vary (Figure 2). For Kvädöfjärden, concentrations are decreasing in Blue mussel for all metals (statistically significant for arsenic, cadmium, chromium and lead) except for zinc. On the contrary, for Eelpout sampled at the same site, arsenic, cadmium, copper, selenium and silver are significantly increasing over time.



Figure 2: 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). The value of the slope is shown if the trend is significant ($p < 0.05$).

For **Herring** sampled at **Holmöarna**, almost all metal concentrations are elevated the last two years and can have a disproportionate influence on the statistical evaluation of those time series. As an example, the time trend for silver is now statistically significant with an upward trend of 36 % per year. Time trends from this site should therefore be treated with caution. When it comes to biological factors potentially explaining these elevated concentrations, the age of the Herring could be an explanation as the specimens sampled the two last years have

in general been older compared to previous years. In 2017, also the fat content was markedly high (detected outlier). In general, the size of the fish has increased over time (time series of 9 years) with significant upward trends for total length, total weight and for Fulton's condition index.

For **chromium**, time trends appear to go upwards according to the heatmaps for all fish at nearly all sites. However, the trends are highly influenced by consistently low values across the time series between the years 2007 and 2009, which might reflect problems related to the analytical method, rather than a true trend. For information on specific time series, see Appendix.

9 Polychlorinated Biphenyls (PCBs)

The PCB concentrations show a general downward trend over time (Figure 3). However, for Holmöarna in the Bothnian Bay and for Väderöarna on the west coast, time trends are going upwards. For Holmöarna, elevated concentrations the last two years are driving the upward, occasionally statistically significant, time trends, whereas for Väderöarna, increasing concentrations over the last ten years are resulting in several statistically significant upward time trends (see Appendix).

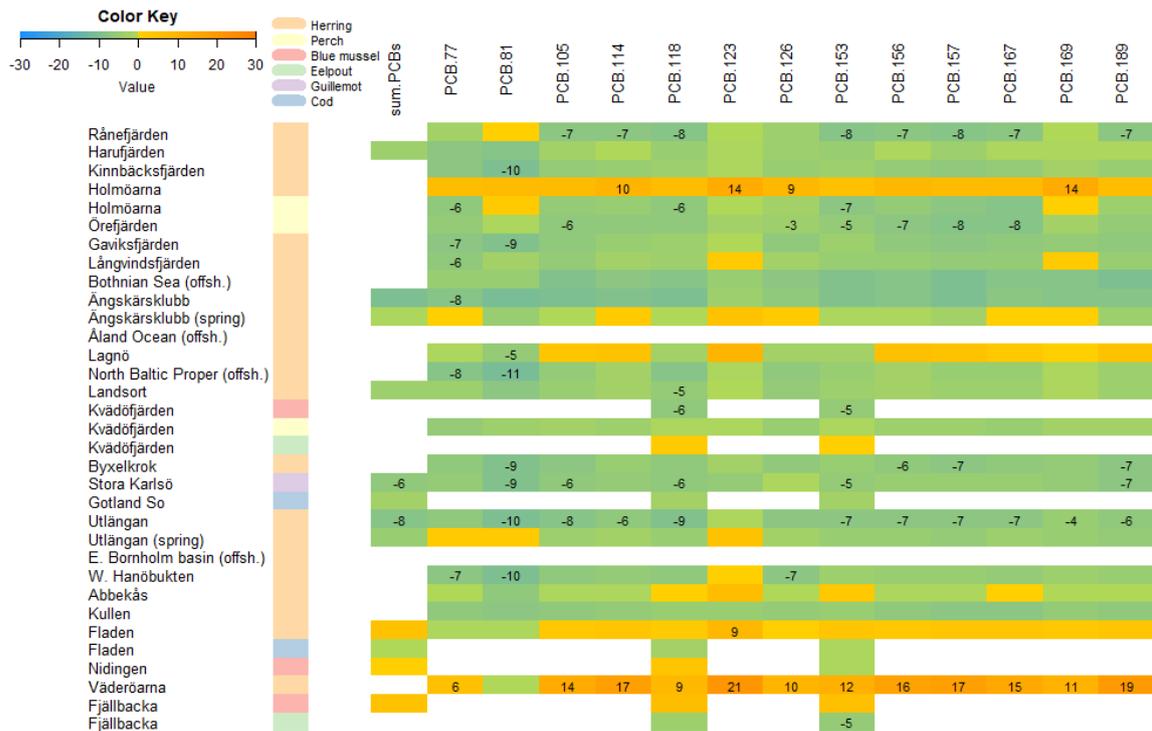


Figure 3: The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). The value of the slope is shown if the trend is significant ($p < 0.05$).

10 Chlorinated Pesticides (DDTs, HCHs, HCB)

For **DDT**, **DDE** and **DDE**, Holmöarna is the only site where concentrations in **Herring** are increasing among the sampling sites in the Bothnian Bay and the Bothnian Sea and is due to elevated concentrations during the two last years. On the west coast, Väderöarna indicates statistically significant (or close to significant ($p < 0.1$) upward time trends for **DDD** and **DDE**, whereas for the parent compound (**DDT**), a concentration drop can be observed for the last year. Elevated concentrations of **DDT** in **Eelpout** sampled in Kvädöfjärden the last years resulted in a significant upward trend over the last ten years.

For α -**HCH**, β -**HCH** and γ -**HCH** (Lindane), concentrations are decreasing and/or are below the level of quantification for most species and sites. No significant upward trends exist for these compounds except for α -**HCH** in Herring sampled at Holmöarna due to elevated concentrations the last two years.

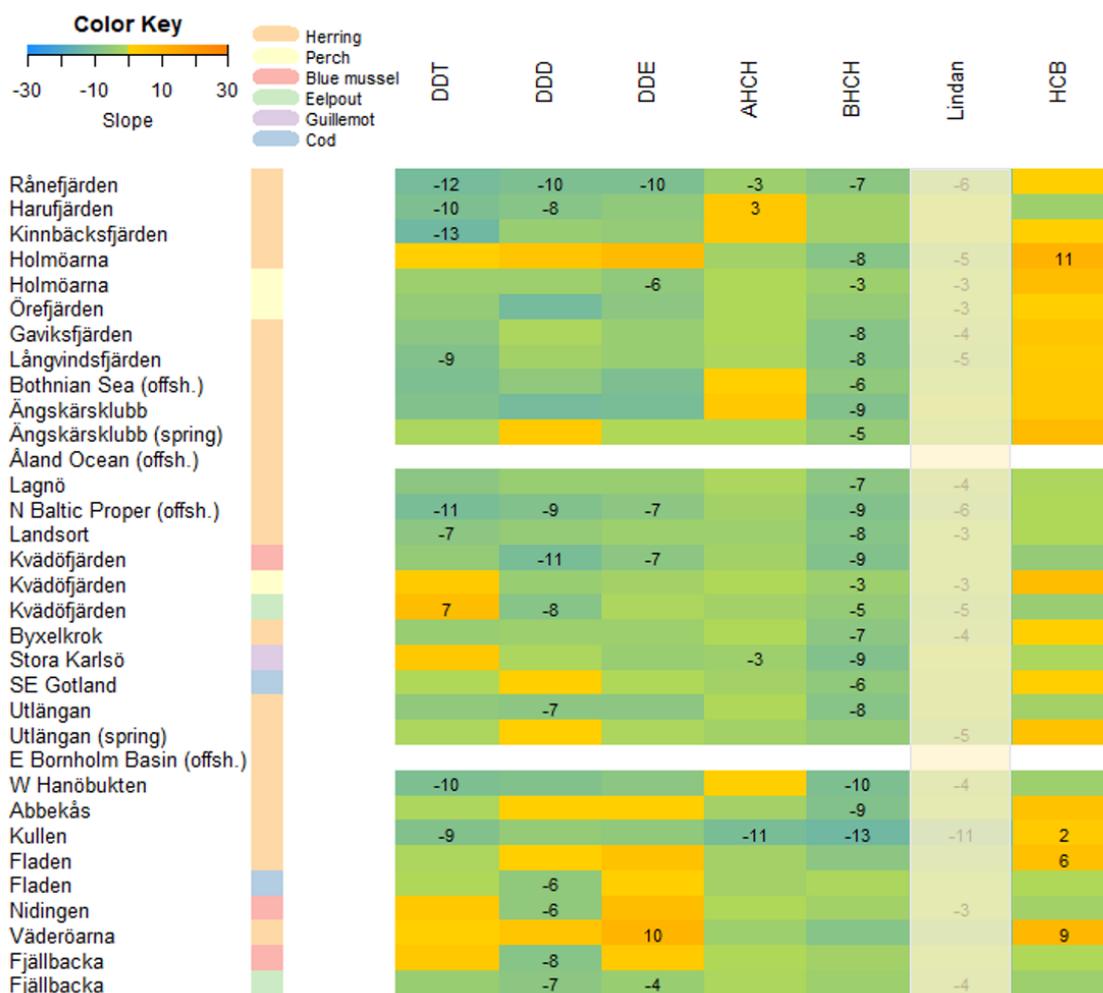


Figure 4: The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). The value of the slope is shown if the trend is significant ($p < 0.05$). Faded areas represent time trends in which all or mostly values are below the LOQ.

For **HCB**, concentrations in **Herring** sampled in the Baltic Sea are within the expected within and in-between year variation and therefore no significant upward trends exist, except for Herring sampled at Holmöarna due to increasing concentrations over the last 5 years. On the contrary, Herring sampled on the west coast (Kullen, Fladen and Väderöarna) have significant

upward trends the last 10 years (Fladen and Väderöarna) or for the entire monitoring period (Kullen) (see Appendix).

11 Polychlorinated Dibenzodioxins /-furans (PCDD/Fs)

Generally, concentration time trends of the PCDD/Fs are going downward, with a few exemptions from some scattered substances at some locations (Figure 5). For some of the PCDD/Fs, significant upward time trends can be observed for Herring sampled at Holmöarna and Väderöarna as well as occasional time series at Ängskärsklubb (spring) and Fladen.

