Report to the Swedish EPA (the Health-Related Environmental Monitoring Program)

Temporal trends of Swedish environmental and human milk concentrations of dioxins, furans and dioxin-like PCBs, with forecasts to 2040

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Introduction

The report deals with temporal trends of measured concentrations of representative congeners of dioxin-like PCBs, dioxins (PCDDs) and furans (PCDFs) (here together called dioxins) in biological samples from the Swedish marine and freshwater environment together with human milk and food.

Dioxins are well-known for causing cancer, severe reproductive and developmental problems, damage the immune system and for interfering with hormonal systems. Dioxin exposure has further been suggested to induce birth defects, reduced sperm counts, diabetes, learning disabilities, immune system suppression, lung problems, skin disorders, lowered testosterone levels and more.

The data originates from national monitoring programs on contaminants financed by the Swedish Environmental Protection Agency (EPA) and accomplished by the Swedish Food

Agency (SFA), the Swedish Museum of Natural History (SMNH) and Stockholm University (SU). Temporal trends of the abovementioned chemicals have been studied, also, the trends have been extrapolated in an attempt to forecast the situation during the coming 20 years.

The present report focus on temporal trends and longer time series (often more than 2 decades). Compilations that covers also shorter time-series, relations between various biological samples and geographical distribution can be found elsewhere. A thorough study describing the dioxin situation in the Baltic was carried out by the Swedish EPA 2013 (Wiberg *et al.*, 2013).

Material and Methods

Data from studies on human milk (Gyllenhammar *et al.*, 2017 and Nyberg *et al.*, 2017), food baskets (Livsmedelsverket, 2017), wild species from the Baltic and Swedish west coast like guillemot, herring and from freshwater, pike and perch have been considered for temporal trend analyses (Bignert *et al.* 2018, Nyberg *et al.*, 2018). Dioxin data from cod and blue mussels are not available, 10 - 11 years of dioxins in perch from the Baltic are available but the time-series are too short and variable to make any meaningful trend assessments or predictions.

Trends for human milk samples from Uppsala are based on individual samples whereas the human milk trends from Stockholm and Göteborg are based on pooled samples containing often 10 individuals, never less than 5 (but often substantially more individuals).

TCDD or 2,3,7,8-tetrachlorodibenzo-p-dioxin is among the most toxic chemicals known, but other substances with similar structures, also contribute to the toxic effects of dioxin and dioxin-like substances. To estimate the combined effect of these compounds, toxicity equivalent factors (TEF) (Table 1) are multiplied with the measured concentrations of dioxin-like PCBs, furans (PCDFs) and dioxins (PCDDs) before adding them and the sum is then expressed as a TCDD equivalent concentrations (TEQ concentrations).

Not included here are e.g. poly*brominated* dioxins (PBDDs) that are assumed to contribute to total dioxin-TEQ exposure. The toxicity of PBDDs closely resembles that of the PCDD/Fs as they share a common mechanism of action. In Baltic Proper littoral fish, the levels of PBDDs may exceed those of their chlorinated analogues (PCDDs) and the levels of PBDDs are high in mussels, which possibly increase over time (Haglund *et al.*, 2007).

All concentrations are given as TEQ-values on a lipid weight basis to enable comparisons among matrices. Note that fat content differ a lot between fish/seafood species. Pike and perch are lean species with a fat content less than 1 %, hence the fresh weight TEQ concentrations are considerably lower than in fat-rich species, although the concentrations are the same on a fat weight basis.

The primary assumption is that an ongoing trend follows an exponential path i.e. that concentrations measured at a year following another year has changed a certain percentage of the previous year. This assumption may be fair when the usage of a substance has stopped after a ban or when the production ceases after measured have been taken to reduce discharges of the chemical.

Estimated concentration from the log-linear regression line for the last year of data is reported as e.g. y(17)= 0.60 (0.57,0.63), means that last year's (2017) concentration is estimated to 0.60 with a 95% confidence interval between 0.57 and 0.63.

To forecast future years concentrations, assuming a continuous change at the same rate as during the time series, the regression line has been extrapolated to year 2040, and the concentrations from the upper 95% prediction(population) interval of the regression line at year 2027, 2030 and 2040 are reported (Y(27)=, Y(30)= and Y(40)=). This implies that we assume that 97.5% of the monitored population is below the upper interval.

To consider possible deviations from log-linear trends, a change-point analysis was also carried out. Change-points (CPs) were identified by a technique similar to that reported by Sturludottir *et al.*, 2017. Shown in the figures as CP=year and p-value for the CP-test.

In addition, the regression lines for the two periods 1998 - 2006 and 2007 - 2016 were compared if sufficient data from the two periods were available. If the slope of the regression lines (% change/year) significantly differed from each other, and/or a significant change-point was detected, the forecast from the extrapolation can be questioned. It should be stressed though that in many cases, splitting the time-series in two parts often results in rather short series. The results from this test are reported as b_1=b_2, p=, b_1=, b_2= , the first p-value show if the slopes from the two periods were significantly different, b_1 and b_2 the slope of the first and second period respectively.

The quality of the time-series has been evaluated by estimating the smallest trend possible to detect with an 80% statistical power both for the current time-series (LDT(c)) and for a monitoring period of 10 (LDT(d))years and also the n of years required (YRQ) to detect a yearly trend of 5% with an 80% power.

In 2018, the European Food Safety Authority issued a tolerable weekly intake (TWI) of dioxin-like PCBs, PCDDs and PCDFs of 2 pg TEQ/kg body weight/week (EFSA, 2018). In the present report, a target value of 5.9 pg TEQ/g lipid in mother´s milk was used, representing the TEQ concentration reached after maternal TEQ intake at the TWI for 35 years (EFSA, 2018). This target value was used in the extrapolations of temporal trends of total TEQs in mother´s milk. In the extrapolations, the upper 95% prediction (population) interval was used for the sum of TEQ (non-ortho-PCBs + mono-ortho-PCBs + PCDFs + PCDDs). To account for populations with higher exposure than the mothers from Uppsala, imaginary populations being exposed to twice as high dioxin TEQ concentrations as the mothers from Uppsala were also considered.

Further detailed information on material and methods is found in Glynn *et al.* (2020).

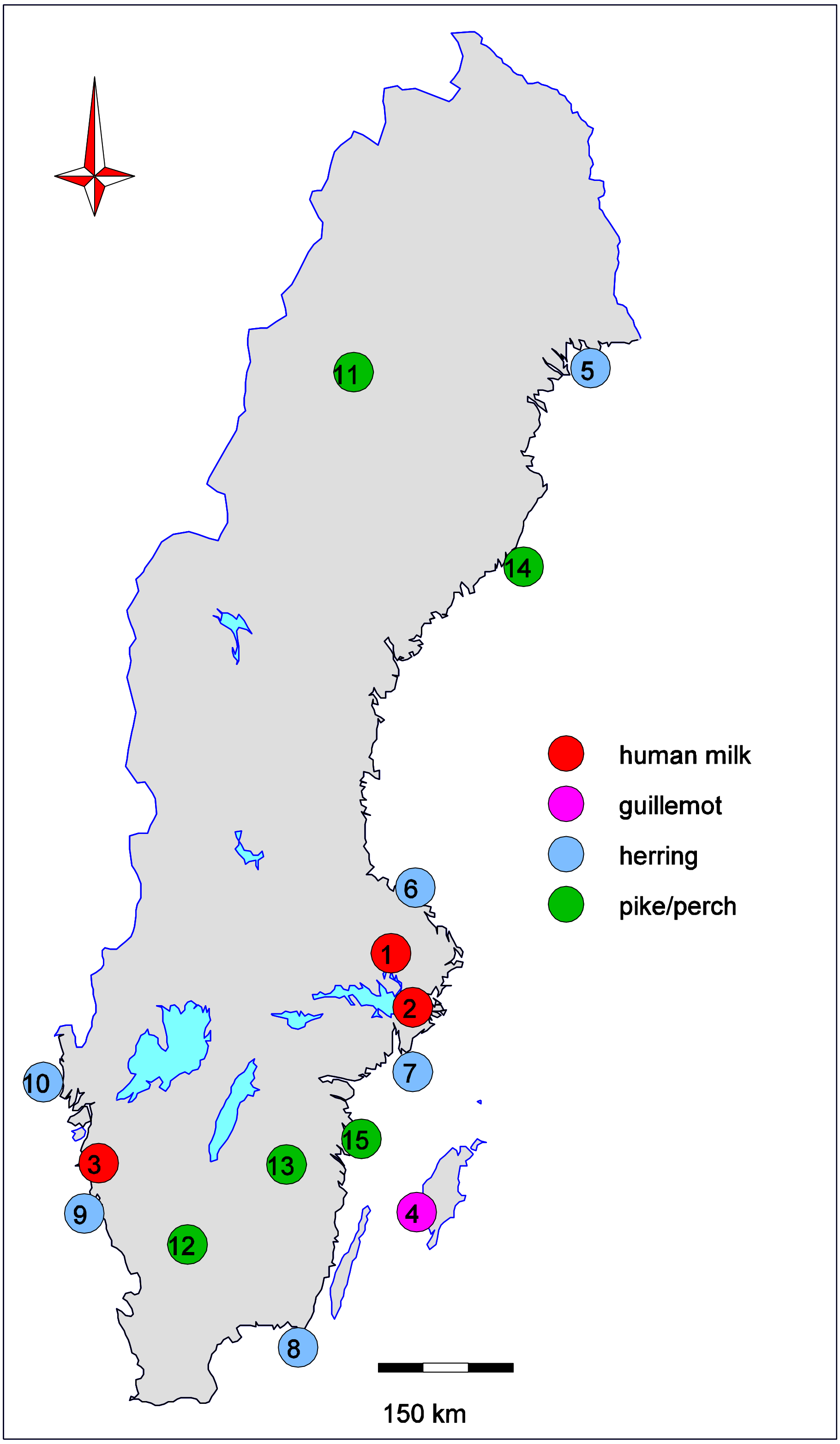

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Figure 1.1. Map showing the various sampling sites. 1=Uppsala, 2=Stockholm, 3=Göteborg, 4=St Karlsö, 5=Harufjärden, 6=Ängskärsklubb, 7=Landsort, 8=Karlskrona, 9=Fladen, 10=Väderöarna, 11=Storvindeln (pike), 12=Bolmen (pike), 13=Skärgölen (perch), 14=Holmöarna (perch), 15=Kvädöfjärden (perch)

Substance	WHO-2005	Substance	WHO-2005
Dibenso-*p*-dioxins		**Dibenso-furans**	
2,3,7,8-TCDD	1	2,3,7,8-TCDF	0.1
1,2,3,7,8-PeCDD	1	1,2,3,7,8-PeCDF	0.03
1,2,3,4,7,8-HxCDD	0.1	2,3,4,7,8-PeCDF	0.3
1,2,3,6,7,8-HxCDD	0.1	1,2,3,4,7,8-HxCDF	0.1
1,2,3,7,8,9-HxCDD	0.1	1,2,3,6,7,8-HxCDF	0.1
1,2,3,4,6,7,8-HpCDD	0.01	1,2,3,7,8,9-HxCDF	0.1
OCDD	0.0003	2,3,4,6,7,8-HxCDF	0.1
		1,2,3,4,6,7,8-HpCDF	0.01
		1,2,3,4,7,8,9-HpCDF	0.01
		ODCF	0.0003
		Mono-*ortho* PCBs	
Non-*ortho* PCBs		PCB 105	0.00003
PCB-77	0.0001	PCB 114	0.00003
PCB-81	0.0003	PCB 118	0.00003
PCB-126	0.1	PCB 123	0.00003
PCB-169	0.03	PCB 156	0.00003
		PCB 157	0.00003
		PCB 167	0.00003
		PCB 189	0.00003

Table 1. TEF-values (Toxicity Equivalent Factors) (US EPA, 2010) used in the following TEQ calculations.

Results

The results are shown as figures with an explanation and evaluation below.

1 Relative contributions to the sum of TEQ

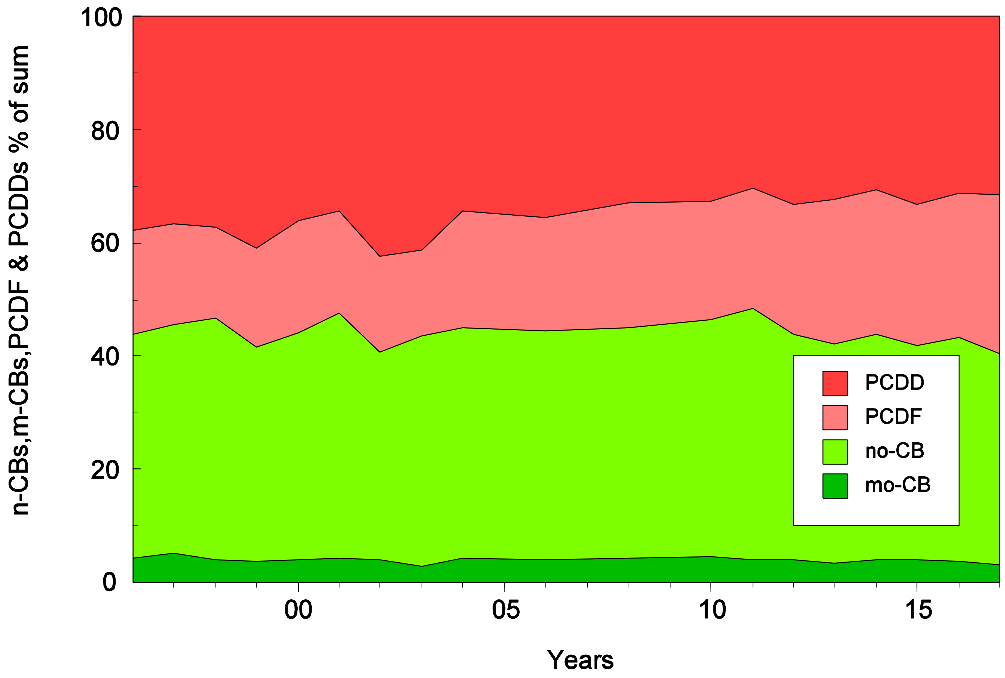


Figure 1.2. Human milk 1996-2017 from Uppsala. Relative contributions to the sum of TEQ from mo-PCBs (~5%), no-PCBs (~35%), PCDFs (~25%) and PCDDs (~35%). The relative contributions have been relatively constant over time except that the contribution from the PCDFs has increased somewhat during recent years. See more details below.

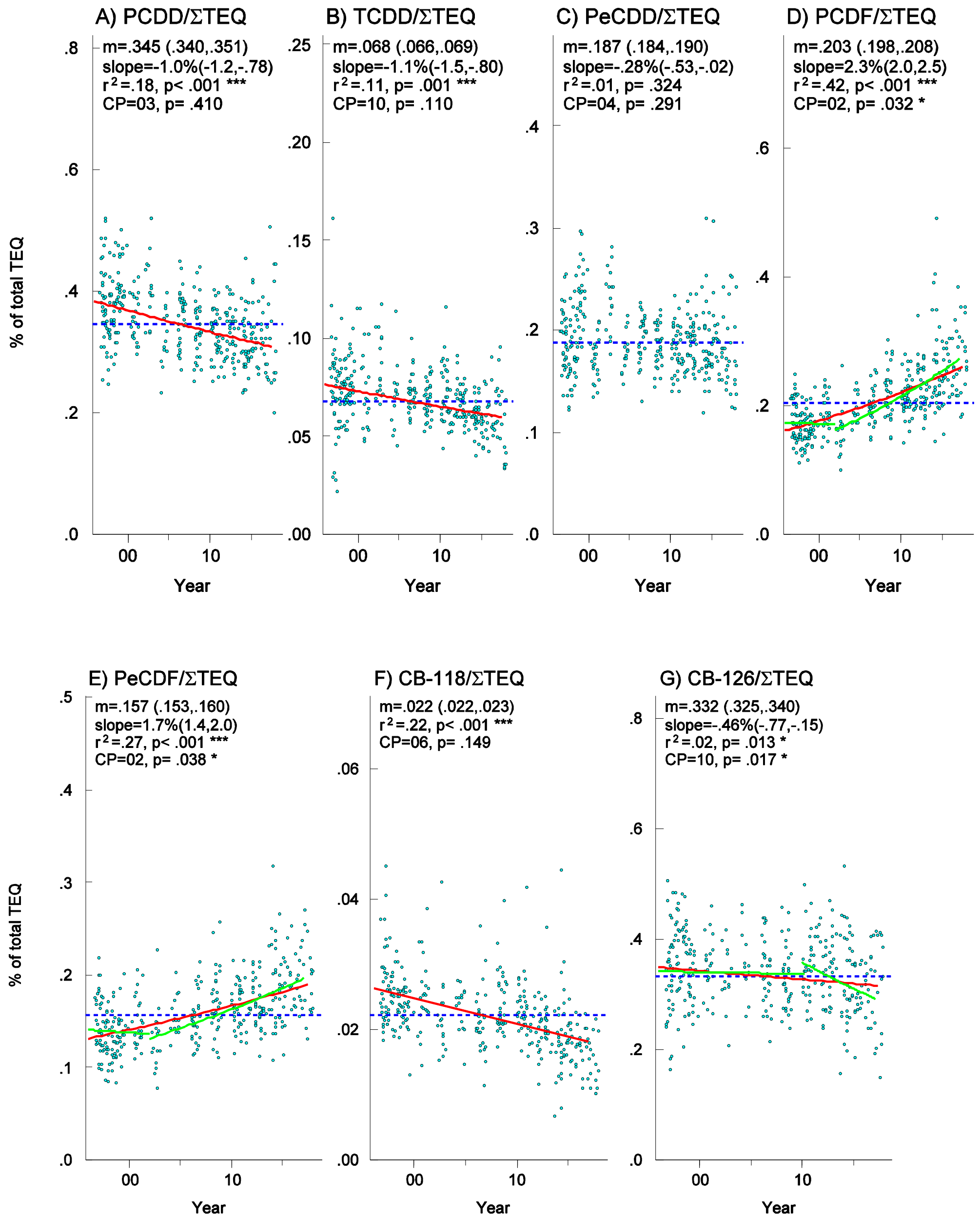


Figure 1.3. Human milk 1996-2017 from Uppsala (n=367, 19 years with data). Ratios between PCDDs, PCDFs and dl-PCBs (TEQ) and the sum of TEQ. Note that these ratios deviates somewhat from human milk from Stockholm and guillemot eggs from the Baltic (Fig.1.3).