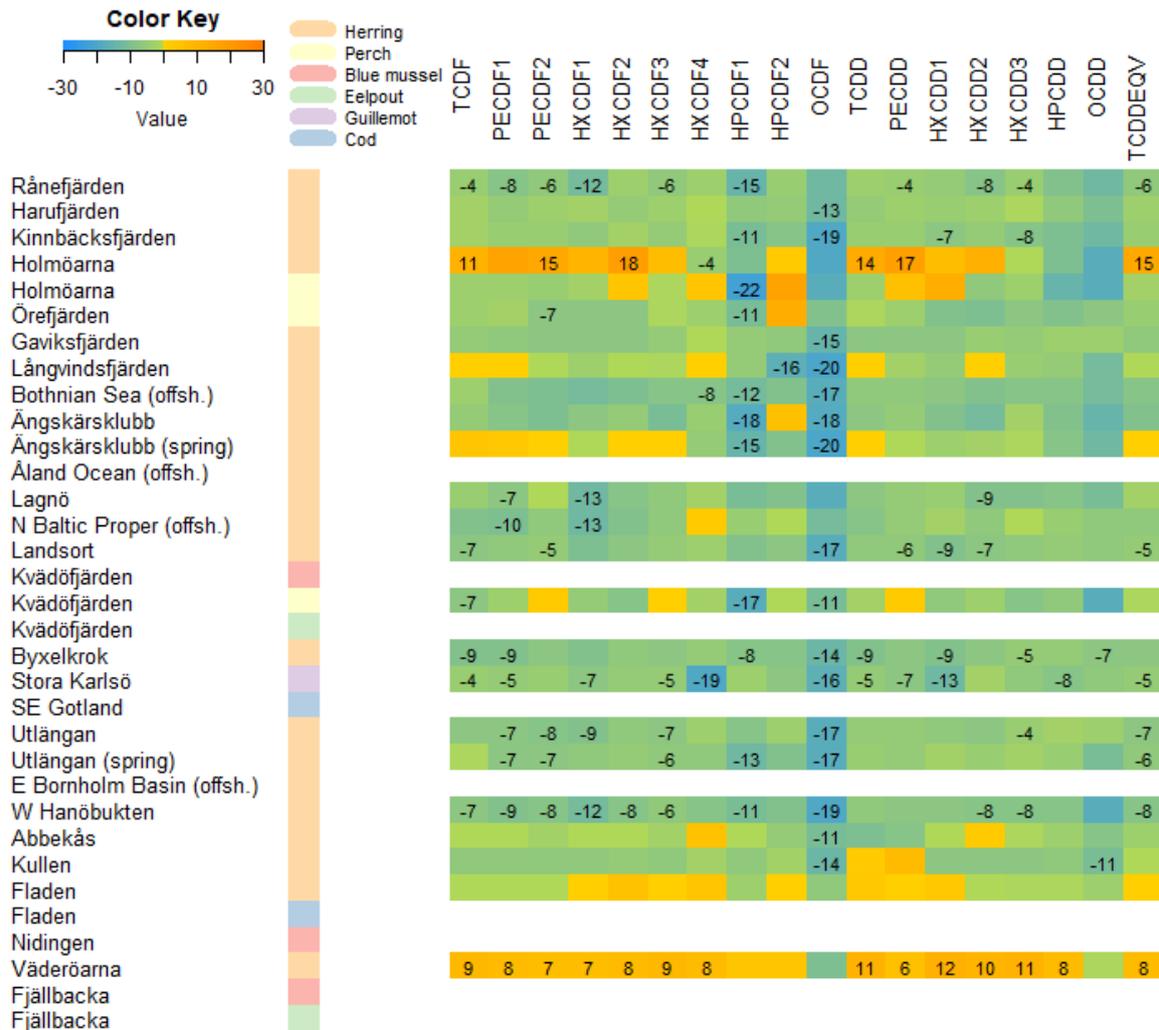


Figure 5: The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). The value of the slope is shown if the trend is significant ($p < 0.05$). **TCDF**, 2,3,7,8-TCDF; **PECDF1**, 1,2,3,7,8-PeCDF; **PECDF2**, 2,3,4,7,8-PeCDF; **HXCDF1**, 1,2,3,4,7,8-HxCDF; **HXCDF2**, 1,2,3,6,7,8-HxCDF; **HXCDF3**, 2,3,4,6,7,8-HxCDF; **HXCDF4**, 1,2,3,7,8,9-HxCDF; **HPCDF1**, 1,2,3,4,6,7,8-HpCDF; **HPCDF2**, 1,2,3,4,7,8,9-HpCDF; **OCDF**, 1,2,3,4,6,7,8,9-OCDF; **TCDD**, 2,3,7,8-TCDD; **PECDD**, 1,2,3,7,8-PeCDD; **HXCDD1**, 1,2,3,4,7,8-HxCDD; **HXCDD2**, 1,2,3,6,7,8-HxCDD; **HXCDD3**, 1,2,3,7,8,9-HxCDD; **HPCDD**, 1,2,3,4,6,7,8-HpCDD; **OCDD**, 1,2,3,4,6,7,8,9-OCDD; **TCDDDEQV**, PCDD/Fs WHO-98 TEQ.

In **Herring** at Kullen, several of the PCDD/Fs have elevated concentrations in 2017 (see Appendix). This is likely the result of the very low fat content measured for these samples (see chapter 7). While in Herring at Kinnbäcksfjärden, N Baltic Proper, and Utlängan, concentrations were lower in 2017 compared to the previous years, and for Byxelkrok, this

low concentration was seen for the second year in a row. PeCDD concentrations at Lagnö, North Baltic Proper (offshore) and Utlängan were at an all-time low in the respective Herring time series. Some other sites and compounds showed a similar picture.

In **Perch** for all sites, many of the **HpCDD/Fs** and **HxCDD/Fs** concentrations in 2017 were elevated and even statistical outliers.

TCDD equivalents expressed on a lipid weight basis are three times higher in 2017 compared to 2016 at Kullen, but the levels are still below the threshold for EQS_{hh} but not secondary poisoning (QS). At Kinnbäcksfjärden, Ångskärsklubb, North Baltic Proper (offshore) Landsort and Utlängan, the geometric mean TCDD equivalents in Herring are below the threshold for secondary poisoning (QS_{biota, sec pois}) this year.

12 Brominated Flame Retardants (BFRs)

The time trends for the Brominated flame retardants are varying for the compounds, sampled species and sampling sites (Figure 6). Similar to other substance groups, **Herring** sampled at **Holmöarna** has elevated concentrations the two last years, because of which many time trends become unjustifiably significant due to the disproportionate influence of those two data point (Figure 6 and Appendix). Therefore, Holmöarna is not discussed in further detail below.

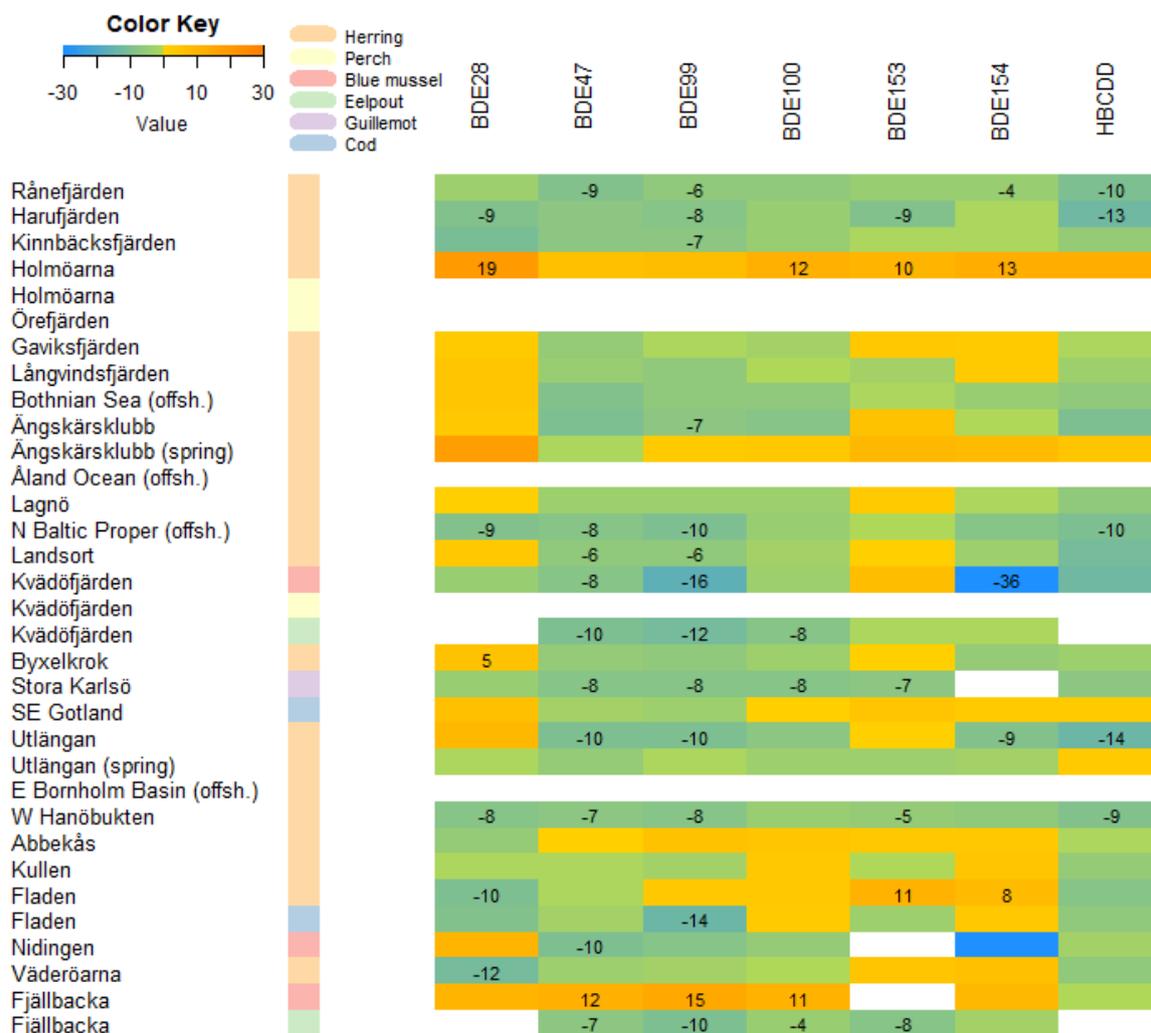


Figure 6: The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). The value of the slope is shown if the trend is significant ($p < 0.05$).

For **PBDE-28**, **PBDE-99** and **PBDE-100** most of the data points were below the LOQ and the time trends are either stable or going downward. A similar pattern is observed for **PBDE-47**, most sampling matrices and sites showed significant downward trends. An exemption from this is **Blue mussel** sampled at **Fjällbacka**, for which a significant increasing time trend exists for the last 10 years (Figure 6), however, this time trend is largely influenced by high concentrations in 2014 and again in 2016 (although not always detected as statistical outliers).

For **fish** and **Blue mussel**, measured concentrations of **PBDE-153** were often below LOQ, occasional statistically or close to statistically significant upward time trends exists (Ängskärsklubb (spring) and Fladen, see Appendix). For **PBDE-154**, time trends are either

stable or going downward, with the exemption of the statistically or close to statistically significant upward time trends for **Herring** sampled at Fladen and Väderöarna. The PBDE-154 concentration in Herring from N. Baltic Proper was lower than all previous years (now below LOQ). The steepest downward trends were found in both cases for **Blue mussel** and PBDE-154 at **Nidingen** and at **Kvädöfjärden**, of which the latter sampling site had an annual significantly decreasing time trend of 36 % (Figure 6). However, this decrease is due to a very high value in 2002 and all the following years after that are much lower (see Appendix).

For **HBCDD**, most sample matrices and sites showed either no statistical trend or statistically significant downward trends. For **Cod** sampled at SE Gotland, a significant non-parametric upward trend exists.

For several of the PBDEs, the **threshold** that relates to the **sum of the PBDEs** is already exceeded by one of the compounds and hence will the threshold always be exceeded regardless if any other individual substance would be below (see time trends and spatial visualisation in the Appendix).

13 Polycyclic Aromatic Hydrocarbons (PAHs)

PAHs are analysed in Blue mussel from three different sites (Kvädöfjärden, Nidingen and Fjällbacka) along the Swedish coast. For all three sites, Σ PAHs is stable or decreasing over time (see Appendix). In 2016, concentrations of several PAHs were elevated on the west coast (Nidingen and Fjällbacka); these are back to the levels of the years before in Blue mussels from Nidingen. However, concentrations of several PAHs are still elevated compared to the five years prior to 2016 (see Appendix). Therefore, the trends for the last 10 years illustrated in the heatmap are especially for Fjällbacka highly influenced by the outlier year 2016 and not statistically significant (Figure 7 and Appendix). In contrast to this, Kvädöfjärden has significant downward trends for the majority of the analysed PAHs (see Figure 7 and Appendix).

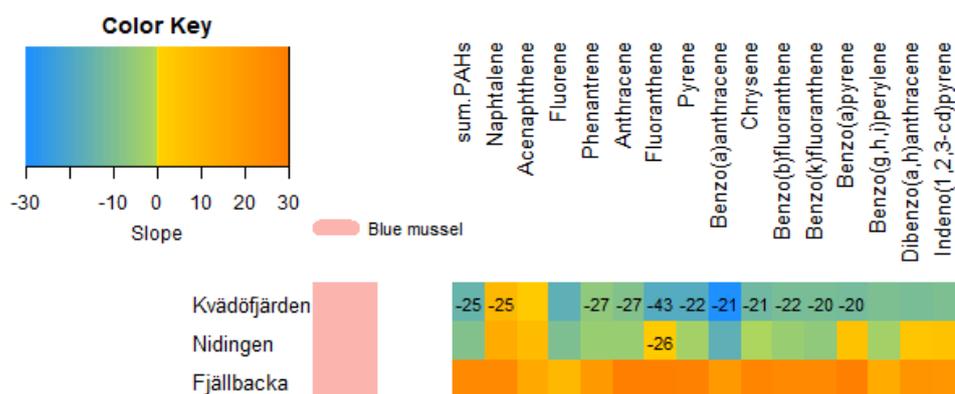


Figure 7. The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). The value of the slope is shown if the trend is significant ($p < 0.05$).

14 Perfluoroalkyl Substances (PFASs)

Generally, the perfluoroalkyl substance concentrations in Herring and Guillemot eggs show downward trends at the majority of the sites (see the green and blue colour in Figure 8). The negative trends of PFDS, PFHpA, PFHxA, and PFBS should be interpreted with caution, as the concentrations are mostly lying below the LOQ (or at a similar level as LOQs in some years), so therefore, simply the decrease of the LOQ due to analytical advances leads to a downward trend. For PFBS, the LOQs were comparably high during the 2017 sampling year. Among the long-chain perfluoroalkyl carboxylic acids (PFCAs), nearly all compounds from PFDA onwards seem to decrease over time with the exemption from very few sites. PFDoDA is most prominently with significant decreases at nearly all sites and with a maximum annual decrease of up to 17 %. PFPeDA and PFTeDA trends might also be influenced by the high LOQs in 2009 compared to the years after, during which concentrations could be quantified lying below this limit. PFOA and PFNA still seem to be on an upward trend at several sites, so seems PFOS among the perfluoroalkane sulfonic acids (PFSA) as well as the perfluorooctane sulfonamide (FOSA). The significant decreasing PFOA trends are highly influenced by the higher LOQ concentrations before 2010, thereafter the measured concentrations are lower than the previous LOQ, but above the new lower LOQs. At these sites, it would be interesting to see the future 10-year trends without any influence by the levels of the LOQ.

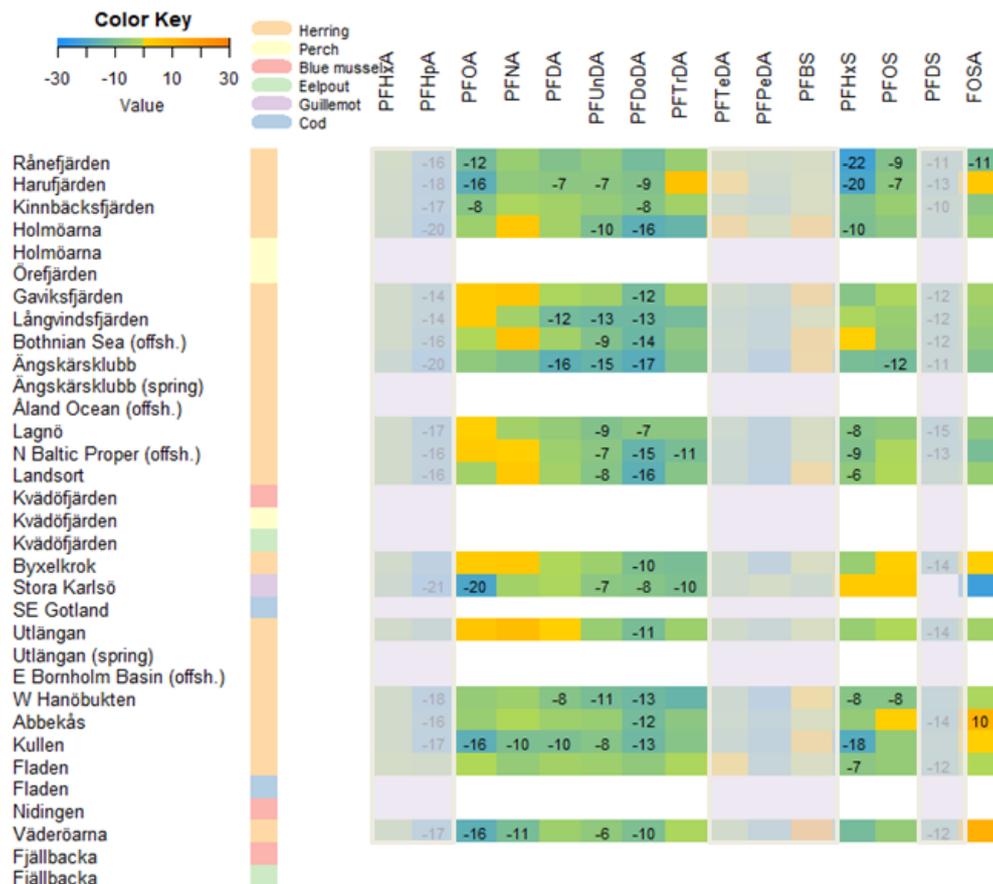


Figure 8: The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). The value of the slope is shown if the trend is significant ($p < 0.05$). Faded areas represent time trends in which all or mostly values are below the LOQ

Noteworthy changes in the PFAS concentrations in Herring and Guillemot eggs in comparison to the previous report are described below.

PFHxS concentrations in Herring caught at Gaviksfjärden were outliers in 2016 (high) and this year (low).

The **sum-PFOS** concentrations were outlying high in Herring from Fladen in 2017, the opposite was true for Rånefjärden, Ängskärsklubb and Landsort. The fitted smoother trend line for Guillemot eggs' sum-PFOS and linear PFOS concentrations seems to be on an increasing trend since 2013.

Linear PFOS concentrations in Herring follow the same trends as sum-PFOS, and statistical outliers at the lower end were found from Rånefjärden, Bothnian Sea (offsh.) and Ängskärsklubb. The **branched PFOS** isomers concentrations had statistical outliers at the lower end in Herring from Harufjärden, Ängskärsklubb and Kullen. Branched PFOS concentrations in Herring were below the LOQ at Harufjärden, Ängskärsklubb and Kullen for the first time in the time series as well as already during 2016 in Fladen and 2015 in Väderöarna.

Guillemot egg **PFOA** concentrations at Stora Karlsö were outlying low and for the first time below the LOQ (after the previous LOQ was lowered in 2006).

PFDA in Herring from Ängskärsklubb was comparably high in 2016, this year though, the concentration was lying even below the LOQ.

Among the longer long-chain perfluoroalkyl carboxylic acids (**PFDA onwards**) indicated several sites lower concentrations during this sampling year than the latest previous years, partly even below the LOQ. This could be a first indication among the marine biota concentrations that mirrors the production shift towards the shorter chain PFASs, which is already ongoing for nearly two decades. However, the next sampling years should be awaited to corroborate this hypothesis.

After the high concentration in 2015, br-FOSA concentrations in Herring seem to decrease, especially in the Baltic Proper and Kullen. The lin-FOSA concentrations in Herring were lower at all sights compared to 2015 data.

14.1 PFASs in Cod, Perch and Eelpout

This year's report is the first to show **PFAS concentrations in Cod, Perch and Eelpout**, the figures can be found in the PFAS chapter of the Appendix.

PFBS concentrations were below the LOQ in all fish species during 2016. The concentrations measured in the two liver sample pools from 2017 differed for all fish species with up to an order of magnitude (maximum measured concentration: Perch from Kvädöfjärden with more than 10 ng/g ww liver).

Among all sites and fish species, **PFHxS** concentrations are highest for Perch from Kvädöfjärden in 2017, with a geometric mean at about 0.25 ng/g ww liver, all other geometric means are below 0.1 ng/g ww liver. Kvädöfjärden Perch is also noteworthy due to the higher concentrations during 2017 in comparison to 2016.

In all fish, the **sum-PFOS** concentrations are lying below the threshold; the concentrations in Perch at Kvädöfjärden are highest with up to about 125 ng/g ww liver. In the Northern part of the Baltic Sea, at Holmöarna, the sum-PFOS concentrations in Perch are a factor of 10 lower (in the low 2-digit range) compared to the Baltic Proper, as were the concentrations in Cod from SE Gotland, compared to the west coast. The sum-PFOS concentration in Eelpout from Kvädöfjärden (in the upper 1-digit range) is a factor of 15 lower than in Perch from this site. Interestingly, the PFOS concentrations in Cod and Eelpout (only from Kvädöfjärden) are higher in 2016 than 2017; the opposite is the case for Eelpout from Fjällbacka and Perch from both sampling sites.

PFDS concentrations are below the LOQ in all species at all sites.

Among the **PFCAs**, PFHxA concentrations were below the LOQ in all species at all sites, except from Cod caught at Fladen in 2016 (pools: ca. 0.7 and 1.1 ng/g ww liver).

The PFOA concentrations in Cod and Eelpout were lower in 2017 than the previous year, which was the opposite for Perch. Highest PFOA concentrations were measured in the Perch sample pools in 2017 with about 0.5 and 1.0 ng/g ww liver.

PFNA, PFDA, PFUnDA and PFDoDA geometric mean concentrations were found to be higher in 2016 than 2017 for the majority of the sampling sites and species (exemptions: Eelpout: PFDA at Kvädöfjärden; Perch: PFNA at both sites, PFDA at Holmöarna and PFUnDA at Holmöarna). Maximum geometric mean concentrations were detected for PFNA in Cod from SE Gotland and Perch from Kvädöfjärden with between 4 and 5 ng/g ww liver in 2016 and 2017, respectively, and at the same sites the PFDA concentrations were highest as well (maximum between 2 and 3 ng/g ww liver). PFUnDA and PFDoDA concentrations were highest in Perch from Kvädöfjärden, with up to 8 ng/g ww liver and up to 2 ng/g ww liver, respectively.

As for the other **long-chain PFCAs**, PFTrDA and PFTeDA concentrations were lower in 2017 than 2016. Most PFTeDA concentrations were even below the LOQ in 2017. In 2016, the measured concentrations were highest in Perch from Kvädöfjärden with both sample pools lying below 0.3 ng PFTeDA/g ww liver and between 3 and 4 ng PFTrDA/g ww liver.

PFPeDA concentrations were below the LOQ or in the low or even below the one decimal place ng/g ww liver concentration range.

FOSA concentrations were higher on the West coast sampling sites and highest for Cod at Fladen with about 3.5 ng linear-FOSA/g ww liver in 2016. Concentrations during 2017 were generally lower than the previous year. Several geometric mean concentrations were below the LOQ at both sampling sites for Perch (br-FOSA both years and linear and sum FOSA in 2017) and for Eelpout the branched FOSA concentration in 2017.