A) The contribution from PCDD TEQ to the sum of TEQ is on average about 35% but decreases significantly from about 40% in the beginning of the monitoring period to 31% at the end.

B) Ratio between 2,3,7,8-TCDD TEQ and the sum of TEQ. The contribution from TCDD is about 7% during the monitoring period, significantly decreasing from 8 to 6%.

C) Ratio between 1,2,3,7,8-PeCDD TEQ and the sum of TEQ. The contribution from PeCDD is about 19% of the sum of TEQ during the monitoring period.

D) The contribution from sum of PCDFs TEQ is on average about 20% but increases significantly on average 2.3% a year from about 17% in the beginning of the monitoring period to 26% at the end.

E) Ratio between 2,3,4,7,8-PeCDF (TEQ) (the major part of PCDF) and the sum of TEQ. The contribution from PeCDF is on average about 16%, but increases significantly on average 1.7% a year from about 13% in the beginning to 19% at the end of the monitoring period. This increase is slower than for the sum of PCDFs implying that the contribution of other PCDFs to the sum is also increasing.

F) Ratio between CB-118 TEQ with the sum of TEQ. The contribution from CB-118 is about 2.2%, 2.5% in the beginning of the monitoring period and decreases to about 1.7%.

G) Ratio between CB-126 TEQ with the sum of TEQ. The contribution from CB-126 is about 33%. A slow but significant downward trend of 0.5% a year was detected and a change-point 2010 indicating a faster decrease since 2010.

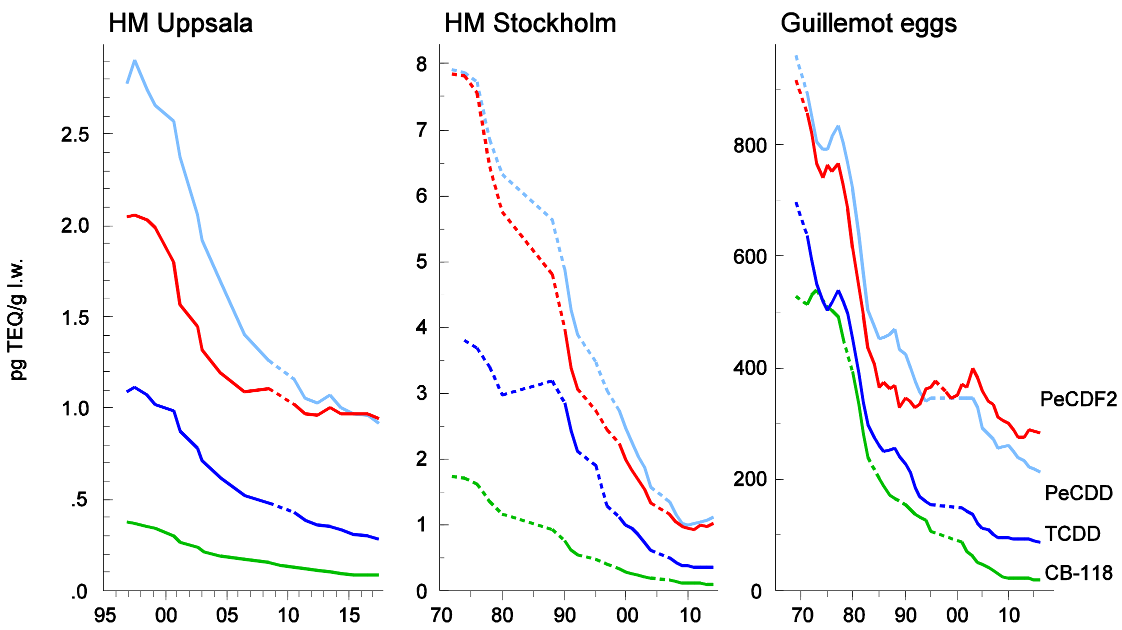


Figure 1.4. Contribution of TCDD (blue), PeCDD (light blue), 2,3,4,7,8-PeCDF(red) and CB-118 (green) in pg TEQ/g in human milk from Uppsala, Stockholm and in guillemot eggs from the Baltic Proper. Note that the Uppsala time-series is shorter. The three time-series showing rather similar patterns of PeCDD, PeCDF, TCDD and CB-118. The longer time-series of human milk from Stockholm and guillemot are somewhat more similar but in 1970-1990, CB-118 contributed more to the total TEQ in guillemot than in the monitored human milk samples. Note also the slower decrease of 2,3,4,7,8-PeCDF in human milk from Uppsala and guillemot eggs compared to human milk from Stockholm.

2 The sum of PCDD/F, mo-PCBs and no-PCBs

For summary of results see Table 3. The sum PCDD/Fs is only reported for human milk from Uppsala, from the "food basket" project and in guillemot eggs.

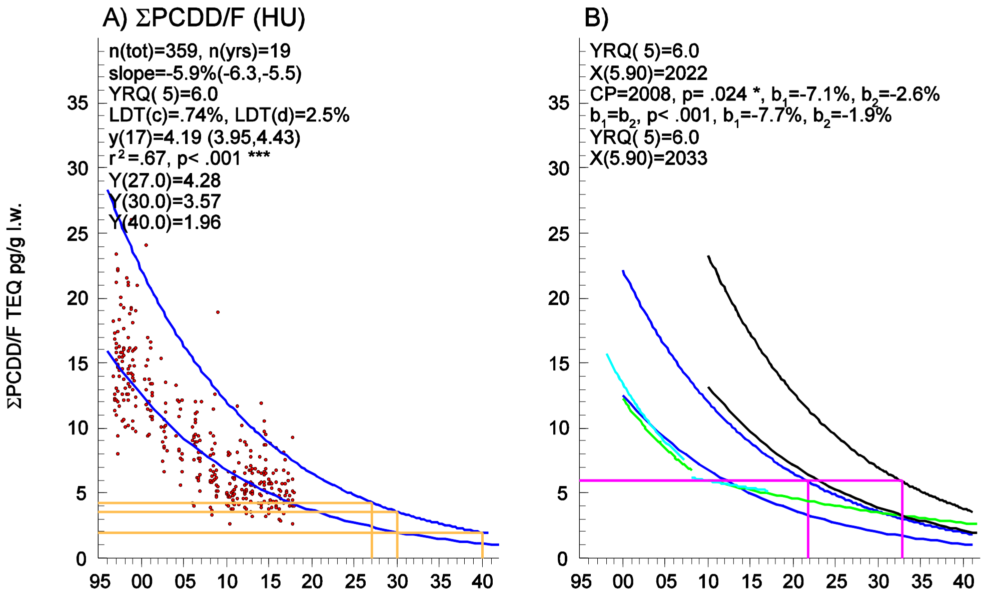


Figure 2.1. Human milk 1996-2017 from Uppsala (n=359). Average concentrations of ΣTEQ (no-CBs+mo-CBs + PCDFs + PCDDs) (adjusted for mother's age, BMI, weight gain during pregnancy and education) and upper 95% prediction(population) interval extrapolated to year 2040. Concentrations for 2027 (Y(27)), 2030 (Y(30)) and 2040 (Y(40)) were estimated through interpolation from this extrapolated upper prediction interval (See Table 3 for a summary).

A) A highly significant decreasing trend of almost 6% a year was detected. The sample sizes (i.e. the number of observations each year) and sampling frequency could potentially detect a considerably smaller trend i.e. 0.74% per year, with an 80% power for the current period of 19 years and 2.5% per year during a period of 10 years. A minimum number of 6 years would be required to detect a yearly change of 5%. Concentrations were estimated from the upper population interval at year 2027 (4.3), 2030 (3.6) and 2040 (2.0 pg TEQ/g lipid) respectively.

B) Estimated year when approximately 97.5% of the population have reached concentrations below 5.9 pg TEQ/g lipid is 2022.

Black lines simulate a "worst case scenario" from an imaginary population being exposed to twice as high dioxin TEQ concentrations as the mothers in Uppsala. In such a case 97.5% of the population is estimated to have concentrations below 5.9 pg TEQ/g lipid in 2033.

There is a significant difference between the slopes calculated for the period 1998 - 2006 (-7.7%) and the period 2007 - 2016 (-1.9%) (light blue lines).

Furthermore, a significant change-point for the whole time series was detected with a faster decrease, -7.2% until 2008 followed by a slower decrease -2.6% (green lines). Hence, the estimated concentrations at year 2027, 2030 and 2040 and the predicted years to reach the target concentration of 5.9 pg TEQ/g lipid above may be underestimated.

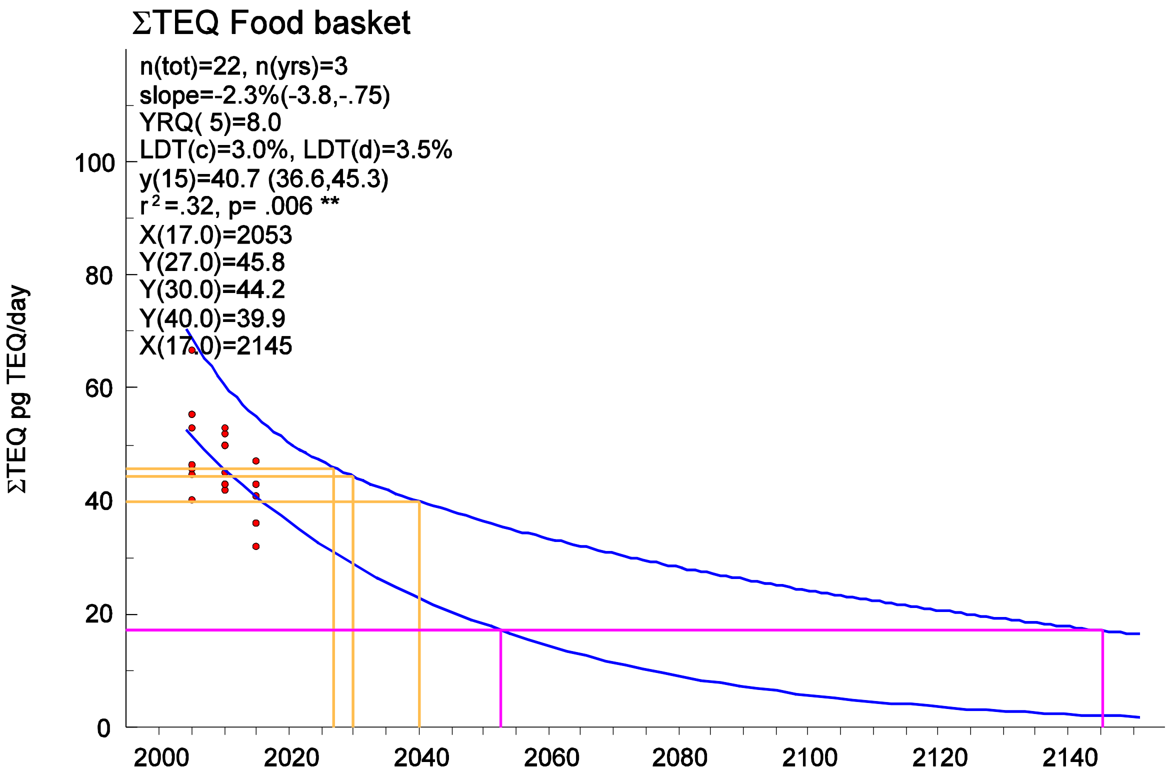


Figure 2.2. Data from the "Food basket"-project (SFA) (n=22, year 2005, 2010, 2015). Temporal trend of estimated per capita intake of ΣTEQ (no-CBs + mo-CBs + PCDDs + PCDFs) (pg TEQ per day) and upper 95% prediction (population) interval extrapolated to year 2040. A significant decreasing trend of 2.3% per year was detected. Per capita intake was estimated from the upper population interval at year 2027 (46), 2030 (44) and 2040 (40 pg/g TEQ/day) respectively. For a woman with a body weight of 60 kg the daily intake at TWI is 17 pg per day. For the average concentrations this level is reached in year 2053, for the upper population interval, not until 2145, provided the trend continue over the years.

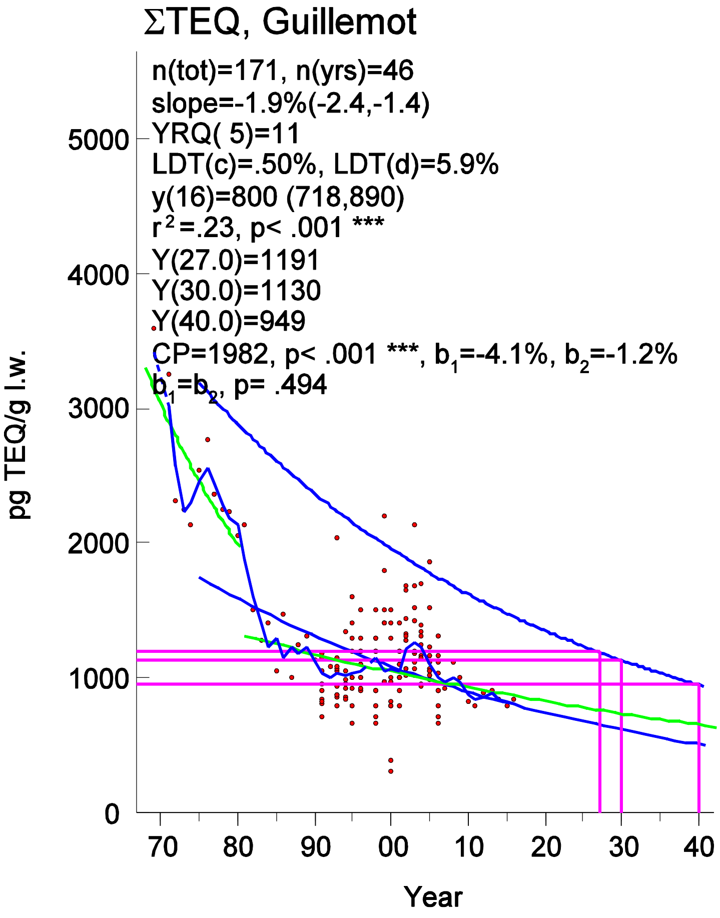


Figure 2.2. Average concentrations of ΣTEQ (n-o-CBs+m-o-CBs + PCDFs + PCDDs) in guillemot eggs from the Baltic Proper. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively (see statistics in figure above (Y(27)=, Y(30)=, Y(40)=). This long time-series starting already 1969, show a significant decreasing trend of on average 1.9% a year, with a significant change-point 1982. From 1969 to 1982 the decrease was on average 4.1% and after 1982 the decrease is only 1.2% a year. No significant difference was detected between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

Geographical distribution

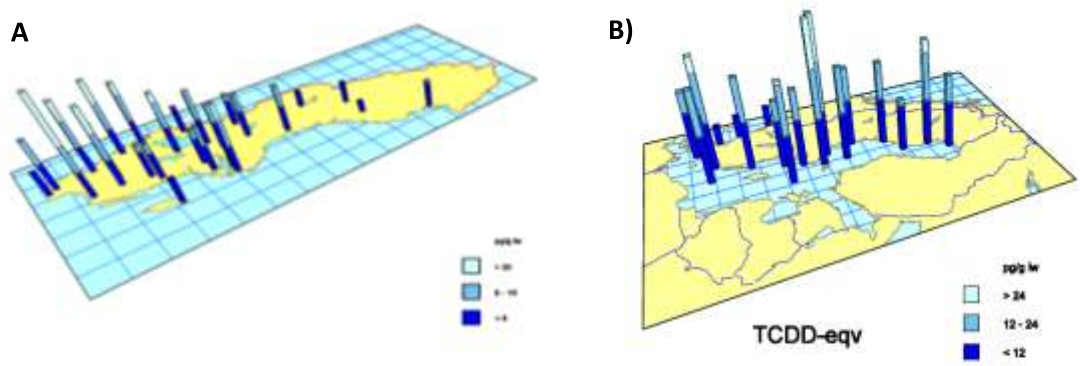


Figure 2.3. ΣTEQ in Sweden around 2017 **A)** in perch muscle. Data from the National Swedish Contaminant Monitoring Programme for Freshwater Biota (Nyberg *et al.* 2018). For freshwater fish (perch), there is an apparent geographical trend with concentrations generally decreasing from south to north. **B)** In herring from the Baltic and the Swedish west coast. The freshwater perch trend is not as evidently seen in Baltic herring (Bignert *et al*., 2018) indicating that local Swedish sources are added to the atmospheric deposition from the industrialized western Europe reported in Wiberg *et al.* (2013).