Generally, concentrations of each PFAS compound in Cod, Perch and Eelpout were in a similar range with at the most up to one order of magnitude difference in the Baltic Proper stations. Among the detected PFASs, Perch has the highest concentrations within the Baltic Proper sampling sites compared to Cod and Eelpout, except for FOSA. FOSA concentrations are highest in Cod, i.e. double as high as in Eelpout and nearly 15 times higher than in Perch (geometric mean of 2016 and 2017 data). As in Herring, FOSA concentrations in Cod and Eelpout were higher in fish from the west coast compared to fish from the Baltic Proper.

15 Organotin Compounds (OTCs)

Organotin compounds were measured at three sites (Holmöarna, Örefjärden and Kvädöfjärden) in Perch. Mean annual concentration trends are varying depended on the compound and site, though none of the trends is statistically significant. For most of the organotin compounds, the values were lying below or at the level of the LOQ during all years. Therefore, one should not talk about any tendencies for these compounds indicated in the heatmap, as they are highly LOQ dependent. The only exemptions are possibly TBT and DiPT, where concentrations are decreasing (see the detailed time trends in the Appendix). Even for DiPT, the heatmap might be misleading, considering that only concentration data after 2012 appear to be decreasing (during the years before 2012 most measured values were lying below the LOQ). TPT trends appear to have strongly increased in 2017 compared to previous years, which might be due to a contamination in the laboratory when preparing the pooled samples for the analytical laboratories with newly bought plastic tweezers (still under investigation).

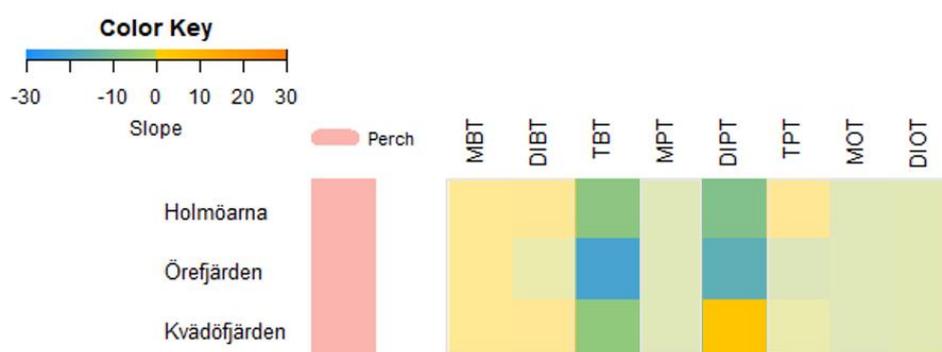


Figure 9: The heatmap illustrates the trends of the entire substance group for the last 10 years by colour. Sites are ordered according to their geographical position starting from the Bothnian Bay to the Skagerrak Strait (North Sea). The value of the slope is shown if the trend is significant ($p < 0.05$). **MBT**, Monobutyltin; **DIBT**, Dibutyltin; **TBT**, Tributyltin; **MPT**, Monophenyltin; **DIPT**, Diphenyltin; **TPT**, Triphenyltin; **MOT**, Monoctyltin; **DIOT**, Dioctyltin. Faded areas represent time trends in which all or mostly values are below the LOQ.

16 Common tern and Eurasian oystercatcher

This is the first report within the Swedish National Monitoring Programme of Contaminants in marine biota presenting environmental contaminant data in eggs of Common tern and Eurasian oystercatcher. This separate chapter is dedicated to those two species due to their novelty within the monitoring programme and upcoming evaluation for further sampling. The figures can be found in the Appendix in the according chapter of the substance class of interest.

There are several advantages of using bird eggs for monitoring of contaminants: 1) Eggs contain high concentrations of bioaccumulative contaminants and hence serve as an early detection of new contaminants that may not be sufficiently enriched in other matrices in order to be detectable. 2) The contaminants in eggs are representative of female exposure to contaminants and indicative of the next generation's exposure. 3) In addition, eggs mostly can be easily collected, handled and stored. For these reasons, bird eggs have proven to be a good matrix for the measurement of environmental contaminants (Helander et al. 1982, Miller et al. 2014). However, there are several factors that have to be considered when using bird eggs in monitoring: 1) the effect of egg laying sequence on the contaminant concentration, 2) the suitability of eggs to represent the contaminant exposure at the sampling site; 3) the trophic position of the bird species in the food web, and; 4) the food sources of the bird species which affect both the contaminant concentration but also the energy content in the food ; 5) the effect of the bird's strategy to allocation energy into the egg on the contaminant concentration.

Two terms are commonly used to describe reproductive investment strategies: "capital" breeders invest in reproduction, i.e. their eggs, from resources that are stored in their bodies (endogenous), whereas "income" breeders invest in reproduction directly from externally acquired resources (Stephens et al. 2009). However, recent literature suggests that these terms are not dichotomous, but rather lie at opposite ends of a continuum (Klaassen et al. 2006). From a monitoring perspective, income breeders are favourable given that the aim is to represent a local and current contaminant status in the environment.

Common tern migrates long distances. The breeding strategy of Common tern is disputed; Hobson et al. (2000) describes the species as capital breeders and Bond and Diamond (2010) as income breeders. According to the continuum hypothesis, differences could occur due to local adaptations. The species feeds mainly on small fish and crustaceans and is considered a top predator in the marine food web (Dittmann et al. 2011). The Eurasian oystercatcher is also a migratory species, but is regarded as a capital breeder. It mainly feeds on macrozoobenthic organisms, such as mussels and worms (Dittmann et al. 2011).

16.1 Temporal variations of contaminants in bird eggs

The Common tern and the Eurasian oystercatcher eggs were collected at Tjärnö and analysed between 2011 (or 2012) and 2017, so the presented data is based upon only a few, mostly six, years. Due to the few data points, the time trends should be considered as a first indication only. The figures of all compounds can be found in the Appendix in the according chapter of the substance class of interest.

For half of the analysed **metals** (zinc, selenium, copper, cadmium and silver), no time trends could be detected for any of the bird species' eggs (e.g. for cadmium see Figure 10).

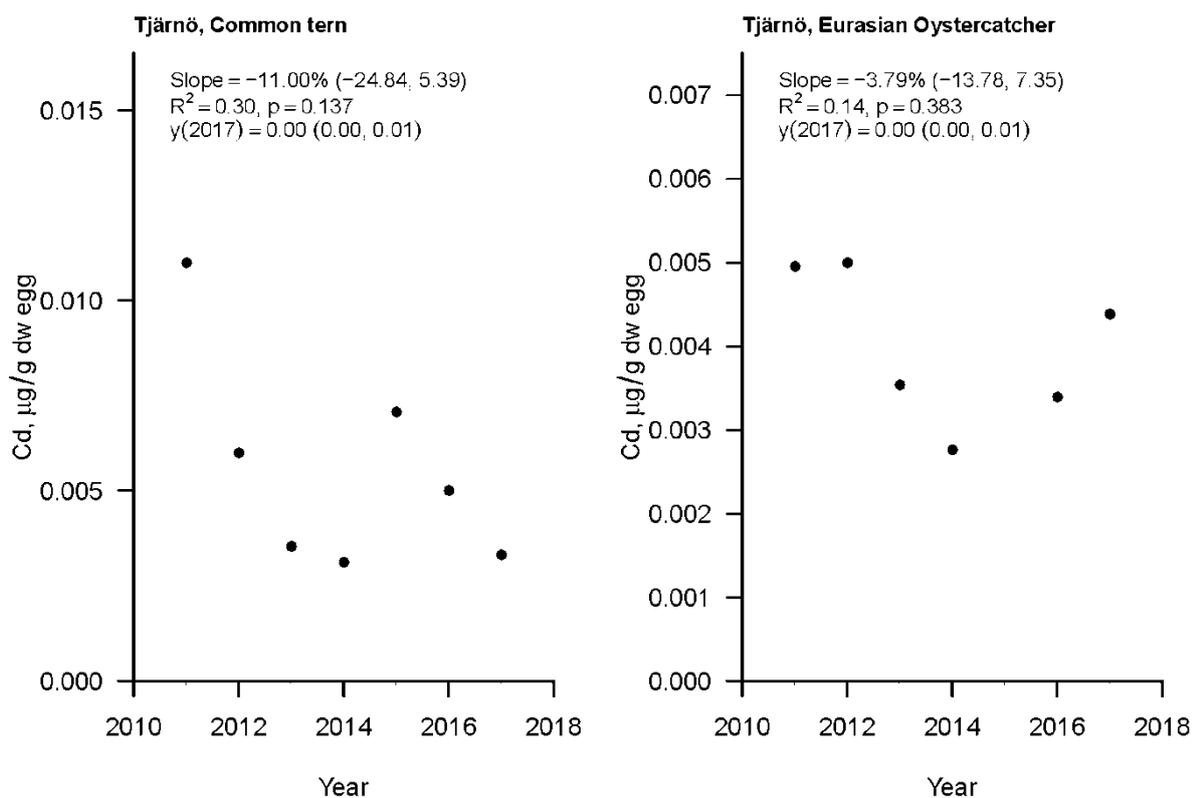


Figure 10: Time trends of cadmium in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per dry weight.

The measured concentrations of these compounds are also similar for the two species. For lead, there is a downward trend in Common tern eggs, with concentrations lying below the LOQ during the last five years, while no trend is seen for lead in Eurasian oystercatcher eggs. The concentrations of lead are approximately 5-times higher in the Eurasian oystercatcher eggs compared to the Common tern. The mercury concentration over time in Common tern eggs is close to a significant downward trend, while there is no trend in Eurasian oystercatcher eggs; concentrations of the two species were similar (Figure 11).

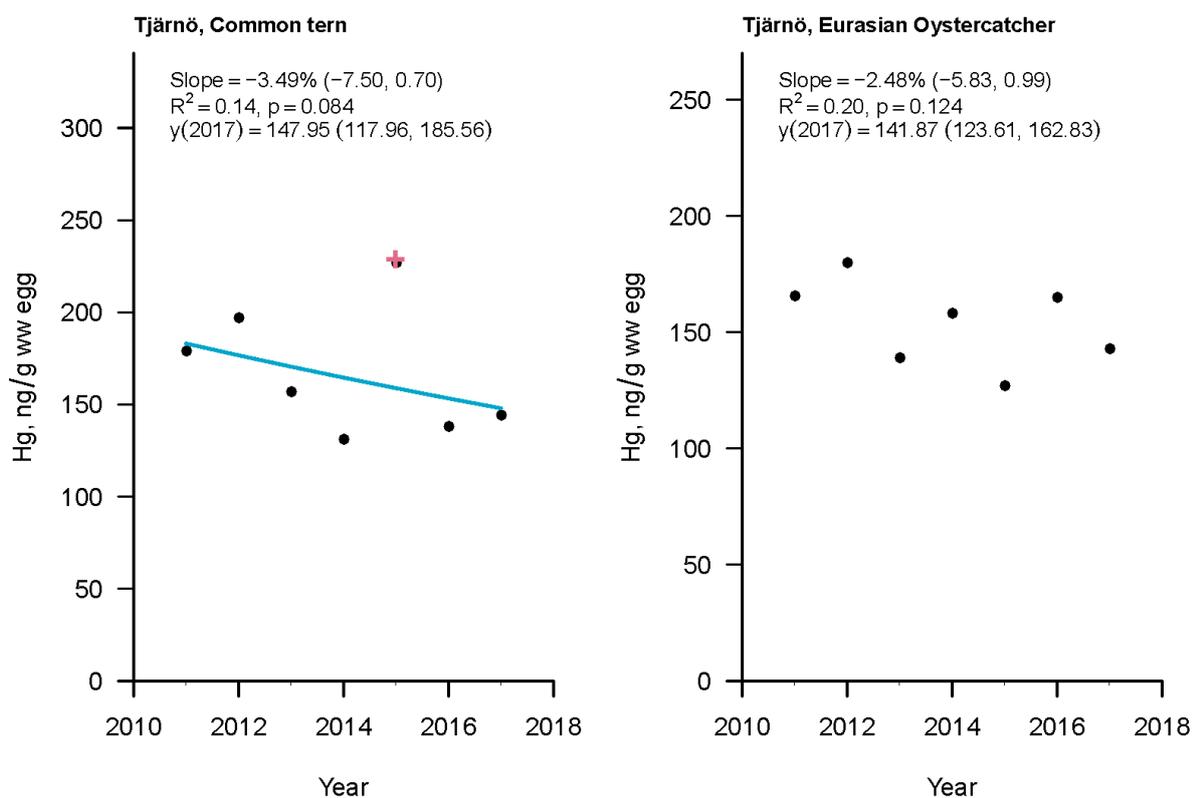


Figure 11: Time trends of mercury in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per wet weight.

For nickel, concentrations are decreasing in Common tern eggs, but not in Eurasian oystercatcher eggs; concentrations are similar for the two species. Chromium is the only metal for which the concentrations show downward trends in both species with concentrations in a similar range. Concentrations of arsenic in Common tern eggs show a significant downward trend, while in the Eurasian oystercatcher eggs, there is a close to significant upward trend. The arsenic concentrations are slightly higher in Eurasian oystercatcher eggs over time than for Common tern. The concentrations found in the eggs of Common tern and Eurasian oystercatcher are in most cases comparable with concentrations found in Guillemot eggs, with the exception of silver and arsenic, which were slightly lower in Guillemot eggs.

For most of the analysed **PCBs**, no time trends exist with the exception of PCB-52 and PCB-77, for which significant downward trends are detected, only for Common tern eggs (PCB-77, see Figure 12). For most PCBs, the concentrations are similar for the two species; exceptions are PCB-77 and PCB-81, for which concentrations are twice as high in Eurasian oystercatcher compared to the Common tern eggs.

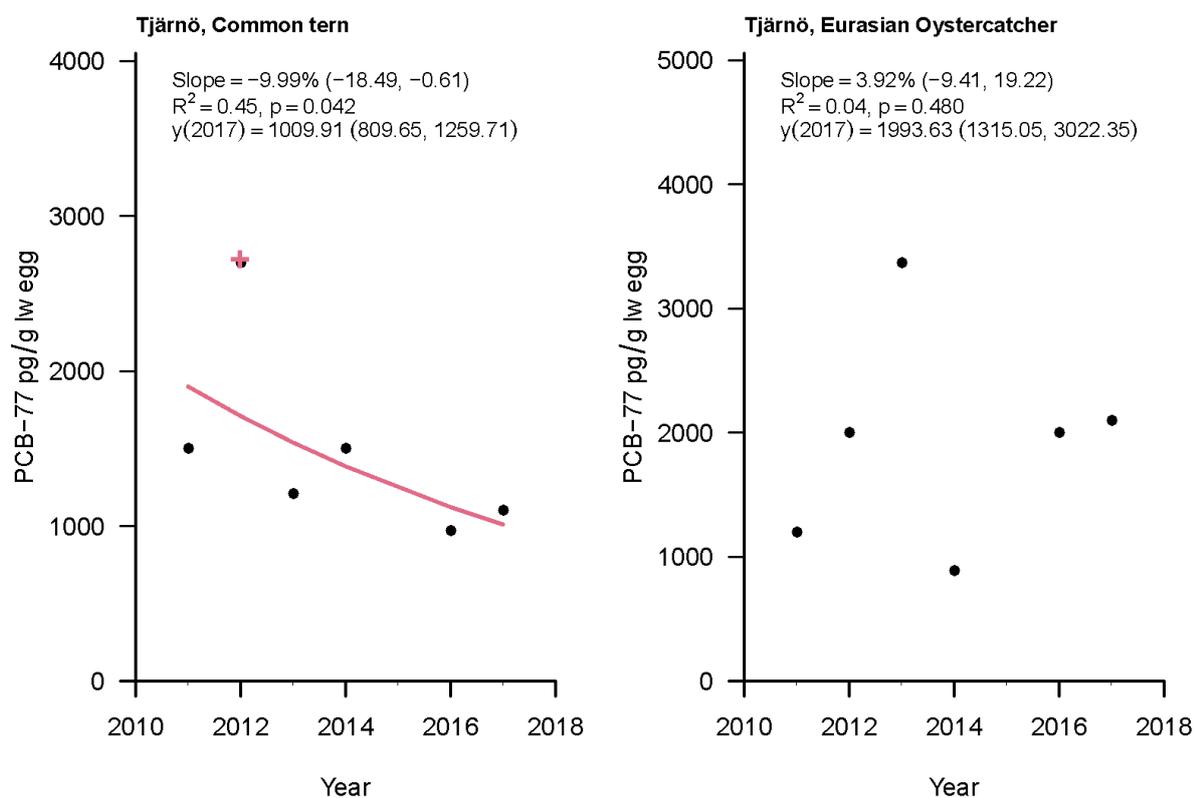


Figure 12: Time trends of PCB-77 in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

Concentrations in Guillemot egg from the Baltic Sea are generally higher than in Common tern and the Eurasian oystercatcher eggs; the extend is dependent on the PCB-congener, but goes up to an order of magnitude.

Among the **chlorinated pesticides**, DDE concentrations do not show any trends and concentrations are slightly higher in Common tern eggs compared to the Eurasian oystercatcher eggs (Figure 13).

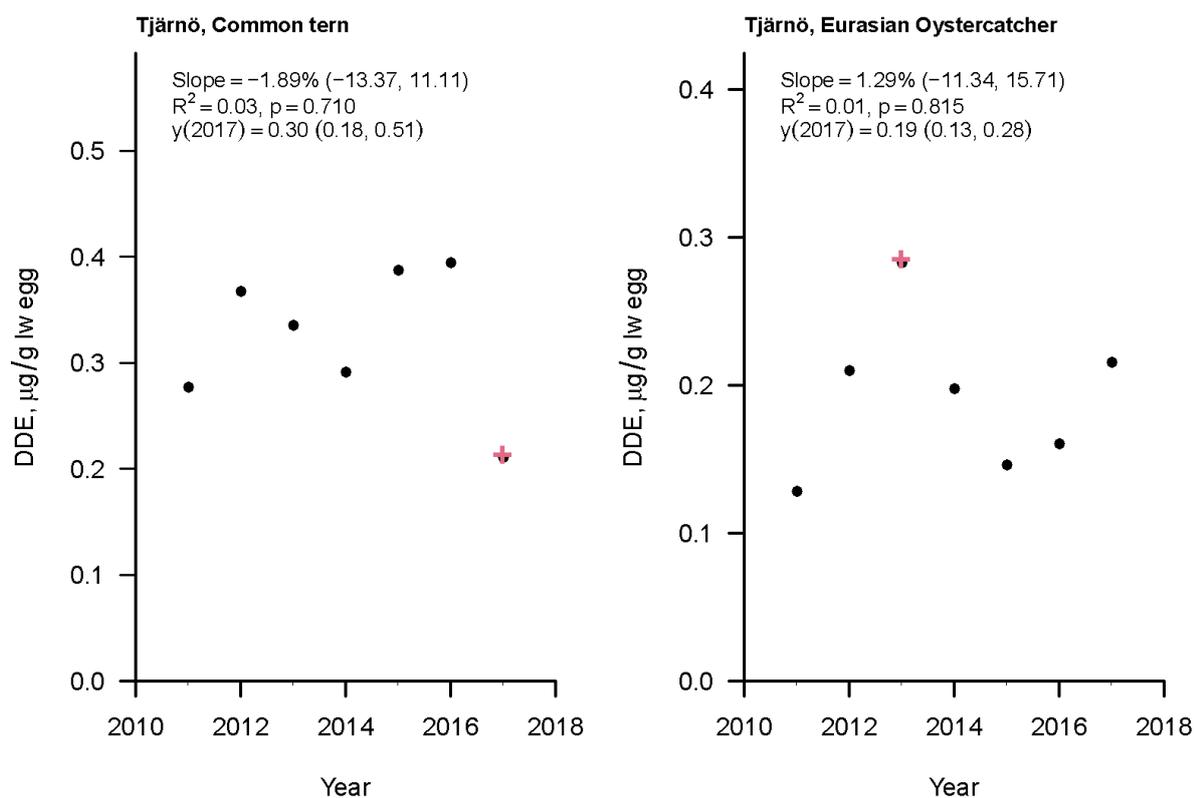


Figure 13: Time trends of DDE in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

DDD, DDT, Lindane and α -HCH are below the LOQ in both species for most years of the monitoring period. β -HCH concentrations, on the other hand, show a downward trend in Common tern eggs while no trend is seen in Eurasian oystercatcher eggs (Figure 14). Concentrations of β -HCH are similar for the two species. For HCB, there are no time trends. However, the HCB concentrations are more than twice as high in Common tern eggs compared to the Eurasian oystercatcher eggs. Concentrations of DDE, HCB and β -HCH are higher in Guillemot eggs compared to Common tern and Eurasian oystercatcher, with up to 20 times higher depending on compound.

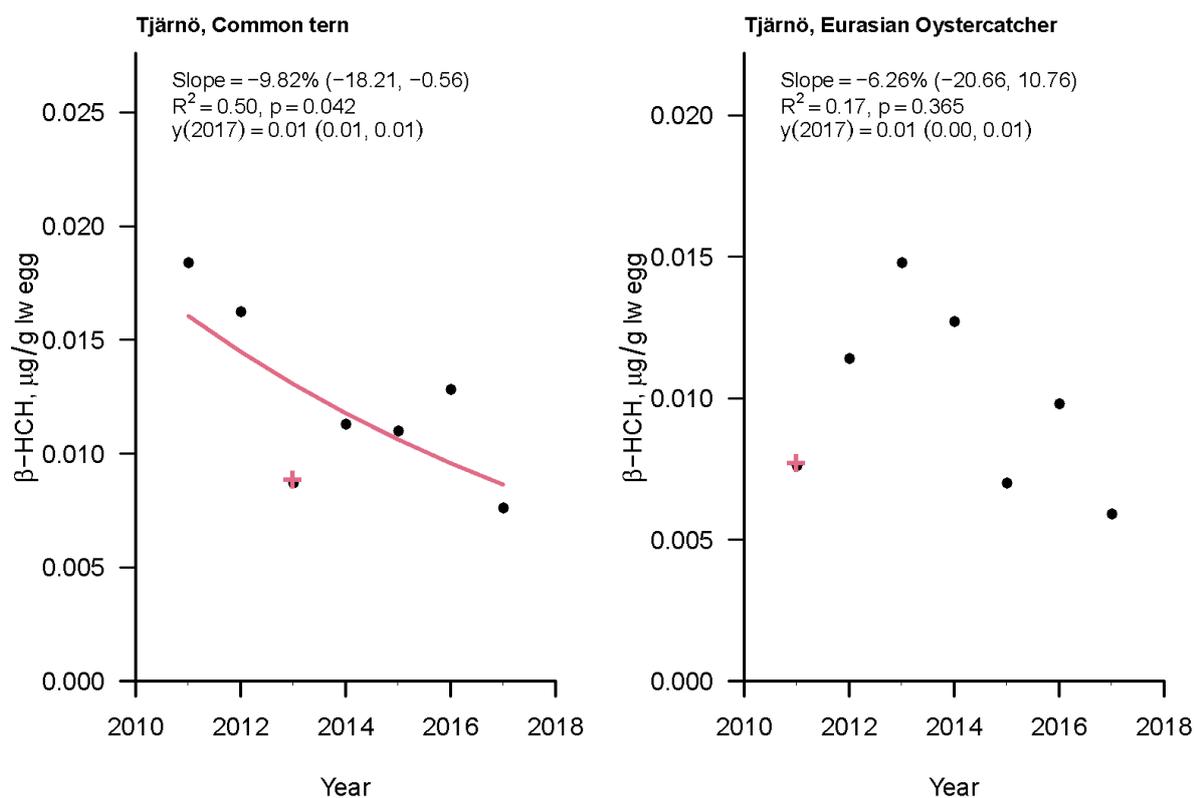


Figure 14: Time trends of β -HCH in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