Note that saltwater fish, like herring, in the northern Baltic Sea with low salinity grow slower than in the southern more saline Baltic Sea, and hence accumulate dioxins during a longer time, leading to higher concentrations in northern Baltic Sea herring when comparing herring of the same size from the southern Baltic Sea.

3 PCDD, chlorinated dibenzo-p-dioxins

ΣPCDD, the sum of chlorinated dibenzo-p-dioxins is only reported for milk from Uppsala

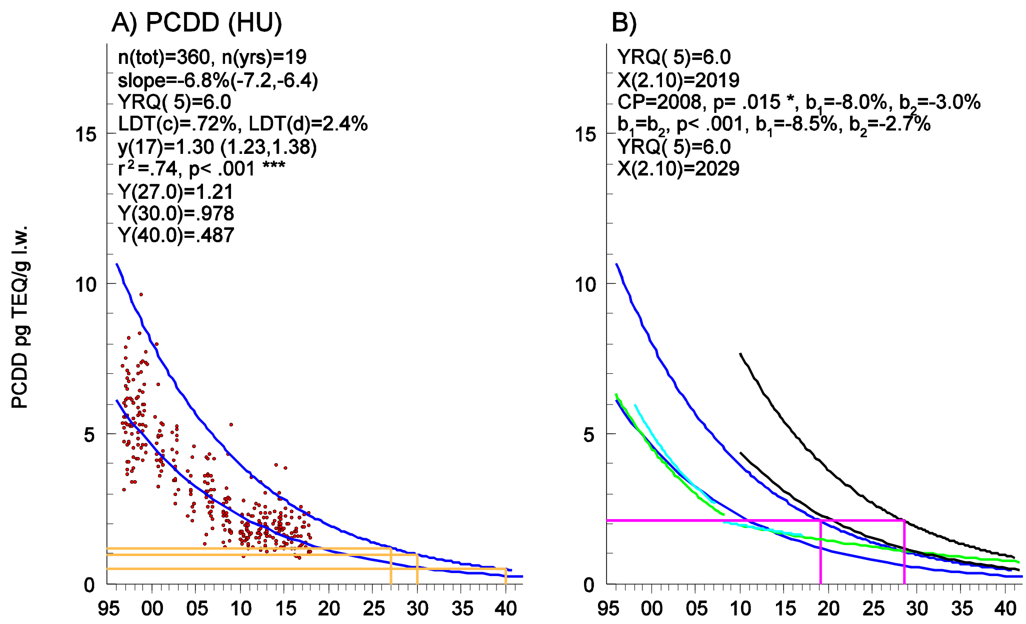


Figure 3.2. Human milk 1996-2017 from Uppsala (n=360). Average concentrations of the sum of PCDDs (adjusted for mother's age, BMI, weight gain during pregnancy and education) and upper 95% prediction (population) interval extrapolated to year 2040.

A) A highly significant decreasing trend of almost 7% a year was detected. The sample sizes and sampling frequency could potentially detect a considerably smaller trend i.e. 0.72% per year, with an 80% power for the current period of 19 years and 2.4% per year during a period of 10 years. A minimum number of 6 years would be required to detect a yearly change of 5%. Concentrations were estimated from the upper population interval at year 2027 (1.2), 2030 (0.98) and 2040 (0.49 pg TEQ/g lipid) respectively.

B) Estimated year when approximately 97.5% of the population are exposed to concentrations below 0.35 x 5.9 (=2.1) pg TEQ/g, is during 2019. Since the contribution from PCDDs to the sum of TEQ is approximately 35% (Fig.1.2 and 1.3), a target value of 2.1 pg TEQ/g lipidwas chosen. Below the target, exposure from dioxins, furans and dioxin-like PCBs would likely be safe provided that the relative contributions from the three groups remains similar.

Black lines simulate a "worst case scenario" from an imaginary population being exposed to twice as high PCDD TEQ concentrations as the mothers in Uppsala. In such a case 95% of the population is estimated to go below the target value in 2029.

There is a significant difference between the slopes calculated for the period 1998 - 2006 (-8.5%) and the period 2007 - 2016 (-2.7%) (light blue lines).

Furthermore, a significant change-point was detected with a faster decrease, -8.0% until 2008 followed by a slower decrease -3.0% (green lines). Hence, the predicted years above to reach the target values may be underestimated.

TCDD (2,3,7,8-Tetrachlorodibenzo-p-dioxin)

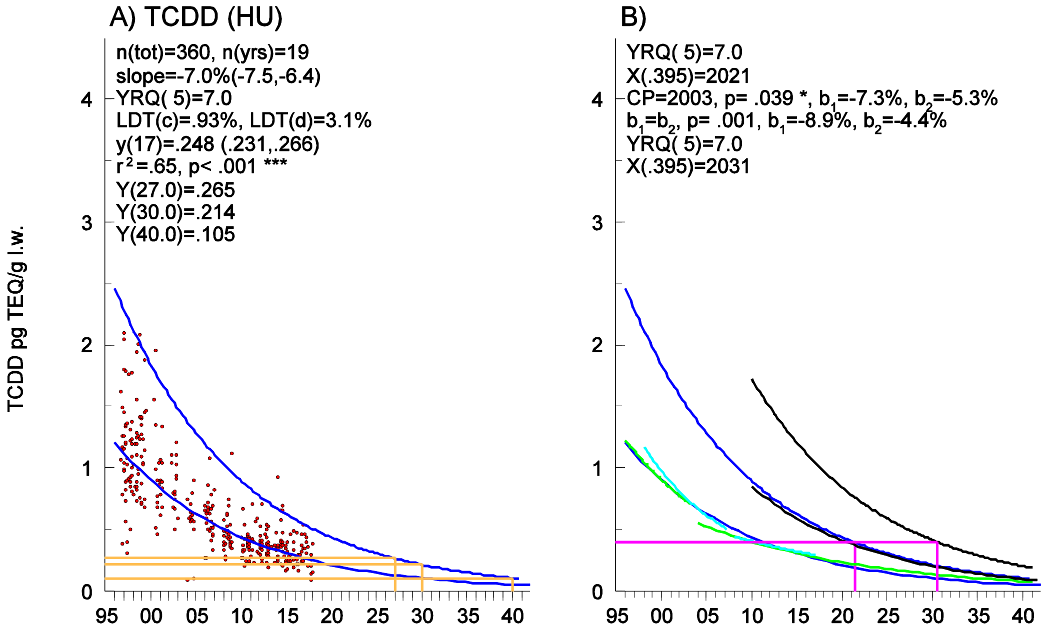


Figure 3.4. Human milk 1995-2017 from Uppsala (n=396). Average concentrations of TCDD (adjusted for mother's age, BMI, weight gain during pregnancy and education) and upper 95% prediction (population) interval extrapolated to year 2040.

A) Concentrations were estimated from the upper population interval at year 2027 (0.26), 2030 (0.21) and 2040 (0.10 pg TEQ/g lipid) respectively (see statistics in figure above (Y(27)=, Y(30)=, Y(40)=).

B) Estimated year when approximately 97.5% of the population are exposed to concentrations below 0.07 x 5.9 (=0.41) pg TEQ/g, is during 2021. Since the contribution from TCDD to the sum of TEQ is approximately 7% (Fig 1.2 and 1.3), a target value of 0.41 pg TEQ/g lipid was chosen. Below the target, exposure from dioxins, furans and dioxin-like PCBs would likely be safe provided that the relative contributions from the three groups remains similar.

Black lines simulate a "worst case scenario" from an imaginary population being exposed to twice as high PCDD TEQ concentrations as the mothers in Uppsala. In such a case 95% of the population is estimated to reach the target value in 2031.

There is a significant difference between the slopes calculated for the period 1998 - 2006 (-8.9%) and the period 2007 - 2016 (-4.4%) (light blue lines).

A significant change-point was detected but the estimated trend after the change-point is not much different from the estimated trend for the whole period (-7.3% and -5.3%, respectively) (green lines).

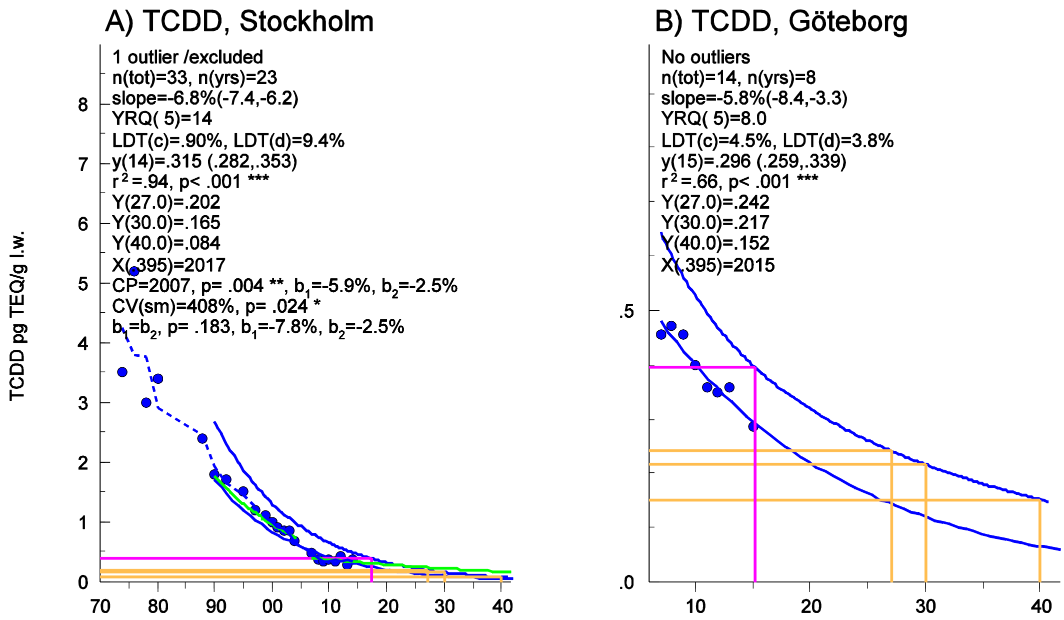


Figure 3.5. A) Human milk 1972-2014 from Stockholm (n=35) and **B)** from Göteborg 2007-2015 (n=14), pooled samples. Average concentrations of TCDD and upper 95% prediction (population) interval extrapolated to year 2040.

A) Significant decreasing trend on average 6.2% per year. The longer time-series from Stockholm show that the TCDD levels today are less than 10% of the levels during the 80-ties. Concentrations were estimated from the upper population interval at year 2027 (0.20), 2030 (0.16) and 2040 (0.084 pg TEQ/g lipid) respectively. These concentrations are somewhat lower than comparable estimates from the Uppsala population though the slopes from the two populations are similar.

There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

A significant change-point was detected at year 2007 (-5.9% and -2.5%, before and after the change-point (green lines).

B) Significant decreasing trend on average 5.8% per year. Concentrations were estimated from the upper population interval at year 2027 (0.24), 2030 (0.22) and 2040 (0.15 pg TEQ/g lipid) respectively i.e. in the middle of and quite similar to the Uppsala and Stockholm samples.

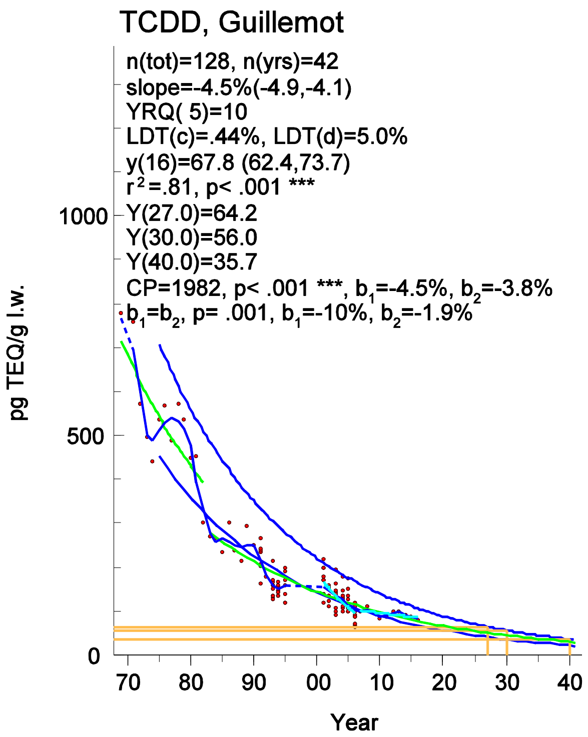


Figure 3.6. TCDD in guillemot eggs from the Baltic Proper. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively (see statistics in figure above (Y(27)=, Y(30)=, Y(40)=). This long time-series starting already 1969, show a significant decreasing trend of on average 4.5% per year.

A significant change-point is detected with a somewhat steeper slope up to 1985, -4.5% compared to -3.8%, but the second part is not deviating strongly from the overall trend (green lines).

Comparing the periods 1998 - 2006 (-10%) and 2007 - 2016 (-1.9%), show a significant difference in the slopes (light blue lines), but the difference seems to be driven by the somewhat exaggerated downward slope during 1998-2006, compare concentrations during the beginning of the 90-ies.

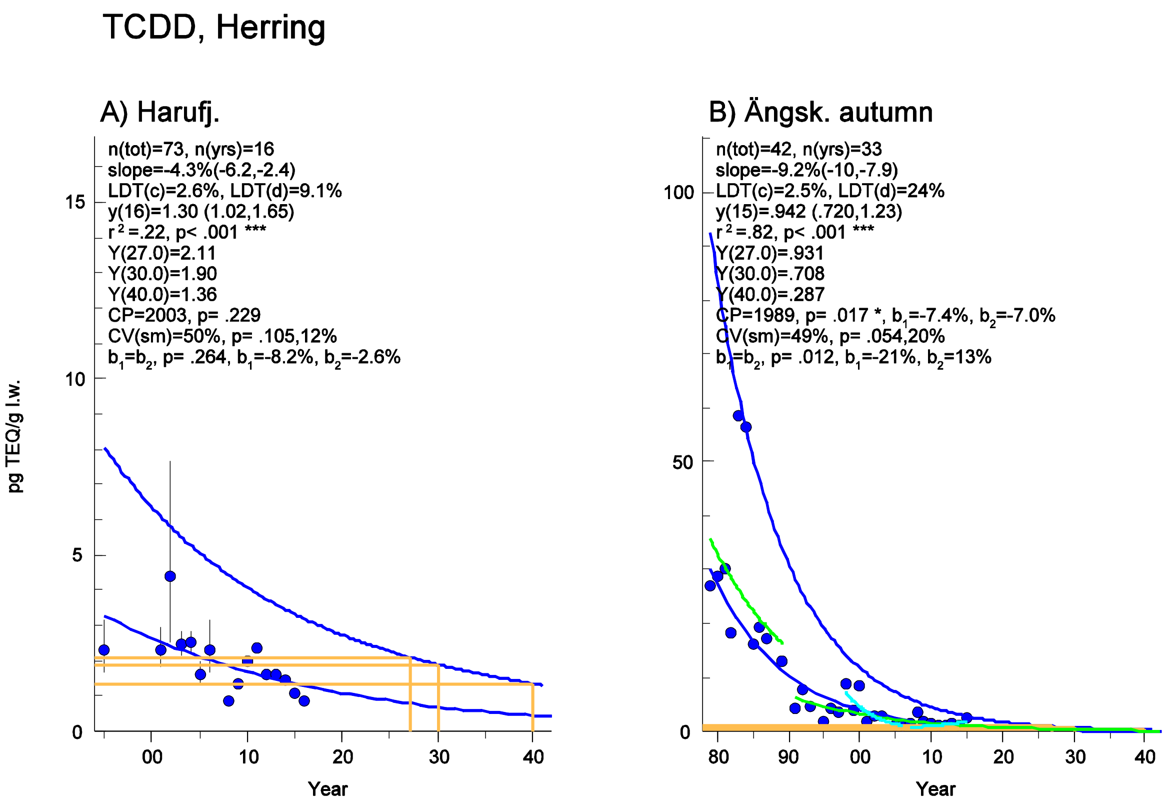


Figure 3.7. TCDD in Baltic herring. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Bothnian Bay (Harufjärden), decreasing trend on average 4.3% per year. Extrapolated concentrations year 2027, 2030, 2040 to 2.1, 1.9 and 1.4 respectively (see statistics in figure above). There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

B) South Bothnian Sea (Ängskärsklubb, autumn), long time-series (33 year), significant decrease, on average more than 9% a year (indicating that high dioxin sediment (Sundqvist *et* al., 2009) concentrations in this area are not leaking extensively to the pelagic herring). Extrapolated concentrations year 2027, 2030, 2040 to 0.93, 0.71 and 0.29 respectively (see statistics in figure above). There is a significant difference between the slopes calculated for the period 1998 - 2006 (-21%) and the period 2007 - 2016 (+13%), but the increasing slope for the last period is not significant. There is also a significant change-point -7.5% before 1989 and -7.0% after.

Two shorter time-series, from the south Bothnian Sea (Ängskärsklubb spring, 12 years), and the north Baltic Proper (Landsort, 11 years), showed no significant change. Figures not shown.