Generally, many PCDD/F compound concentrations are significantly increasing in Eurasian oystercatcher eggs and concentrations in this species are at the most between 2- and 5-times higher than in the Common tern eggs. However, for 2,3,7,8-TCDF and 1,2,3,7,8-PeCDF, the Eurasian oystercatcher eggs have 10- and 15-times higher concentrations, respectively (see TCDF in Figure 15). Concentrations of PCDD/Fs in Guillemot eggs are generally higher compared to Eurasian oystercatcher eggs, with up to an order of magnitude higher.

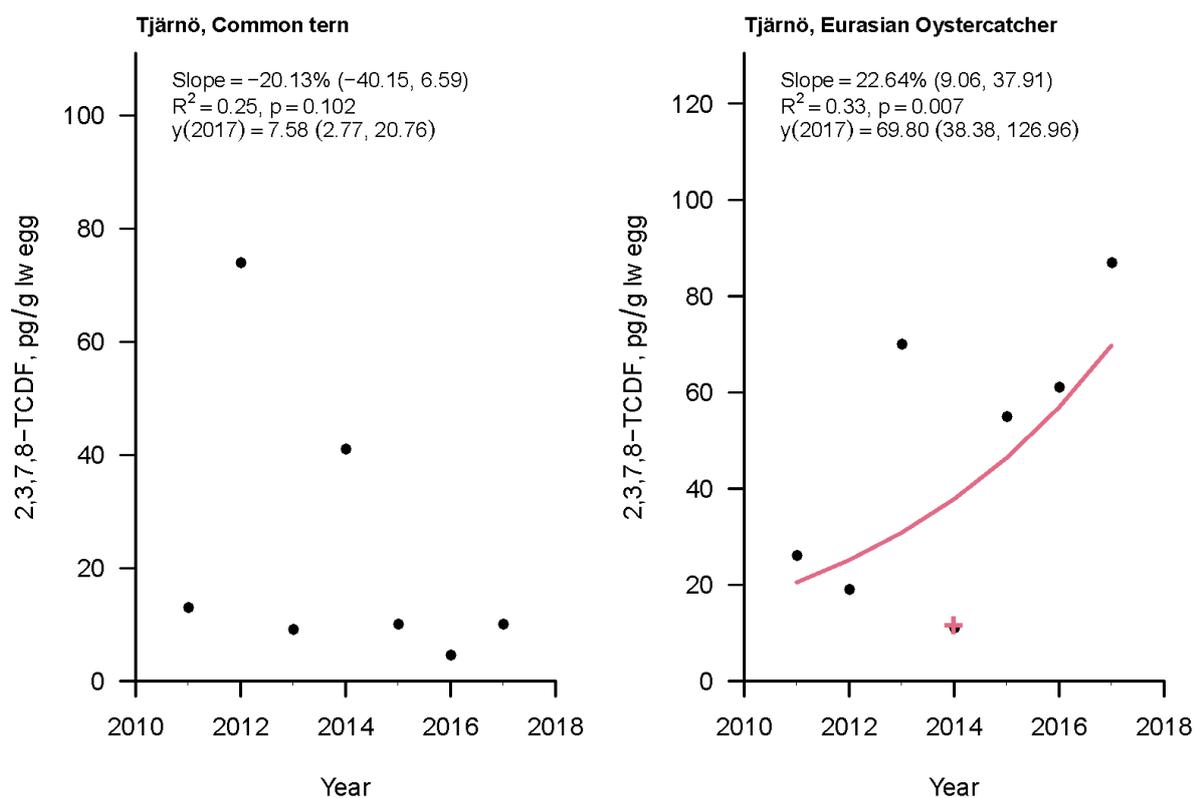


Figure 15: Time trends of 2,3,6,8-TCDF in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

Several of the dioxins and furans with the same number of chlorines had similar patterns over the years. Upward trends in Eurasian oystercatcher egg concentrations are seen for TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDD, and TCDD-equivalents (TEQ) (1,2,3,4,6,7,8-HpCDF see Figure 16, TEQs see Figure 17). In Common tern eggs, downward trends are seen for 1,2,3,4,6,7,8-HpCDF and 1,2,3,4,6,7,8,9-OCDD (1,2,3,4,6,7,8-HpCDF see Figure 16). The 1,2,3,4,7,8,9-HpCDF and 1,2,3,4,6,7,8,9-OCDF concentrations were mostly lying below the LOQ in both species.

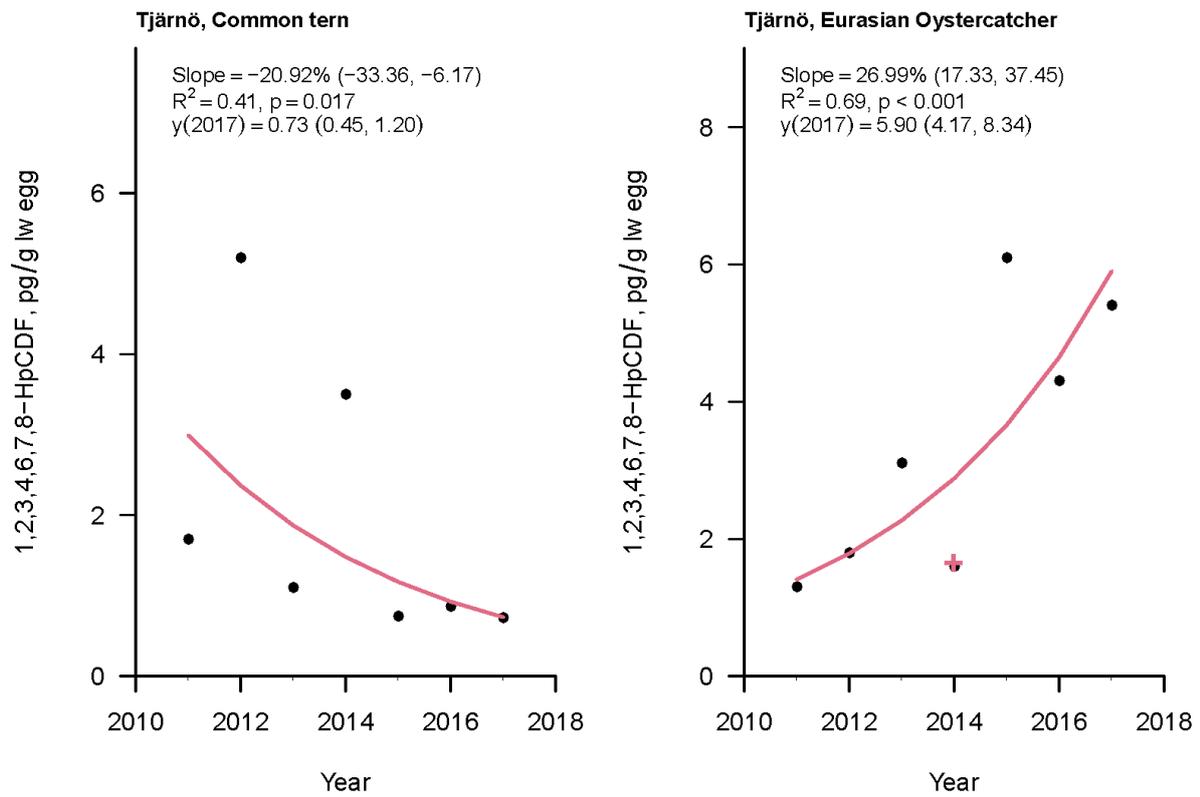


Figure 16: Time trends of 1,2,3,4,6,7,8-HpCDF in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

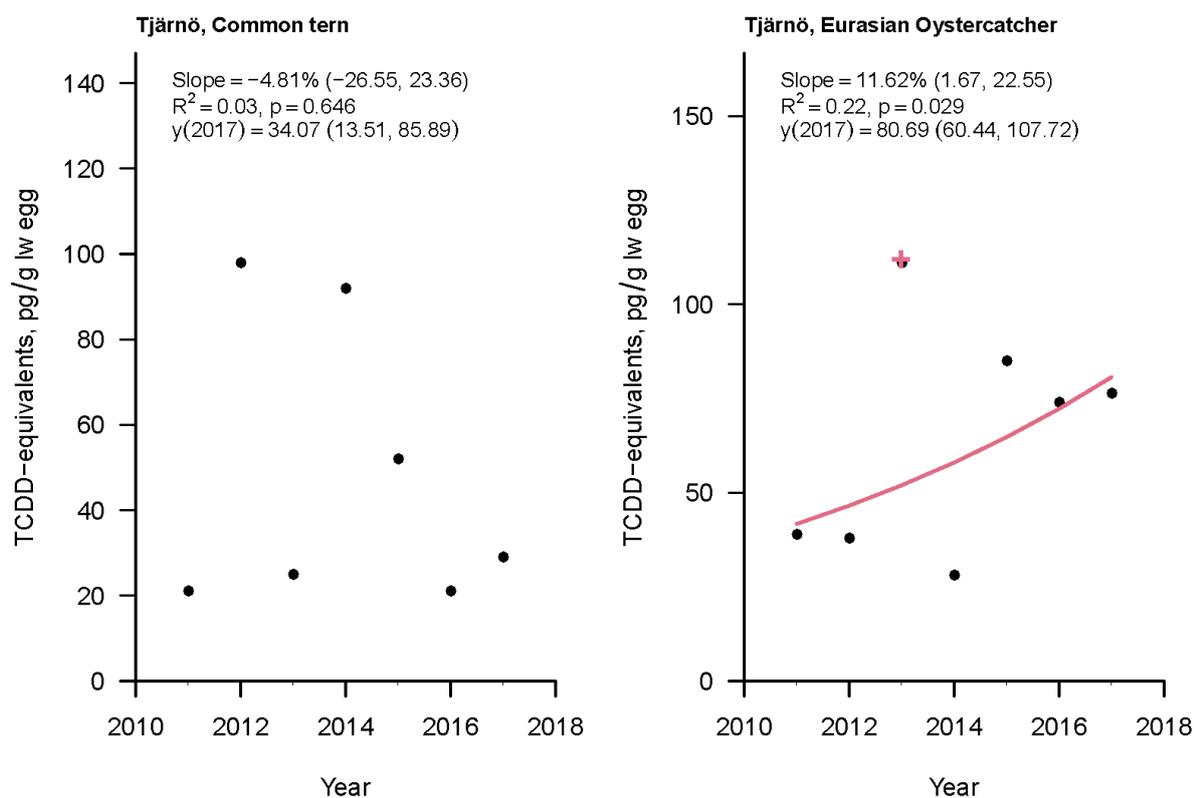


Figure 17: Time trends of TCDD-equivalents (TEQs) in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

For **brominated flame retardants**, concentrations in the Common tern eggs seem higher compared to the Eurasian oystercatcher eggs concentrations, especially for BDE-47, BDE-99, and BDE-100 (Figure 18). The only significant detected time trend (decreasing) existed for HBCDD in Common tern eggs (Figure 19). In Guillemot eggs concentrations of BDE-47 is halved compared to Common tern and concentrations of BDE-99 and BDE-100 in Guillemot eggs are similar as in Eurasian oystercatcher, in contrast the concentration of HBCDD is almost 50 times higher in Guillemot eggs compared to the other two bird species.

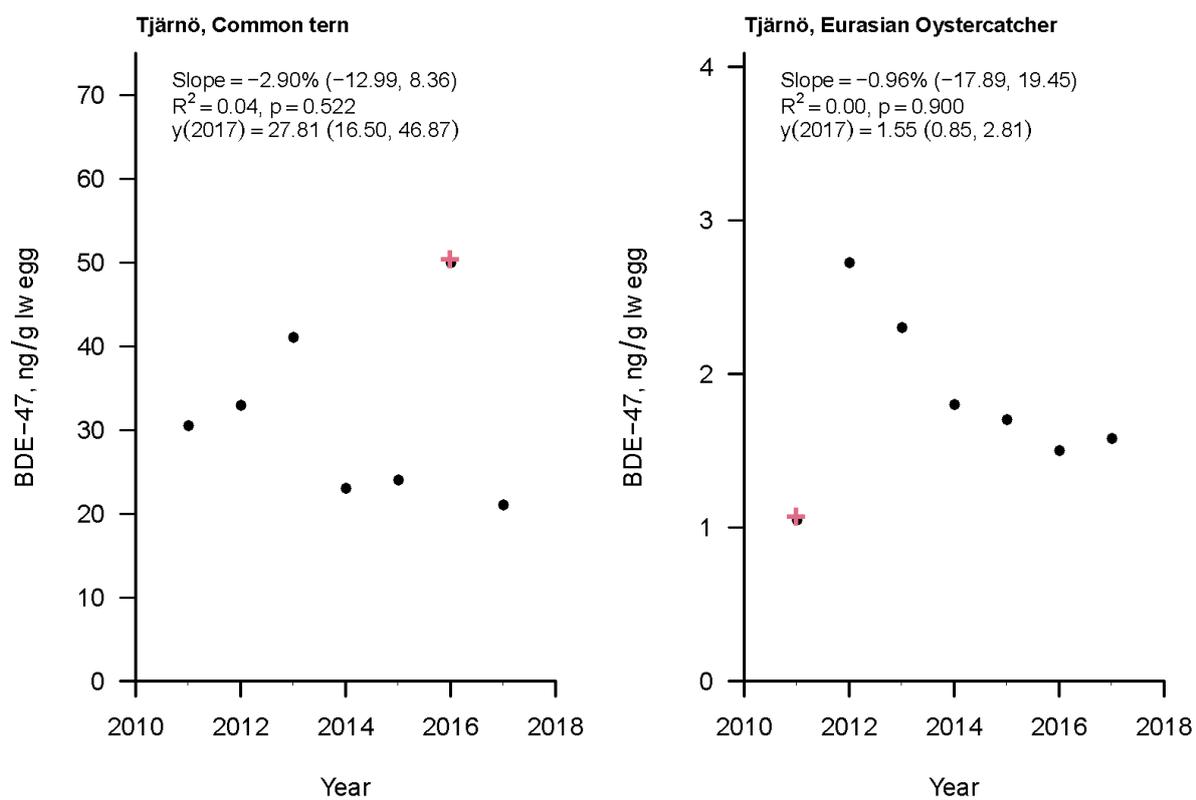


Figure 18: Time trends of BDE-47 in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

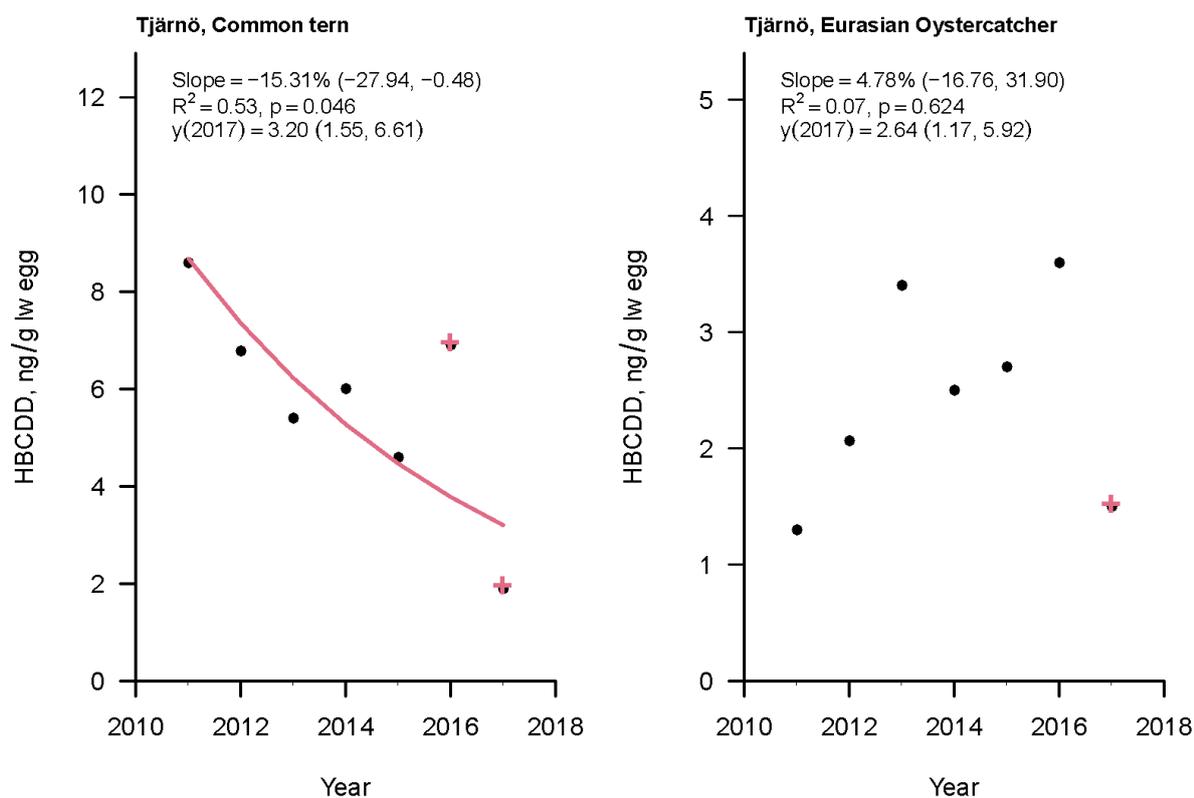


Figure 19: Time trends of HBCDD in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per lipid weight.

For **PFASs**, PFHxA, PFHpA, and PFBS, concentrations are below the LOQ during all years in both species, and for PFDS, PFPeDA, and branched FOSA some of the years' concentrations are below LOQ.

For most **PFCAs**, there are downward trends. PFOA shows downward trends in both species, with concentration below the LOQ for some years in Common tern eggs (Figure 20).

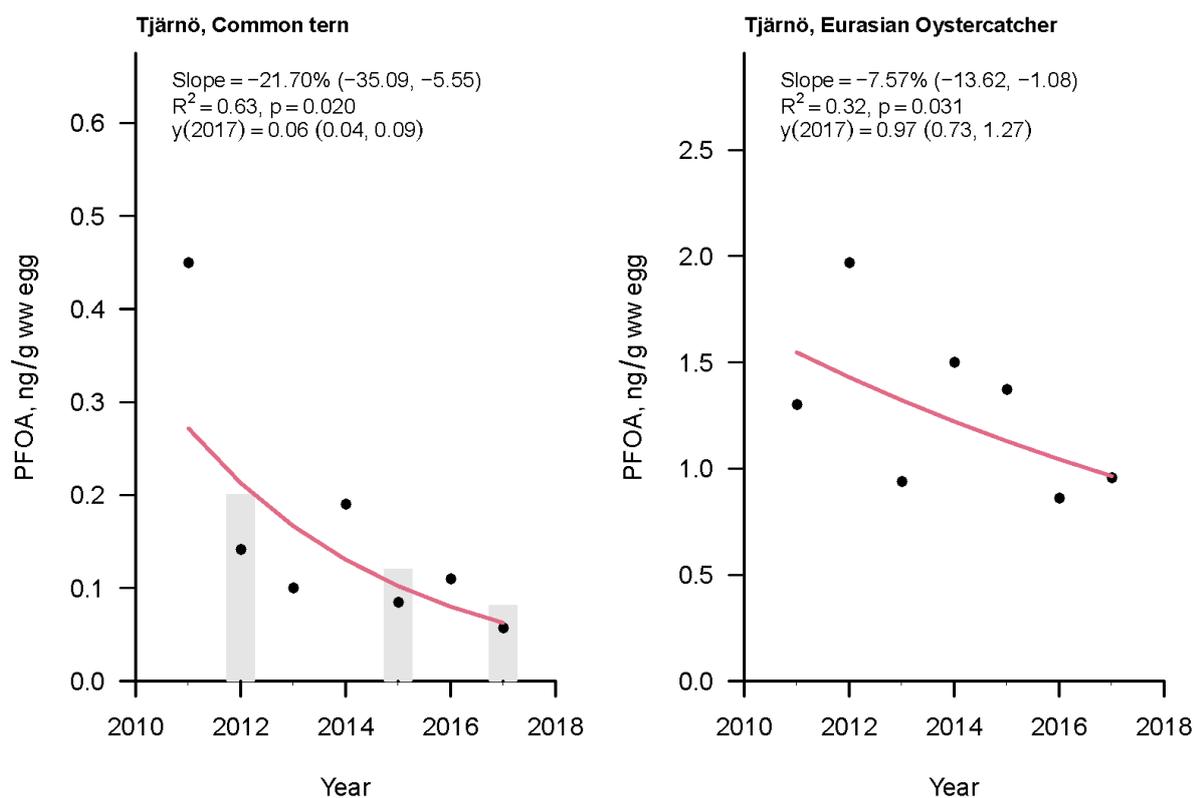


Figure 20: Time trends of PFOA in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per wet weight.

PFNA and PFUnDA (Figure 21) also show downward trends in both species and for PFDA, there is close to significant downward trends in both species' eggs.