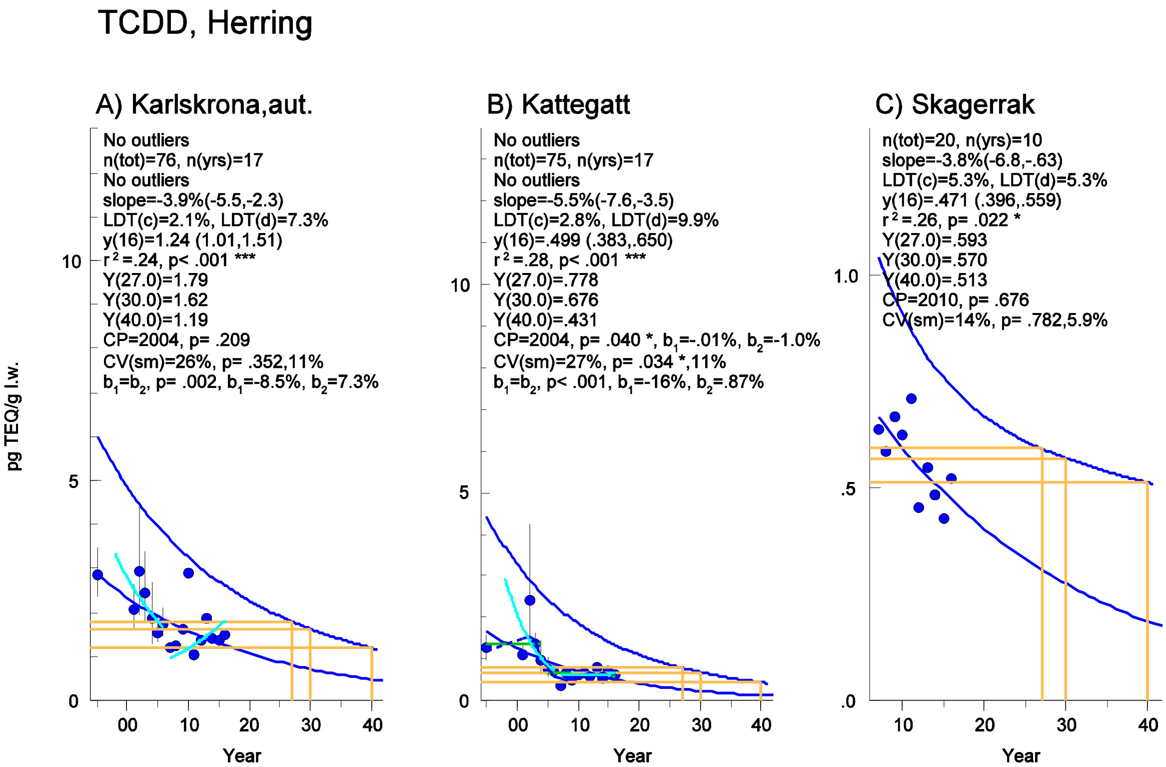


Figure 3.8.1. TCDD in herring from the Baltic Proper and the Swedish west coast. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively (see statistics in figure above (Y(27)=, Y(30)=, Y(40)=). The measured average herring concentrations 2016, from the s. Baltic Proper (Karlskrona) are more than twice as high as the concentrations in herring from the Swedish west coast (Kattegatt and Skagerrak).

A) Autumn caught herring from the s. Baltic Proper (Karlskrona, autumn), decrease significantly, on average 3.9% per year. There is a significant difference between the slopes calculated for the period 1998 - 2006 (-8.5%) and the period 2007 - 2016 (+7.3%), but the indicated increasing slope for the last period is not significant.

B) Kattegat, significant decreasing trend, on average 5.5% per year. A significant difference between the slopes calculated for the period 1998 - 2006 (-16%) and the period 2007 - 2016 (+0.87%) was found.

C) Skagerrak. Low concentrations. Significant decreasing trend after adjustment for age, weight and fat content (fat content variable in the Skagerrak herring and before adjustment an increasing trend was indicated, see Fig 3.8.2, below).

A short (10 years) time-series from southern Baltic Proper, spring caught herring, showed no significant trend. Figure not shown.

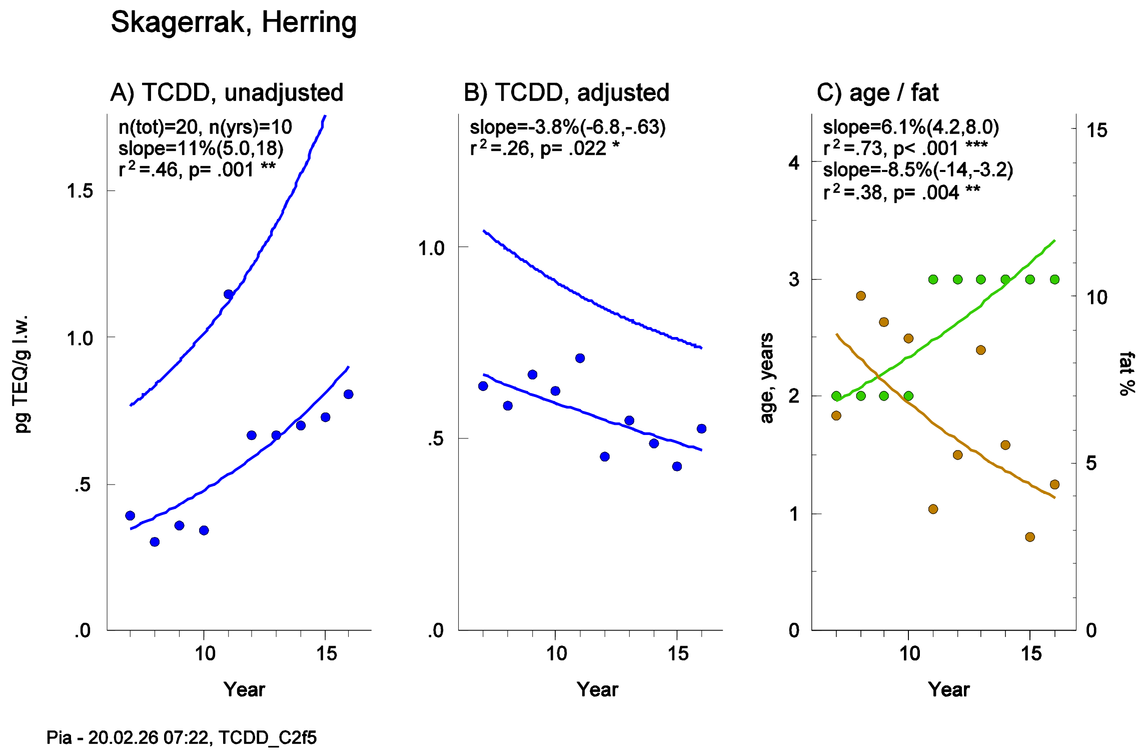


Figure 3.8.2. A) Unadjusted TCDD-concentrations in herring from the Skagerrak.

B) TCDD-concentrations adjusted to concentrations expected if age and fat content were constant.

C) Temporal trends of age (green) and fat content (brown). It is known that dioxins accumulate, i.e. increase with increasing age and that dioxins increase with decreasing fat content (on a lipid weight basis). In the time-series of herring from the Skagerrak the age is increasing over time and the fat content is decreasing, thus the dioxin concentrations can be expected to increase just because of these facts, irrespective of anthropogenic discharges of dioxin. Compensating for this, assuming constant age and fat content result in figure **B**.

The fact that age increases and fat content decreases over time at the Skagerrak sampling station, implies that all the dioxins should be adjusted for age and fat at this site.

(In general, the sampling protocols for fish within the national program for monitoring of environmental contaminants, narrows down the ranges of sizes, age, samling time and so forth making adjustments less important. Adjustments may introduce extra uncertainty, especially with narrow ranges in confounder variables, also some data loss if confounder variables are not measured consistently over time in longer time-series. Therefore adjustments are carried out only after considering these circumstances)

There are two additional time-series of TCDD in perch from the Baltic. However, no significant trends were found but the time-series are too short (10 and 11 years) and variable to make any meaningful trend assessments or predictions. No figures are shown.

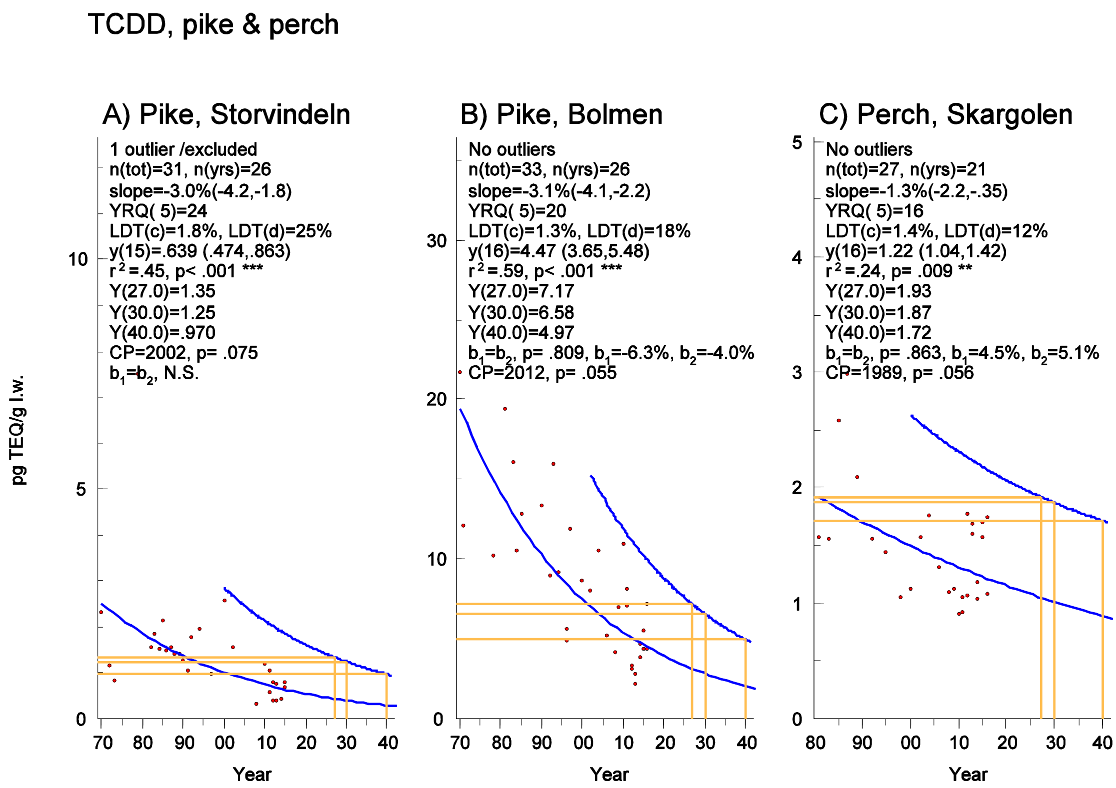


Figure 3.9. TCDD in freshwater fish (adjusted for age, weight and lipid content). Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Pike from n. Sweden (L. Storvindeln), slow (-1.9% a year) but significant decrease.

B) Pike from s. Sweden (L. Bolmen), significant decrease (-3% per year). The calculated average concentration from year 2016 were six times higher in this pike compared to the pike from northern Sweden (L. Storvindeln above) (c.f. Fig. 2.3, geographical pattern in perch).

C) Perch from s. Sweden (L. Skärgölen), show a slow (-1.3% per year) but significant decrease.

The TEQ values, expressed on a lipid weight basis, are quite high, both pike and perch are lean fish (lipid content < 1%) and therefore the fresh weight concentrations are substantially lower compared to fat fish.

PeCDD (1,2,3,7,8-Pentachlorodibenzo-p-dioxin)

This dioxin has a TEF-value of 1.0, the same as TCDD, and the contribution from PeCDD is about 19% of the sum of TEQ (Fig 1.3).

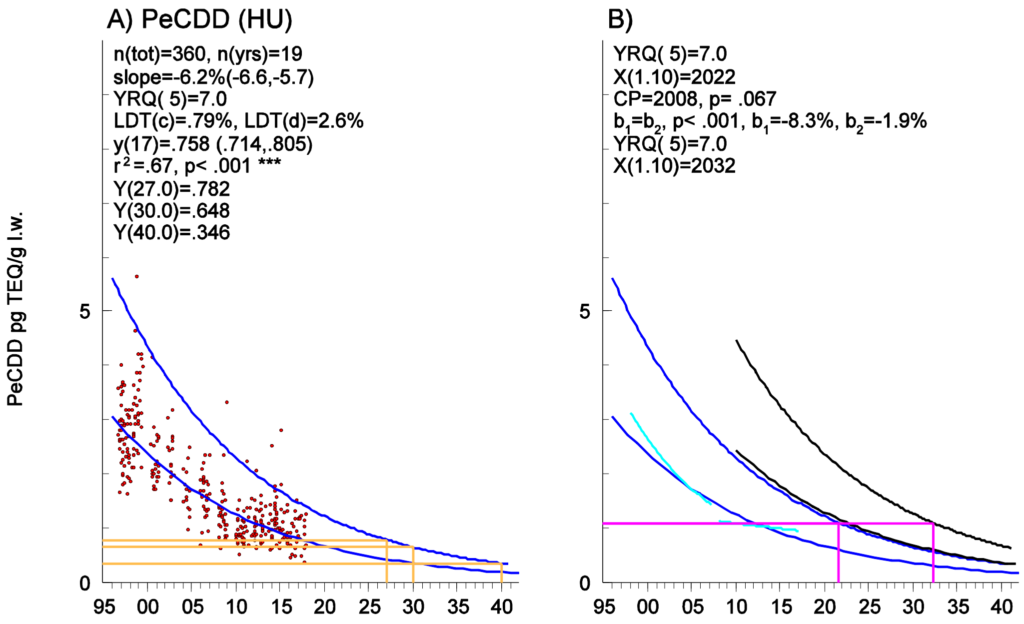


Figure 3.10. Human milk 1996-2017 from Uppsala (n=360). Average concentrations of PeCDD (adjusted for mother's age, BMI, weight gain during pregnancy and education) and upper 95% prediction (population) interval extrapolated to year 2040 (see statistics in figure above (Y(27)=, Y(30)=, Y(40)=).

A) A highly significant decreasing trend of 6.2% per year was detected. The sample sizes and sampling frequency could potentially detect a considerably smaller trend i.e. 0.79% per year, with an 80% power for the current period of 19 years and 2.6% per year during a period of 10 years. A minimum number of 7 years would be required to detect a yearly change of 5%. Concentrations were estimated from the upper population interval at year 2027 (0.78), 2030 (0.65) and 2040 (0.35 pg TEQ/g lipid) respectively.

B) Estimated year when approximately 97.5% of the population were exposed to concentrations below the target value of 0.19 x 5.9 (=1.1) pg TEQ/g lipid i.e. the year 2022.

Black lines simulate a "worst case scenario" from an imaginary population being exposed to twice as high PCDD TEQ concentrations as the mothers in Uppsala. In such a case 97.5% of the population is estimated to reach the target value in 2032.

There is a significant difference between the slopes calculated for the period 1998 - 2006 (-8.3%) and the period 2007 - 2016 (-1.9%), respectively but no significant change-point was shown.

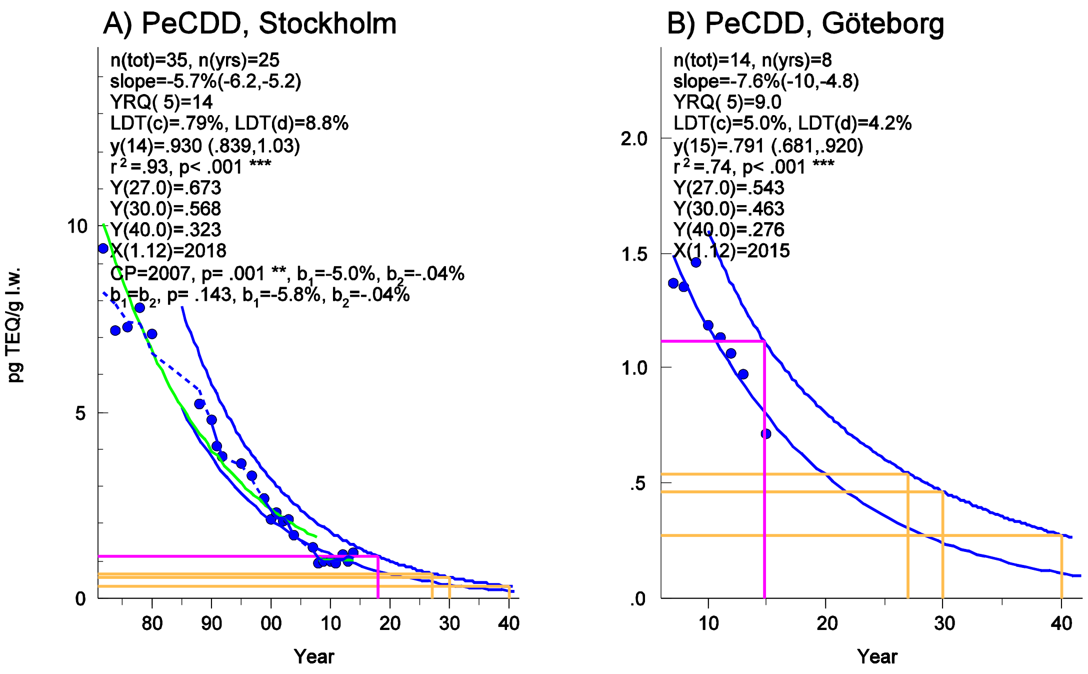


Figure 3.11. Human milk, average concentrations of PeCDD and upper 95% prediction (population) interval extrapolated to year 2040.