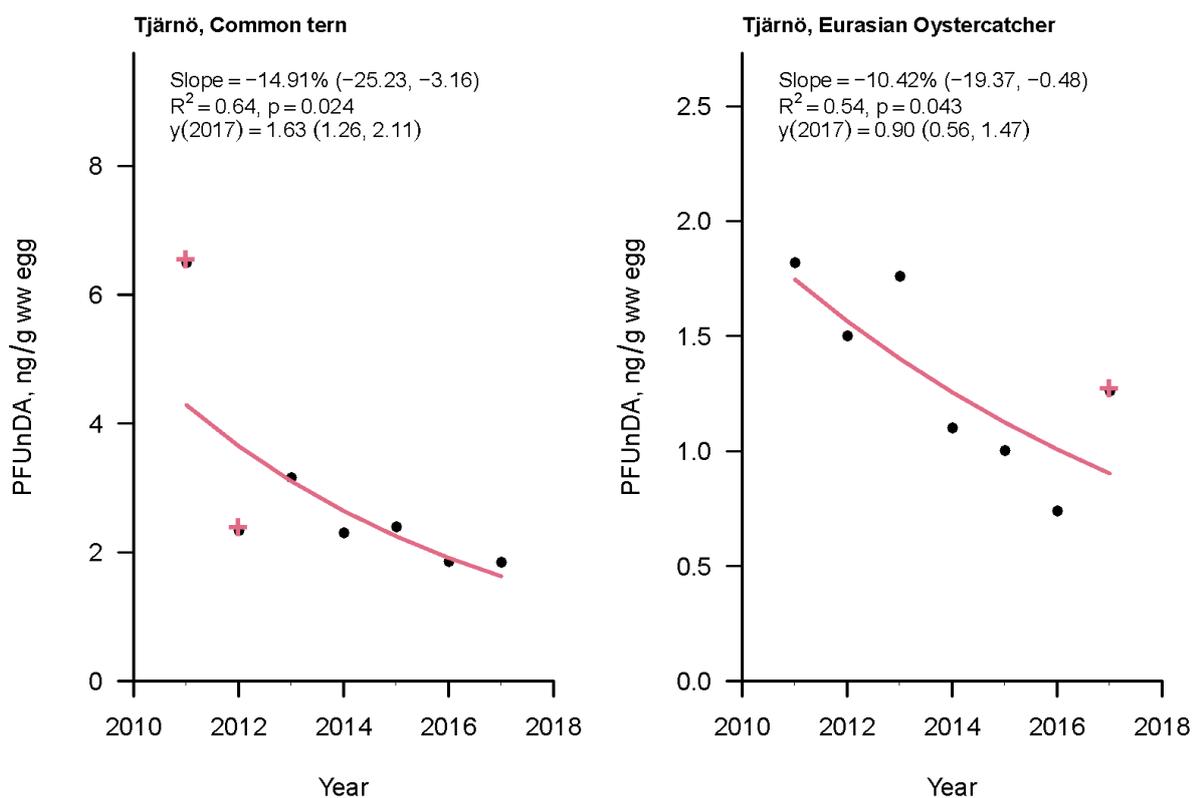


Figure 21: Time trends of PFUnDA in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per wet weight.

The concentrations of PFDoDA over time indicate a downward trend in Common tern while concentrations of PFTrDA are decreasing in Eurasian oystercatcher. For PFTeDA there are no trends in any of the species.

There are no general pattern regarding concentration differences in PFCA's between the two bird species; PFOA, PFNA, and PFTrDA are higher in Eurasian oystercatcher, while PFDA and PFUnDA are higher in Common tern eggs. PFDoDA and PFTeDA show similar concentrations in the bird eggs. For all PFCA's, concentrations in Guillemot eggs are much higher compared to Eurasian oystercatcher and Common tern.

PFSA's concentrations display downward time trends. Sum PFHxS concentrations (Figure 22) are decreasing in both species and the last year's concentration in Eurasian oystercatcher is approximately 10-times higher compared to Common tern.

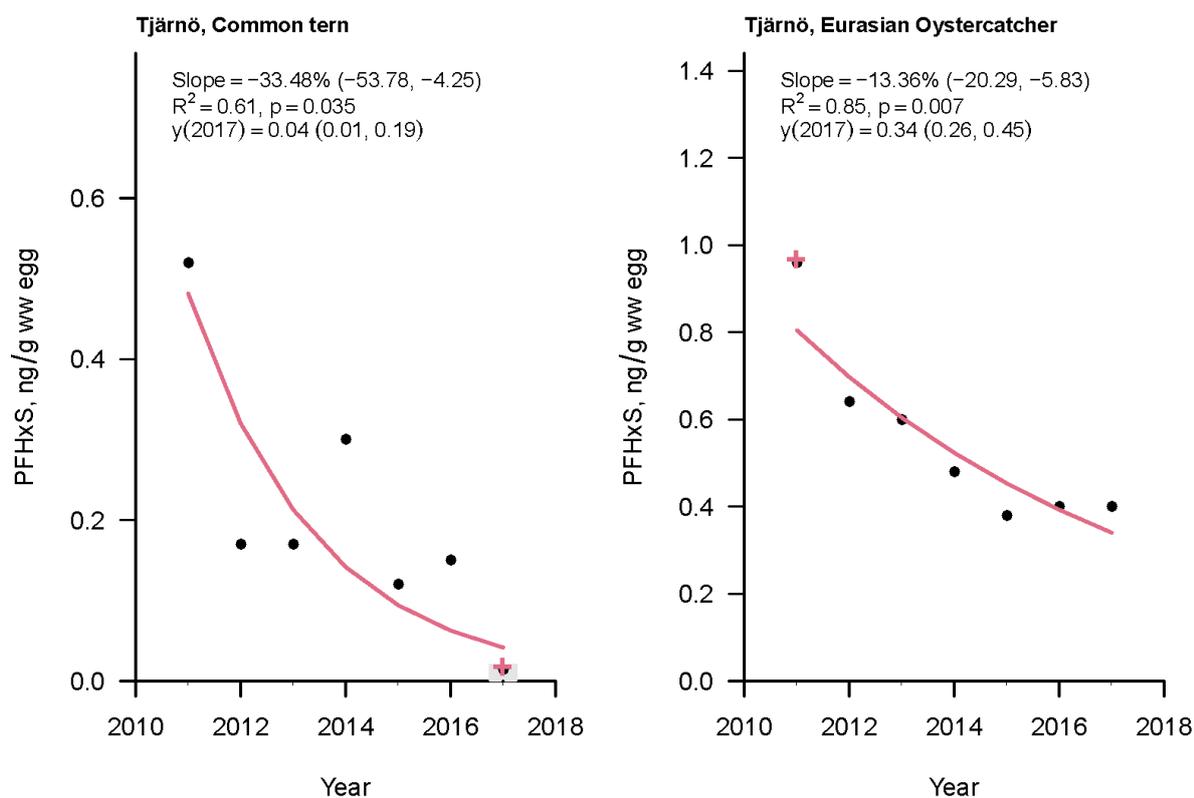


Figure 22: Time trends of PFHxS in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per wet weight.

Sum PFOS (Figure 23) and linear PFOS concentrations are decreasing in both species and concentrations are similar among the two species for sum PFOS, or slightly higher for Common tern in case of linear PFOS. The branched PFOS concentration is on the other hand only decreasing in Common tern eggs and the concentration is more than 3-times higher in Eurasian oystercatcher eggs. The concentration of sum PFOS in Guillemot eggs are almost two order of magnitude higher compared to Eurasian oystercatcher and Common tern.

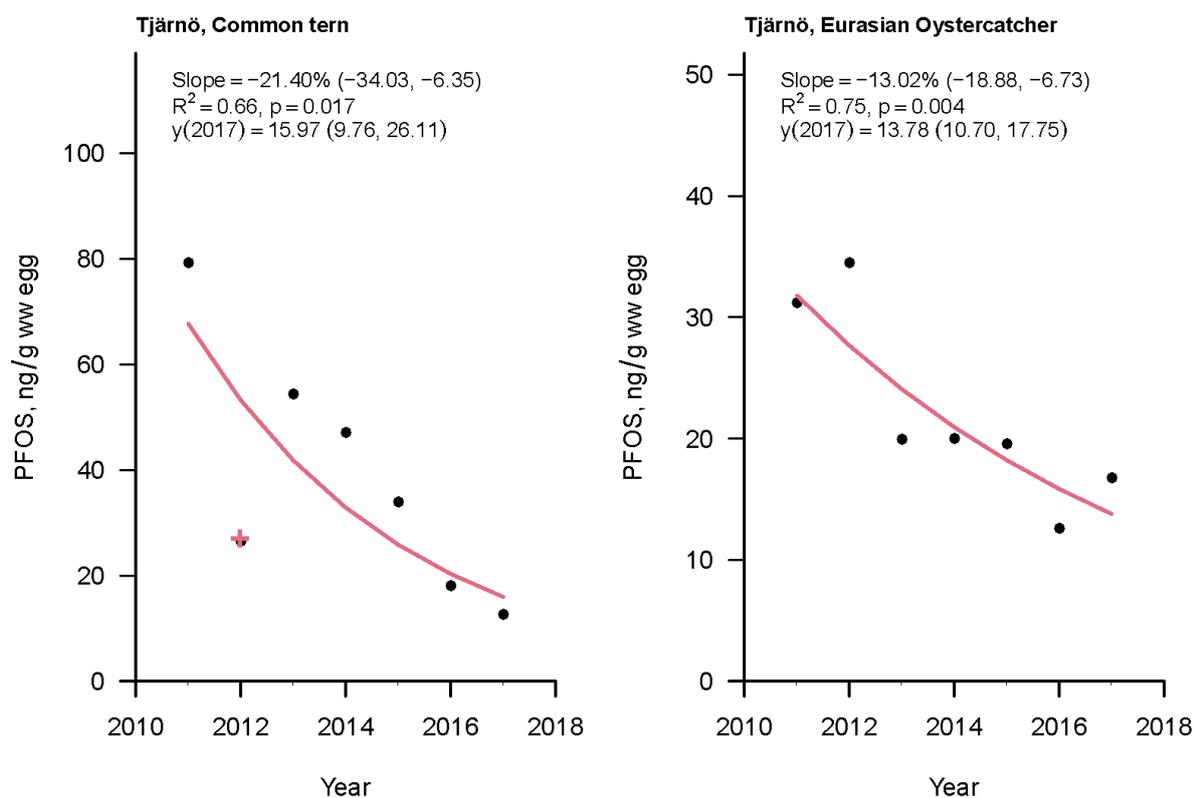


Figure 23: Time trends of PFOS in Common tern and Eurasian oystercatcher eggs from Tjärnö over the entire sampling period, concentrations given per wet weight.

Sum FOSA and linear FOSA show downward trends in Common tern; for sum FOSA the concentration is twice as high in Eurasian oystercatcher, while for linear FOSA concentrations are similar for the species. Sum FOSA concentrations in Eurasian oystercatcher were highly influenced by an extremely outlying high concentration in 2014 among the branched isomers (otherwise on a constant low level, even partly below the LOQ).

In general, many of the compounds within the Swedish National Monitoring Programme of Contaminants in marina biota could be detected in Common tern and Eurasian oystercatcher. Concentrations of several of the contaminants are similar in the two species, but there are exceptions among each contaminant class and even opposing time trends for some contaminants. There are clear differences in contaminant concentrations between species for PCDD/Fs and BFRs; for most PCDD/Fs, concentrations are higher in Eurasian oystercatcher compared to Common tern, while for the BFRs concentrations are higher in Common tern (with the exception of HBCDD).

Dioxins are sediment-bound and Eurasian oystercatcher mainly feeds on macrozoobenthos in the sediment. Common tern, on the other hand, feeds mainly on fish and crustaceans that inhabit the water phase and are therefore likely exposed to a lower amount of dioxins in direct comparison to Eurasian oystercatcher.

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