A) Human milk 1972-2014 from Stockholm (n=35). Significant decreasing trend on average 5.7% per year. Concentrations were estimated from the upper population interval at year 2027 (0.67), 2030 (0.57) and 2040 (0.32 pg TEQ/g lipid) respectively.

There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016, but a change-point was identified with a slope of -5.0% before 2007 and -0.04% after 2007.

B) Human milk from Göteborg, 2007-2015 (n=14). Significant decreasing trend on average 7.6% per year. Concentrations were estimated from the upper population interval at year 2027 (0.54), 2030 (0.46) and 2040 (0.28 pg TEQ/g lipid) respectively. The regression analysis indicates a somewhat faster decrease in this population, affecting of course also the estimated years from the extrapolation. However, only 8 years of data were available and considering this uncertainty, the milk from Uppsala, Stockholm and Göteborg are reasonably similar.

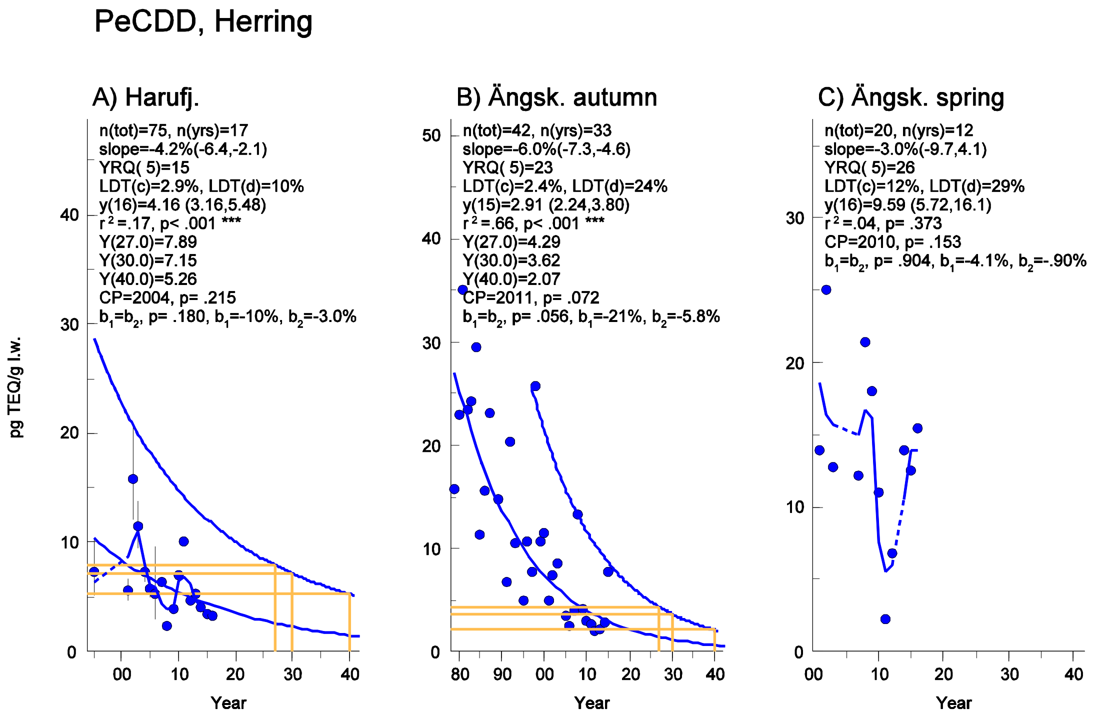


Figure 3.12. PeCDD in Baltic herring. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively (see statistics in figure above (Y(27)=, Y(30)=, Y(40)=).

A) Bothnian Bay (Harufjärden), significant decreasing trend, on average 4.2% per year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016. Concentrations were estimated from the upper population interval at year 2027 (7.9), 2030 (7.2) and 2040 (5.3 pgTEQ/g fat) respectively.

B) Southern Bothnian Sea (Ängskärsklubb autumn), long time-series (33 year), significant decrease, on average 6% a year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016. Concentrations were estimated from the upper population interval at year 2027 (4.3), 2030 (3.6) and 2040 (2.1 pg TEQ/g fat) respectively i.e. lower compared to the site above. The estimated slope is rather fast for this long time-series at Ängskärsklubb, off the coast of Gävle, at least former an area with concentrated pulp industry. The fast slope, should it continue, would bring down the concentrations faster than the concentrations in herring from the Bothnian Bay. Also the slower growth at the northern, less saline site will tend to increase the concentrations at that site.

C) Southern Bothnian Sea (Ängskärsklubb spring), short time-series (12 years), a significant smoother indicate a dip in the temporal trend followed by a steep increase. The extreme dip may be caused by concentrations suddenly falling right beneath the LOD.

Northern Baltic Proper (Landsort), short time-series (11 years) no significant change. Figure not shown.

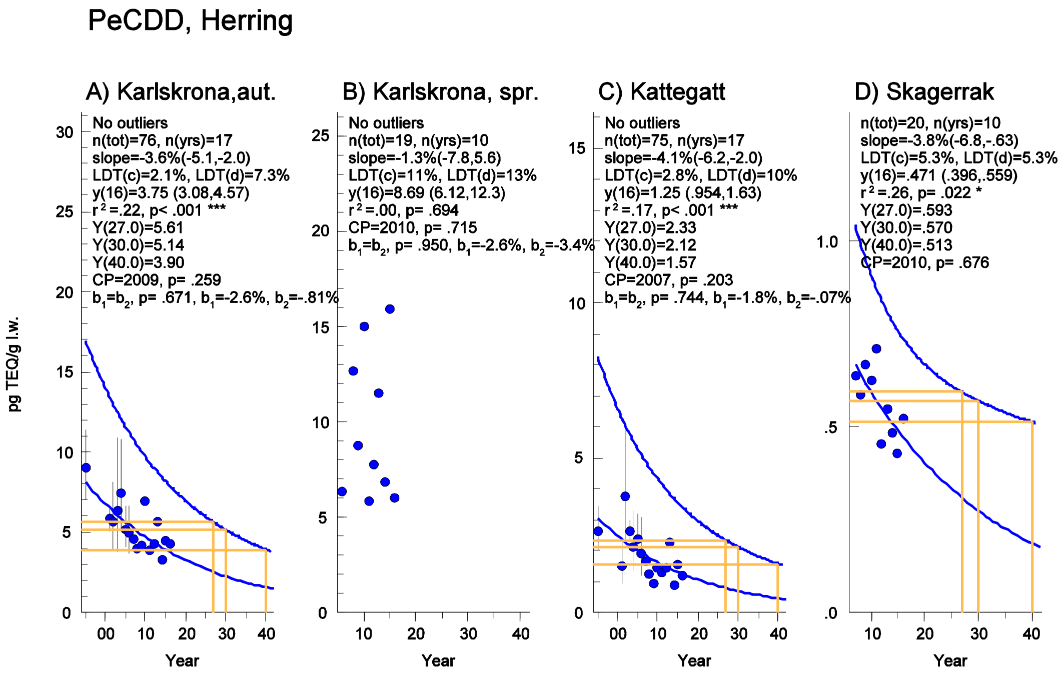


Figure 3.13. PeCDD in herring from the Baltic Proper and the Swedish west coast. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively (see statistics in figure above (Y(27)=, Y(30)=, Y(40)=).

A) Autumn caught herring from the s. Baltic Proper (Karlskrona), decreasing on average 3.6% per year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016. The estimated herring concentrations 2016, from the s. Baltic Proper are more than twice as high as the concentrations from the Swedish west coast.

B) Spring caught herring from the southern Baltic Proper (Karlskrona), short time-series (10 years), no significant trend was shown. Estimated concentrations 2016 (Y(16)=, see statistics in figure above) are more than twice as high in spring caught herring compared to autumn caught herring from approximately the same site. This seasonal variation may have several explanations see e.g. Assefa *et al.*, (2019).

C) Kattegat, significant decreasing trend, on average 4.1% per year. No significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

D) Skagerrak. Low concentrations. Significant decreasing trend, on average 3.8% per year after adjustment for age, weight and fat content (fat content variable in the Skagerrak herring and before adjustment an increasing trend was indicated, see Fig 3.7.2, above).

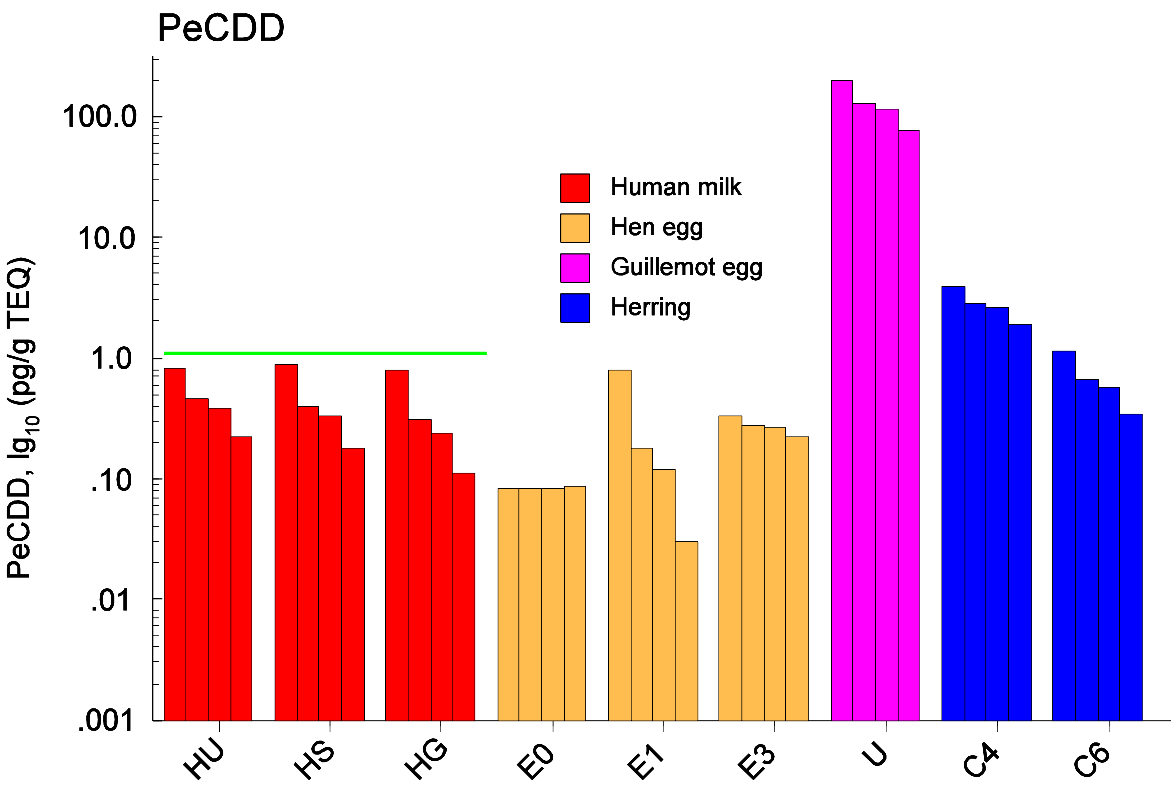


Figure 3.14. Concentrations of 1,2,3,7,8-PeCDD (TEQ lw) in various biological matrices at year 2017, 2027, 2030 and 2040. H=Human milk(red), U=Uppsala, S=Stockholm, G=Göteborg, E=hen eggs (light brown), 0= caged, 1= sputtering, 3="eco", U=guillemot (purple), C4=herring (blue) from s. Baltic Proper, C6=herring from Kattegat at the Swedish west coast. The green line at 0.19 x 5.9 (1.1) pg TEQ/g lipid, indicate the target concentration of PeCDF, considered as "safe" at present in human milk, assuming relative contributions from different dioxin-like substances remain unchanged.

Note the lower levels and faster decreasing concentrations in the herring from the Swedish west coast (C6).

PeCDD in pike and perch

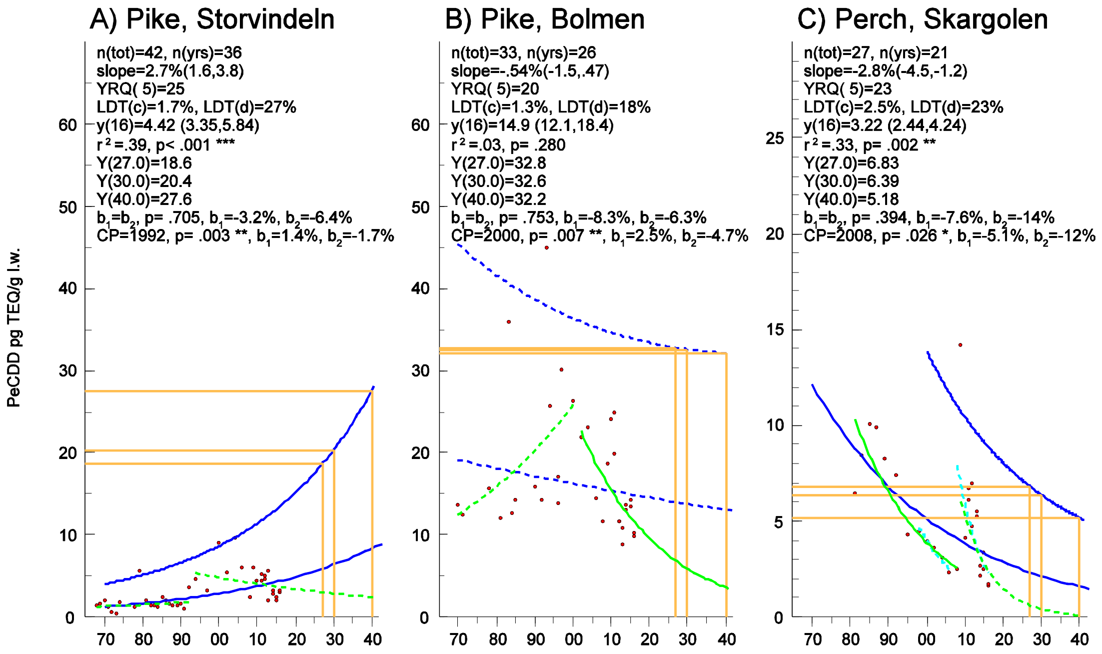


Figure 3.15. PeCDD (adjusted for weight and fat) in freshwater fish. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Pike from n. Sweden (L. Storvindeln) shows a significant ***increase***, but the levels 2027, 2030 and 2040 are still lower than corresponding values for pike from L. Bolmen. Moreover, a change-point was detected year 1992, after which the concentrations seem to decline somewhat albeit not significantly. Up to 1992 the concentrations were generally below LOQ.

B) Pike from s. Sweden (L. Bolmen), considerably higher concentrations compared to pike from the north of Sweden. A significant change-point was detected in year 2000.

C) Perch from s. Sweden (L. Skärgölen), significant downward trend, almost 3% a year. A significant change-point was detected indicating also a jump in the data around year 2008 for which we have no explanation.

The TEQ values, expressed on a lipid weight basis, are quite high, both pike and perch are lean fish (lipid content < 1%) and therefore the fresh weight concentrations are substantially lower.

Summary tables for PCDDs

Dioxin	Matrix	N(tot)	N(years)	Trend(%)	Signif.	Y(X=2027)	Y(X=2030)	Y(X=2040)	
ΣTEQ	HU	359	19	-5.91	---	4.28	3.58	1.96	
	FB	22	3	-2.29	--	45.84	44.24	39.86	1
	U8	171	46	-1.9	---	1191	1130	949	
ΣPCDD	HU	360	19	-6.79	---	1.21	0.98	0.49	
TCDD	HU	360	19	-6.95	---	0.26	0.21	0.11	
	HS	34	25	-6.22	---	0.26	0.22	0.12	
	HG	14	8	-5.84	---	0.24	0.22	0.15	
	U8	128	49	-4.53	---	64.18	56.04	35.71	
	C1	75	17	-4.43	---	2.04	1.83	1.30	
	C2	42	33	-9.19	---	0.93	0.71	0.29	
	V2	20	12	-0.76					
	C3	20	11	1.22					
	C4	76	17	-3.91	---	1.79	1.62	1.19	
	V4	19	10	-4.93					
	C6	75	17	-5.54	---	0.78	0.68	0.43	
	C7	20	10	-3.78	-	0.59	0.57	0.51	
	ES	38	32	-1.86	---	1.48	1.41	1.21	2
	EB	33	26	-3.15	---	7.17	6.58	4.97	2
	PS	27	21	-1.30	--	1.93	1.87	1.72	2
PeCDD	HU	360	19	-6.15	---	0.78	0.65	0.35	
	HS	35	25	-5.72	---	0.67	0.57	0.32	
	HG	14	8	-7.62	---	0.54	0.46	0.28	
	C1	75	17	-4.24	---	7.89	7.15	5.26	
	C2	42	33	-6.00	---	4.29	3.62	2.07	
	V2	20	12	-3.04					
	C3	20	11	-2.98					
	C4	76	17	-3.60	---	5.61	5.14	3.90	
	V4	19	10	-1.28					
	C6	75	17	-4.15	---	2.33	2.12	1.57	
	C7	20	10	-3.78	-	0.59	0.57	0.51	
	ES	42	36	2.69	+++	18.64	20.38	27.59	2
	EB	33	26	-0.54		32.82	32.62	32.16	2
	PS	27	21	-2.85	--	6.83	6.39	5.18	2

Table 3. Trends and predicted concentrations of PCDDs (pg TEQ/g lipid) at 2027, 2030 and 2040 from the upper 95% population interval, i.e. 97.5% of the population assumed to be below these concentrations. Matrix: H=Human milk, U=Uppsala (1996-2017), S=Stockholm (1972-2014), G=Göteborg (2007-2015), FB=Food basket (2005, 2010, 2015), U8=guillemot egg (1969-2016), C=herring, V=spring caught herring, C1=Bothnian Bay (1995-2016), C2=Bothnian Sea(1979-2015), V2 (2001-2016), C3=n. Baltic Proper (2005-2016), C4=s. Baltic Proper (1995-2016), V4 (2006-2016), C6=Kattegat (1995-2016), C7=Skagerrak (2007-2016), ES=pike from n. Sweden (1968-2016), EB= pike from s. Sweden (1970-2016), PS= fresh water perch from s. Sweden (1981-2016). **1** =pg TEQ/day, **2** =lean fish i.e. high concentrations on a lipid weight basis. **---** = decreasing trend p<0.001, **--** p<0.01, **-** p<0.05. Median trend for all tabled matrices = **-** 4.03 % per year

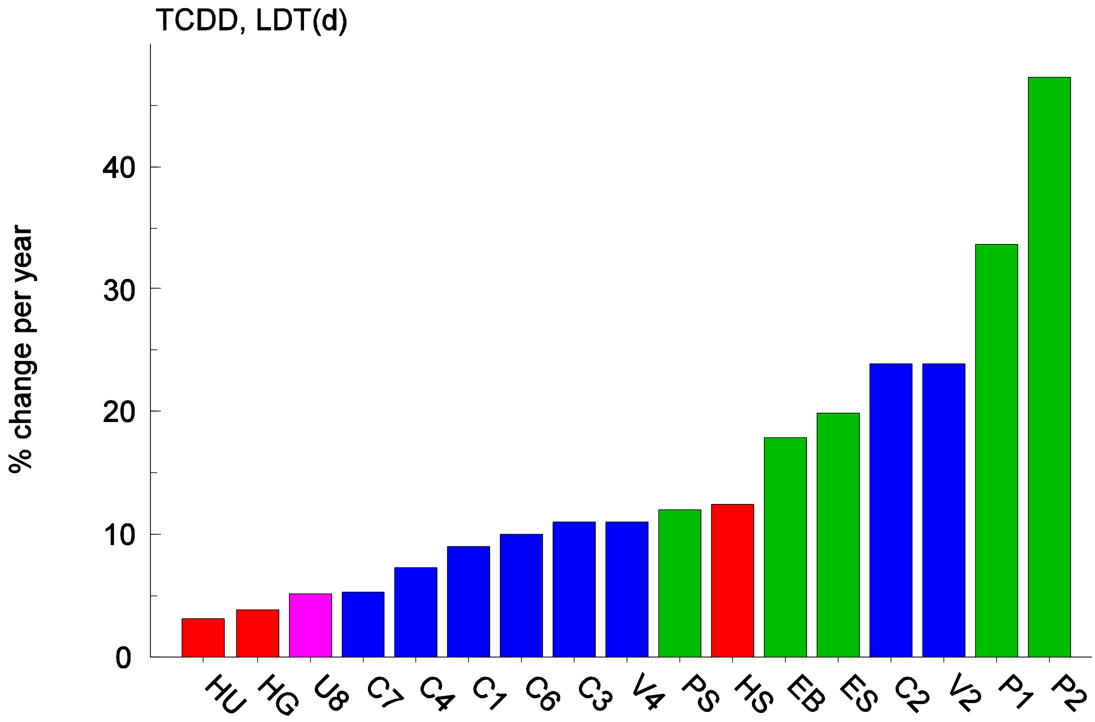


Figure 3.16. This figure shows the LDT(d), the smallest log-linear trend (% per year) a certain time-series can detect, during a ***10 year*** period with an 80 % statistical power, i.e. the lower the better. HU, HS, HG=human milk from Uppsala, Stockholm and Göteborg (red bars); U8=guillemot egg (purple bar); C1-7=autumn caught herring along the east and west coast (dark blue=fat fish); V2, V4=spring caught herring from the Baltic (dark blue); P1, P2=Baltic perch, PS=fresh water perch, ES, EB=freshwater pike (green=lean fish). Various matrices show a various degree of random/unexplained variation along the regression line. Large variation implies that it is more difficult to detect a small trend. The best matrices from this point of view are the fatty matrices of milk from Uppsala and Göteborg and guillemot eggs, the poorest are lean fish, perch from the Baltic (low concentrations and low fat content) (Note that true non-linear trends in the data may spoil the potential to detect linear or log-linear trends (e.g. human milk from Stockholm). Note that several time-series in the current report are considerably longer than 10 years and thus able to detect smaller yearly trends.

4 PCDFs, chlorinated furans

ΣPCDF, the sum of chlorinated furans is only reported for milk from Uppsala. Summary of results see Table 4.

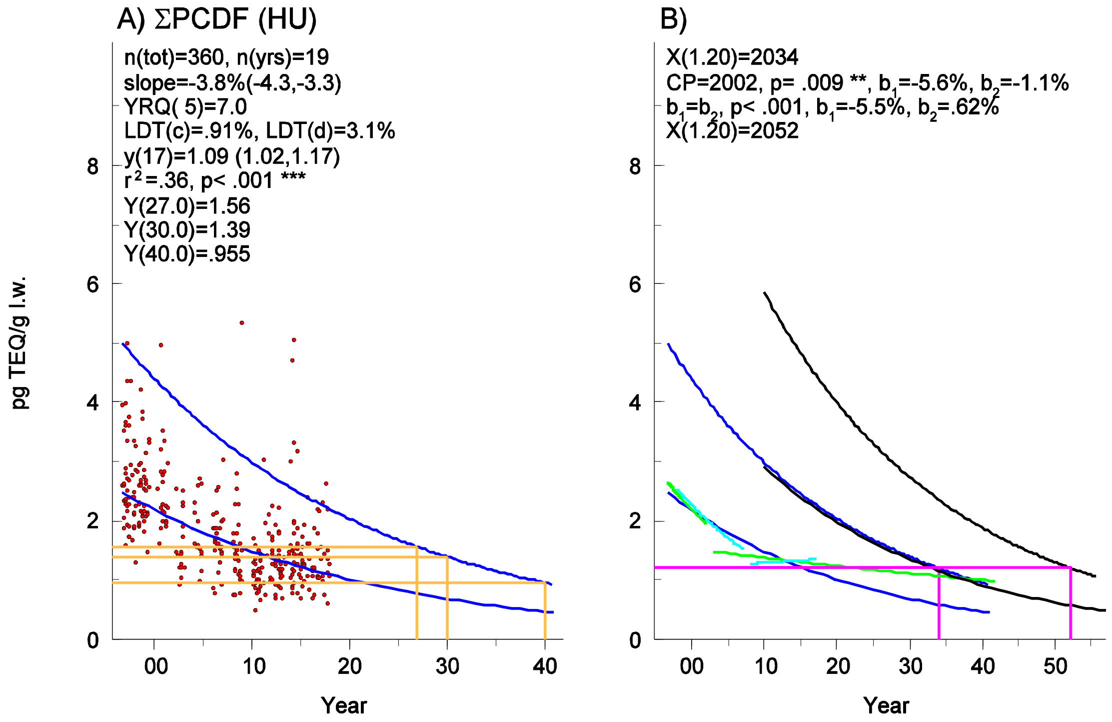


Figure 4.2. ΣPCDF, sum of furans. Human milk 1996-2017 from Uppsala (n=360). Average concentrations of ΣPCDF and upper 95% prediction (population) interval extrapolated to year 2040.

A) A significant decreasing trend of almost 3.8% a year was detected i.e. slower than for PCDD. The sample sizes and sampling frequency could potentially detect a considerably smaller trend i.e. 0.91% per year, with an 80% power for the current period of 19 years and 3.1% per year during a period of 10 years. A minimum number of 7 years would be required to detect a yearly change of 5%. Concentrations were estimated from the upper population interval at year 2027 (1.6), 2030 (1.4) and 2040 (0.96 pg TEQ/g lipid) respectively (light brown lines).

B) Estimated year when approximately 97.5% of the population were exposed to target concentrations below 0.2 x 5.9 (=1.2) pg TEQ/g lipid i.e. during year 2034 (purple line).

Black lines simulate a "worst case scenario" from an imaginary population being exposed to twice as high PCDD TEQ concentrations as the mothers in Uppsala. In such a case 97.5% of the population is estimated to reach the target value in 2052(purple).

A significant (p<0.001) difference between the slopes calculated for the period 1998 - 2006 (-5.5) and the period 2007 - 2016 (+0.86) (light blue lines). Furthermore, a significant change-point was detected up to 2002. The slope was estimated to -5.6% before 2002 and after 2002 to -1.1% a year (green lines). These are signs of a slower decrease of the sum of furans during recent years and also implies that the predicted future concentrations are somewhat overoptimistic.

2,3,4,7,8-PeCDF

This is the furan with the highest TEF: 0.3 and the contribution from PeCDF is on average about 16% of the sum TEQ.

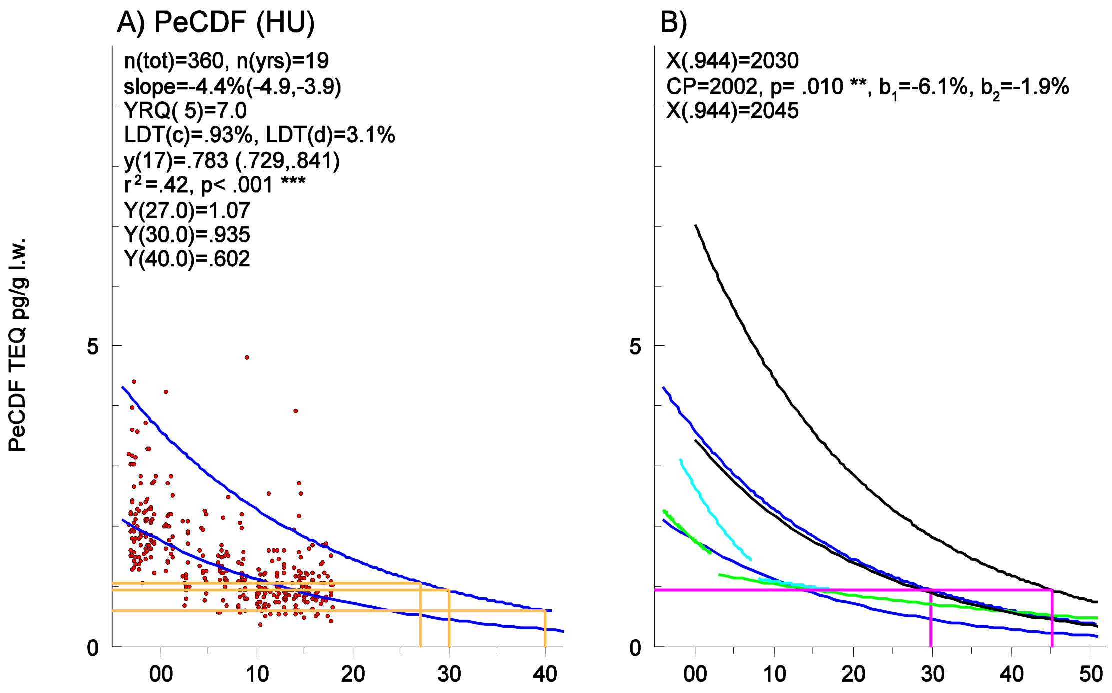


Figure 4.4. PeCDF (pg TEQ/g lipid). Human milk 1996-2017 from Uppsala (n=360). Average concentrations of 2,3,4,7,8-PeCDF and upper 95% prediction(population) interval extrapolated to year 2040.

A) A significant decreasing trend of about 4.4% a year was detected. The sample sizes and sampling frequency could potentially detect a considerably smaller trend i.e. 0.93% per year, with an 80% power for the current period of 19 years and 3.1% per year during a period of 10 years. A minimum number of 7 years would be required to detect a yearly change of 5%. Concentrations were estimated from the upper population interval at year 2027 (1.1), 2030 (0.94) and 2040 (0.60 pg TEQ/g lipid) respectively (light brown lines).

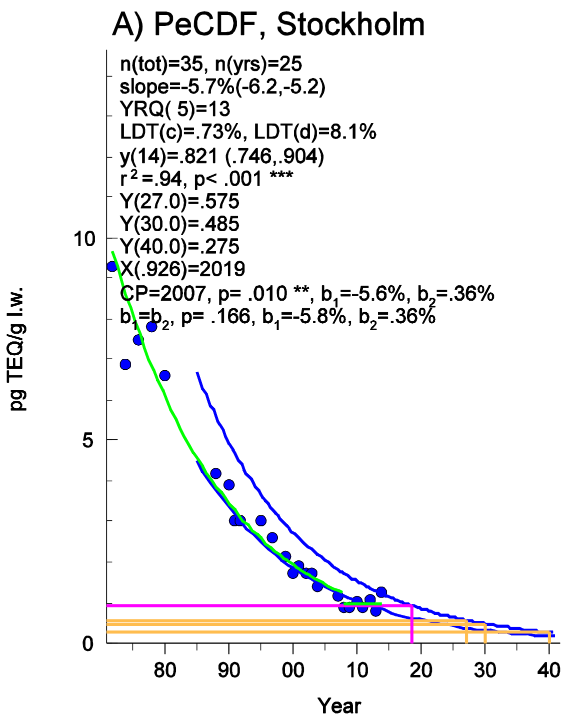
B) Estimated year when approximately 97.5% of the population was exposed to concentrations below 0.16 x 5.9 (=0.94) pg TEQ/g, i.e. around year 2030 (purple lines).

Black lines simulate a "worst case scenario" from an imaginary population being exposed to twice as high PCDD TEQ concentrations as the mothers in Uppsala. In such a case 95% of the population was below the target in 2045 (purple lines).

A significant change-point was shown, indicating a steeper slope -6.1% before 2002 and a slower (-1.9%) decrease after 2002 (green lines).

Comparing the periods 1998 - 2006 (-8.3%) and 2007 - 2016 (-1.9%), show a significant difference in the slopes and slower decrease during the latter period.

The predictions may therefore be too optimistic since the decrease seems to have slowed down.



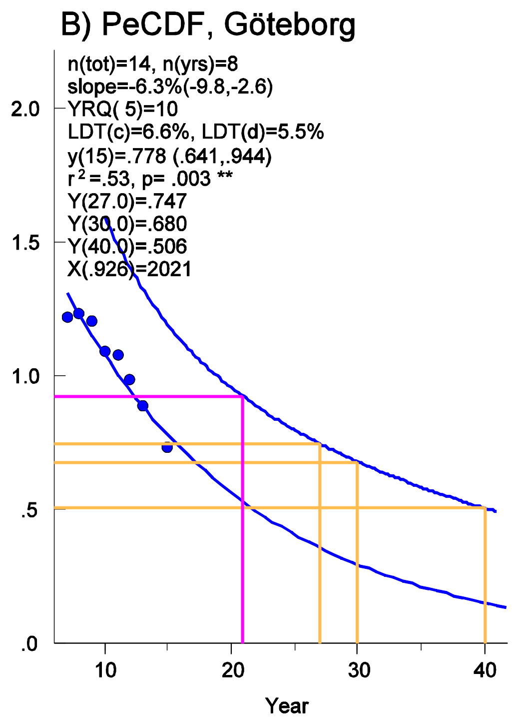


Figure 4.5. PeCDF in human milk. Average concentrations and upper 95% prediction (population) interval extrapolated to year 2040.

A) Stockholm 1972-2014 (n=35). Significant decreasing trend on average 5.7% per year.

There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016, but a change-point was identified at year 2007, with a decrease of 5.6% stops at 2007 followed by almost no trend (+0.36%).

B) Göteborg 2007-2015 (n=14). Significant decreasing trend on average 6.3% per year.

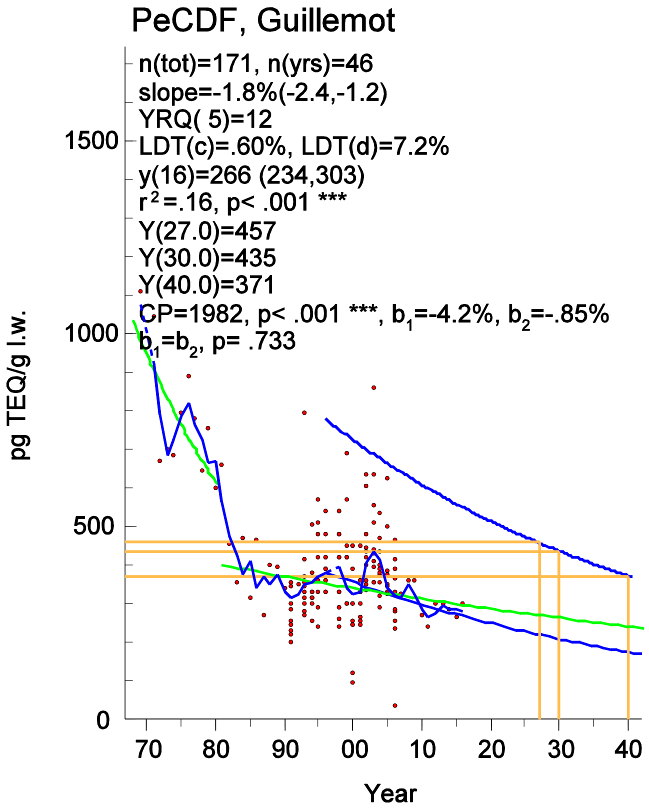


Figure 4.6. 2,3,4,7,8-PeCDF in guillemot eggs from the Baltic Proper. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A significant change-point is detected with an initial decrease of 4.1% up to 1982, and a second half decreasing at a modest pace of about 0.85% a year. Clearly, the decreasing trend has leveled off since 1980-85 but after an almost stable situation up to 2005, possibly is continue decreasing thereafter.

Comparing the periods 1998 - 2006 and 2007 - 2016, showed no significant difference in the slopes.

PeCDF, Herring

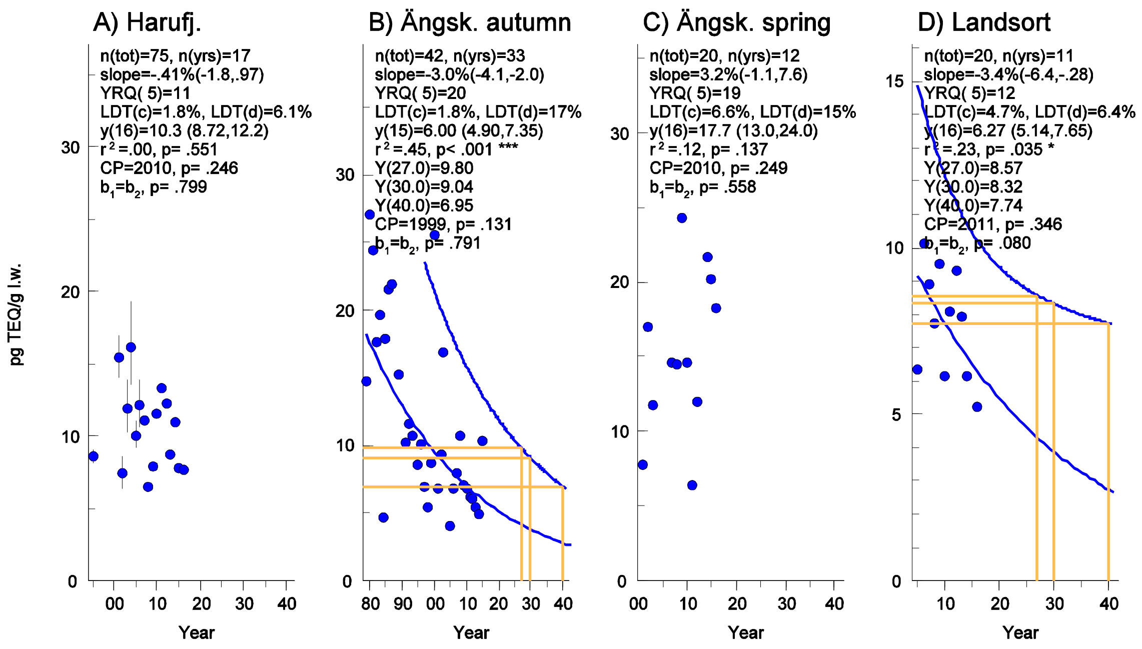


Figure 4.7. PeCDF in Baltic herring. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Bothnian Bay (Harufjärden), no significant trend. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

B) s. Bothnian Sea (Ängskärsklubb autumn), long time-series (33 year), significant decrease, on average 3% a year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

C) s. Bothnian Sea (Ängskärsklubb spring), short time-series (12 years), no significant trends. Around three times higher concentrations in the spring caught herring compared to the autumn caught herring from the same area (2015-2016).

D) n. Baltic Proper (Landsort), short time-series (11 years), significant decreasing trend, on average 3.4% a year.

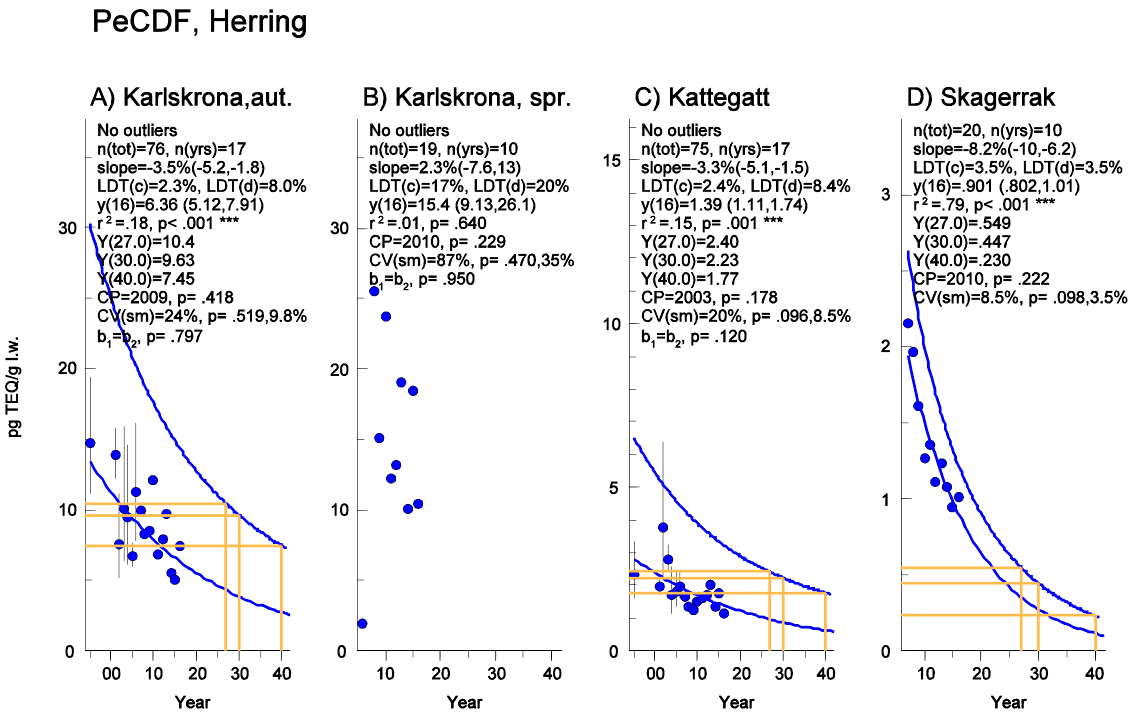


Figure 4.8. PeCDF in herring from the Baltic and the Swedish west coast. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively. The estimated herring concentrations 2016, from the s. Baltic Proper are more than four times as high as the concentrations from the Swedish west coast.

A) Autumn caught herring from the s. Baltic Proper, decreasing significantly with on average 3.5% a year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

B) Spring caught herring from s. Baltic Proper (Karlskrona), (short time-series, 10 years), show no significant trend. Concentrations more than twice as high in spring compared to autumn.

C) Kattegat (Fladen), significant decreasing trend, on average 3.5% per year. No significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

D) Skagerrak (Väderöarna). Low concentrations. Significant decreasing trend 8.2%, after adjustment for age, weight and fat content (fat content variable in the Skagerrak, see Fig. 3.8.2).

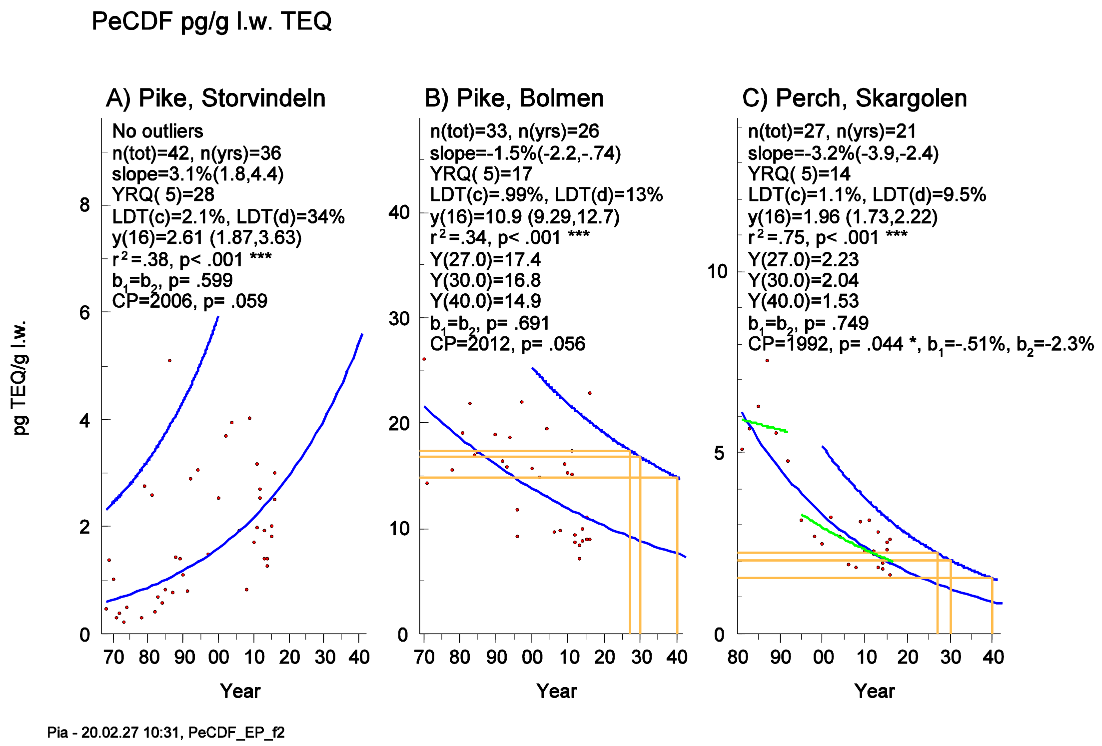


Figure 4.9. PeCDF (adjusted for weight and fat) in freshwater fish. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Pike from n. Sweden (L. Storvindeln) shows a significant ***increase*** of average 3.1% a year. The concentrations are low, several values below LOQ may confuse the picture c.f. Fig. 3.17 (PeCDD in pike from the same site)

B) Pike from s. Sweden (L. Bolmen), considerably (4 times) higher concentrations compared to pike from the north of Sweden.

C) Perch from s. Sweden (L. Skärgölen), significant downward trend of about 3% a year.

The TEQ values, expressed on a lipid weight basis, are quite high, both pike and perch are lean fish (lipid content < 1%) and therefore the fresh weight concentrations are substantially lower.

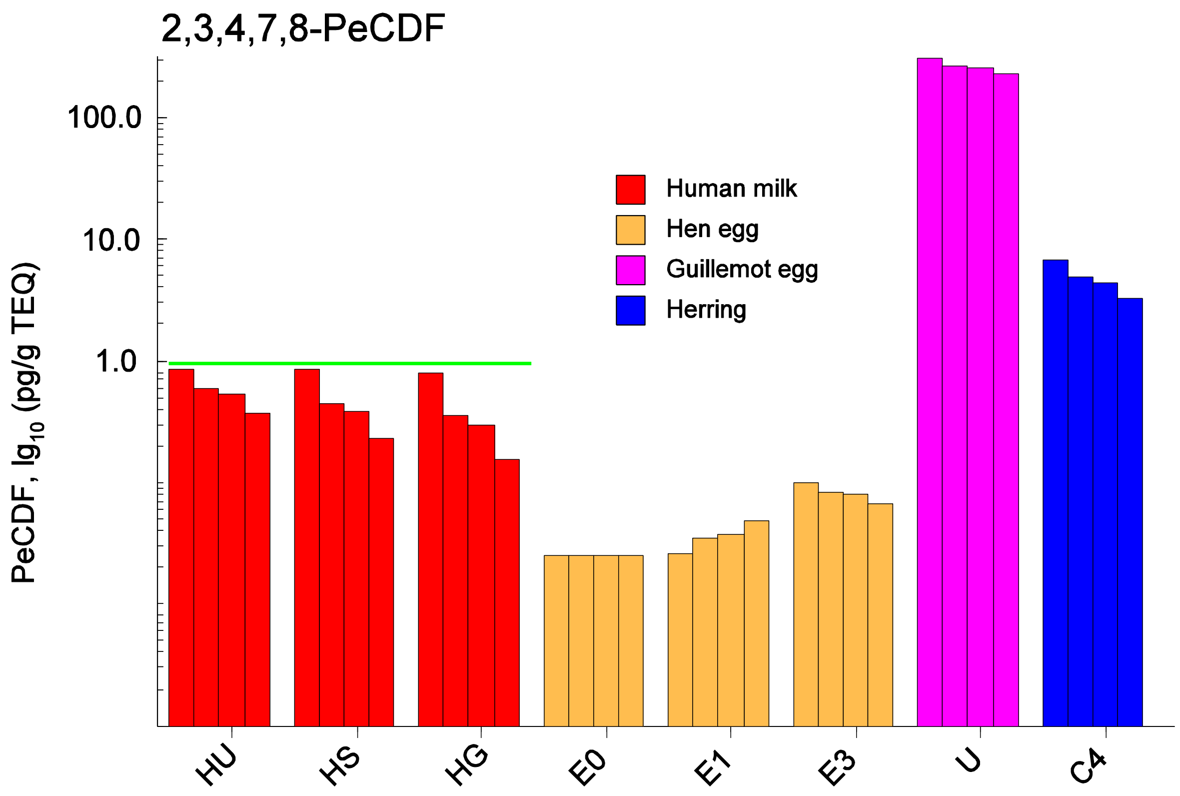


Figure 4.10. Concentrations of 2,3,4,7,8-PeCDF (TEQ lw) for various biological matrices at year 2017, 2027, 2030 and 2040. H=Human milk(red), U=Uppsala, S=Stockholm, G=Göteborg, E=hen eggs (light brown), 0= caged, 1= sputtering, 3="eco", U=Guillemot (purple), C4=Herring (blue) from s. Baltic Proper. The green line at 0.16 x 5.9 (=0.94) pg TEQ/g lipid, indicate the target concentration of PeCDF in human milk, considered as "safe" at present, assuming relative contributions from dioxin-like substances remain unchanged.

The indicated increase among E1 eggs is _not_ significant.

Summary tables for PCDFs

Furan	Matrix	N(tot)	N(grp)	Trend(%)	***	Y(X=2027)	Y(X=2030)	Y(X=2040)
PCDF	HU	360	19	-3.79	---	0.47	0.42	0.29
PeCDF	HU	360	19	-4.41	---	1.07	0.94	0.60
	HS	35	25	-5.72	---	0.58	0.48	0.28
	HG	14	8	-6.29	--	0.75	0.68	0.51
	U8	171	49	-1.78	---	456.7	435.0	371.0
	C1	75	17	-0.41				
	C2	42	33	-3.04	---	32.66	30.13	23.18
	V2	20	12	3.16				
	C3	20	11	-3.39	-	8.57	8.32	7.74
	C4	76	17	-3.49	---	10.44	9.63	7.46
	V4	19	10	2.32				
	C6	75	17	-3.30	---	2.40	2.23	1.77
	C7	20	10	-8.20	---	0.55	0.45	0.23
	ES	42	36	3.11	+++			
	EB	33	26	-1.48	---	17.42	16.78	14.89
	PS	27	21	-3.20	---	2.23	2.04	1.53

Table 4. Trends and predicted concentrations of PCDF and PeCDF (pg TEQ/g l.w.) at 2027, 2031 and 2040 from the upper 95% population interval, i.e. 97.5% of the population assumed to be below these concentrations. Matrix: H=Human milk, U=Uppsala, S=Stockholm, G=Göteborg, U8=guillemot egg, C=herring, V=spring caught herring, 1=Bothnian Bay, 2=Bothnian Sea, 3=n. Baltic Proper, 6=Kattegat, 7=Skagerrak, ES=pike from n. Sweden, EB=pike from s. Sweden, PS= fresh water perch from s. Sweden. --- = decreasing trend p<0.001, -- p<0.01, - p<0.05. Median trend including all matrices in the table= - 3.25 % per year i.e. lower than the PCDDs.

5 Dioxin-like PCBs

CB-118

CB-118 is a mono-ortho PCB with a TEF-value of 0.00003.

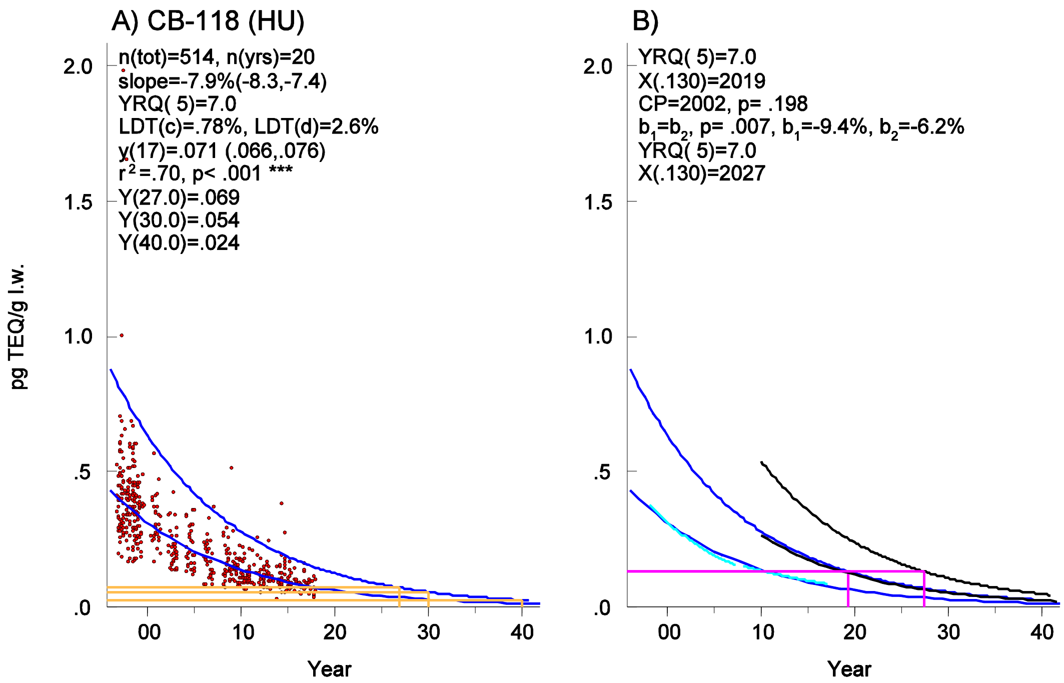


Figure 5.2. Human milk 1996-2017 from Uppsala (n=514). Average concentrations of CB-118 (adjusted for mother's age, BMI, weight gain during pregnancy and education) and upper 95% prediction (population) interval extrapolated to year 2040.

A) Concentrations were estimated from the upper population interval at year 2027 (0.069), 2030 (0.054) and 2040 (0.024 pg TEQ/g lipid) respectively. The sample sizes and sampling frequency could potentially detect a considerably smaller trend i.e. 0.78% per year, with an 80% power for the current period of 19 years and 2.6% per year during a period of 10 years. A minimum number of 7 years would be required to detect a yearly change of 5%.

B) Estimated year when approximately 97.5% of the population were exposed to target concentrations below 0.022 x 5.9 (0.13) pg TEQ/g was during 2019. Since the contribution from CB-118 to the sum of TEQ is approximately 2%, a target value of 0.13 was chosen. Below the target, exposure from dioxins, furans and dioxin-like PCBs would likely be safe provided that the relative contributions from the three groups remains similar.

A highly significant decreasing trend of almost 8% a year was detected. The sample sizes and sampling frequency could potentially detect a considerably smaller trend i.e. 0.78% per year, with an 80% power for the current period of 19 years and 2.6% per year during a period of 10 years. A minimum number of 7 years would be required to detect a yearly change of 5%.

No significant change-point was detected. The periods 1998 - 2006 (-9.4%) and 2007 - 2016 (-6.2%), showed a significant difference in the slopes but this would not affect the predicted concentrations that much.

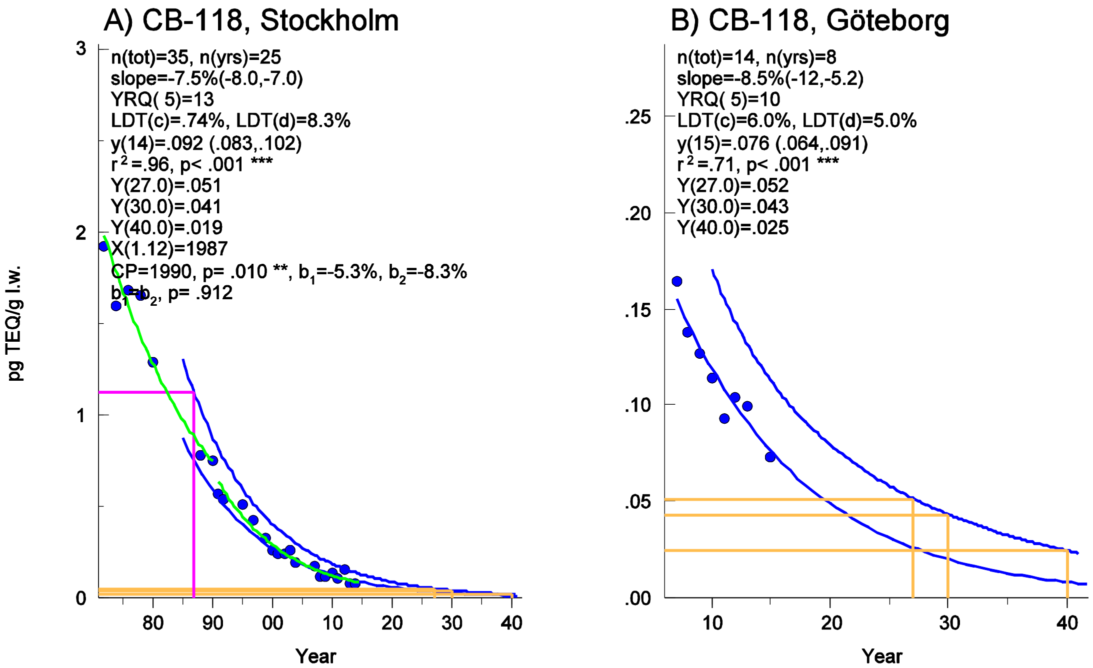


Figure 5.3. CB-118. Human milk. Average concentrations of PeCDD and upper 95% prediction (population) interval extrapolated to year 2040 (see statistics reported in the Figure).

A) Stockholm 1972-2014 (n=35) Significant decreasing trend on average 7.5% per year.

There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016, but a change-point was identified at year 1990, however the second part (-8.3%, steeper than the first period, -5.3%) follows the total regression quite close.

B) Göteborg 2007-2015 (n=14). Significant decreasing trend on average 8.5% per year.

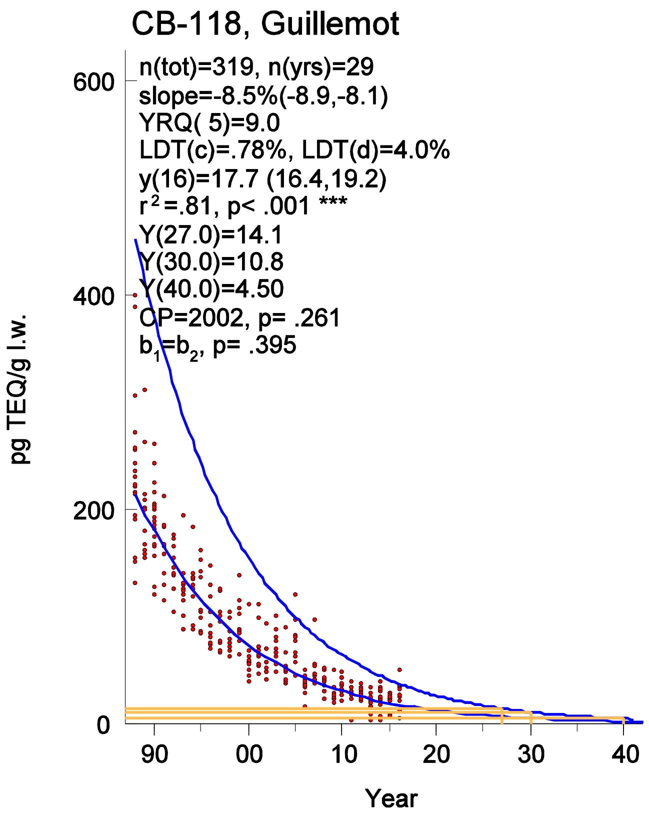


Figure 5.3. CB-118 in guillemot eggs from the Baltic Proper. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively (see statistics reported in the Figure).

No significant change-point was detected. Comparing the periods 1998 - 2006 and 2007 - 2016, show no significant difference in the slopes.

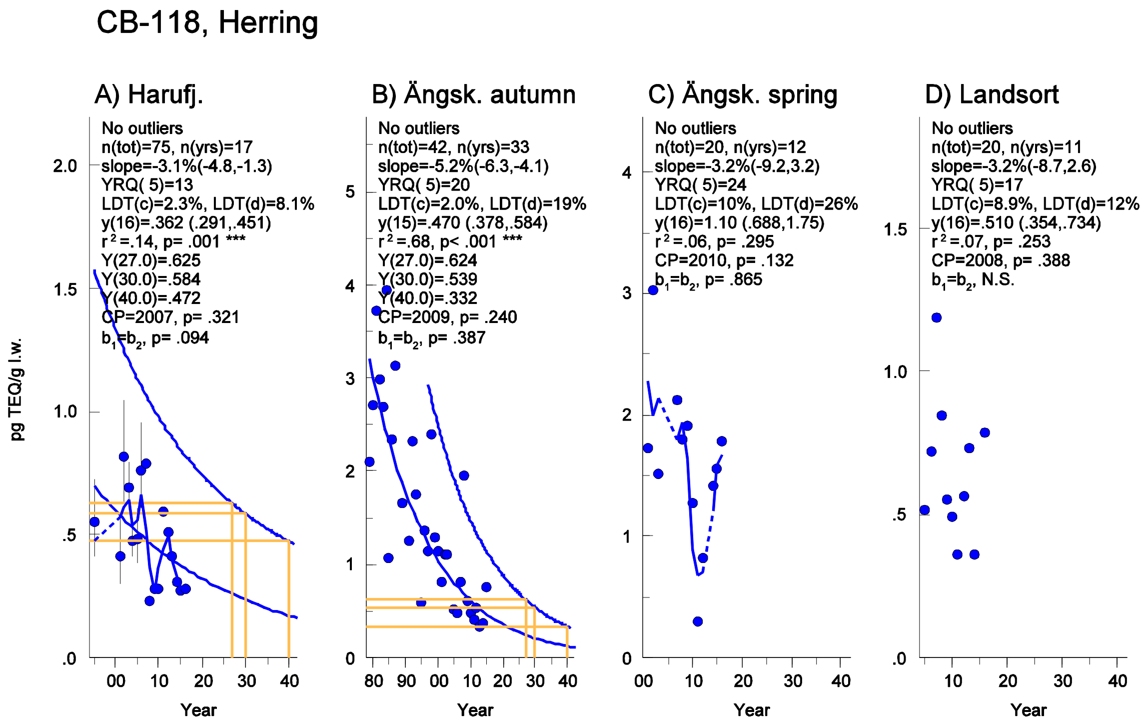


Figure 5.4. CB-118 in Baltic herring. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Bothnian Bay (Harufjärden), significant trend decreasing trend, 3.1% a year.

B) s. Bothnian Sea (Ängskärsklubb autumn), long time-series (33 year), significant decrease, on average 5.2% a year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

C) s. Bothnian Sea (Ängskärsklubb spring), short time-series (12 years), no significant trends.

D) n. Baltic Proper (Landsort), short time-series (11 years), no significant trends.

There were no significant differences between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016 and no significant change-points for any of these sites.

CB-118, Herring

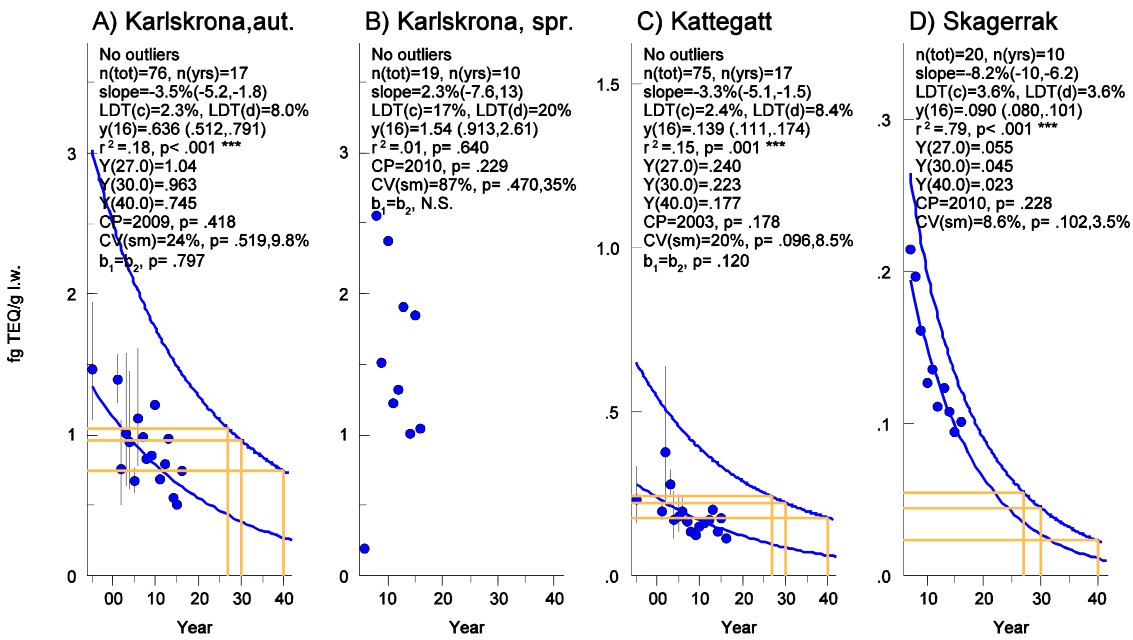


Figure 5.5. CB-118 in herring from the Baltic Proper and the Swedish west coast. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively. The estimated herring concentrations 2016, from the s. Baltic Proper are more than four times as high as the concentrations from the Swedish west coast.

A) Autumn caught herring from the s. Baltic Proper (Karlskrona), decreasing with 3.5% a year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

B) s. Baltic Proper (Karlskrona), spring caught herring, show no significant trend.

C) Kattegat (Fladen), significant decreasing trend, on average 3.3% per year. No significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

D) Skagerrak (Väderöarna). Low concentrations. Significant decreasing trend ***8.2%*** a year, after adjustment for age, weight and fat content (fat content variable in the Skagerrak).

CB-118 pg/g l.w. TEQ

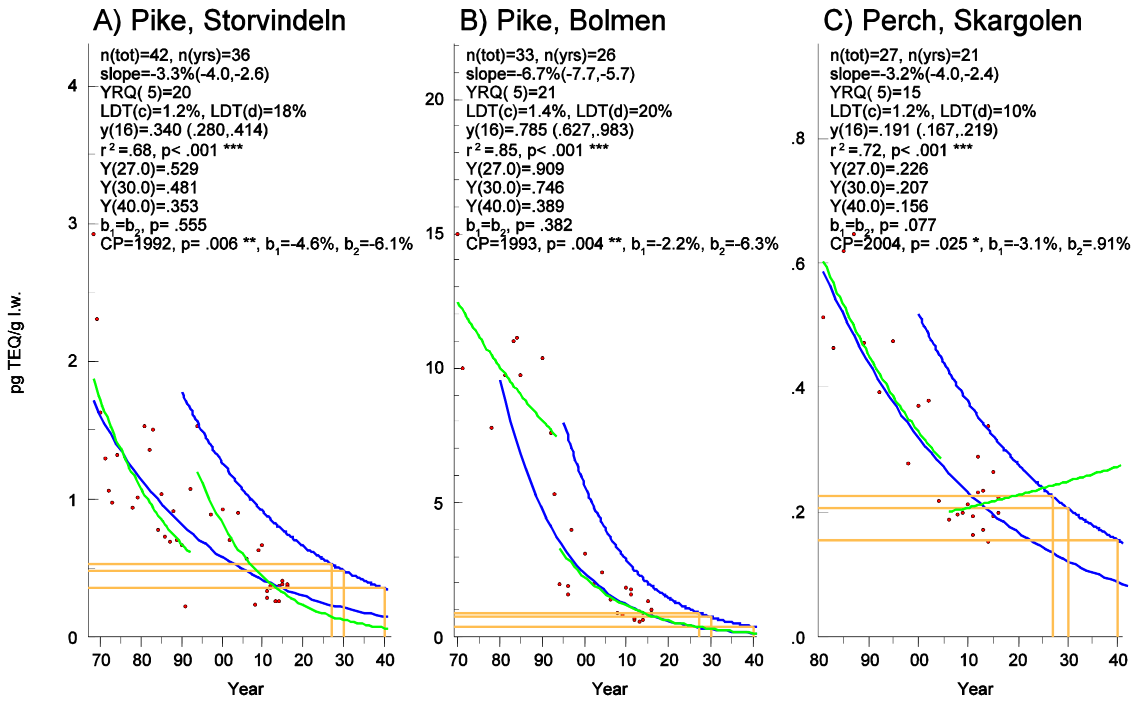


Figure 5.6. CB-118 (adjusted for weight and fat) in freshwater fish. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Pike from n. Sweden (L. Storvindeln) shows a significant decrease of average 3.3% a year.

B) Pike from s. Sweden (L. Bolmen), decreasing with on average 6.7% a year. More than twice as high concentrations compared to pike from the north of Sweden.

C) Perch from s. Sweden (L. Skärgölen), significant downward trend of about 3% a year.

The TEQ values, expressed on a lipid weight basis, are quite high, both pike and perch are lean fish (lipid content < 1%) and therefore the fresh weight concentrations are substantially lower.

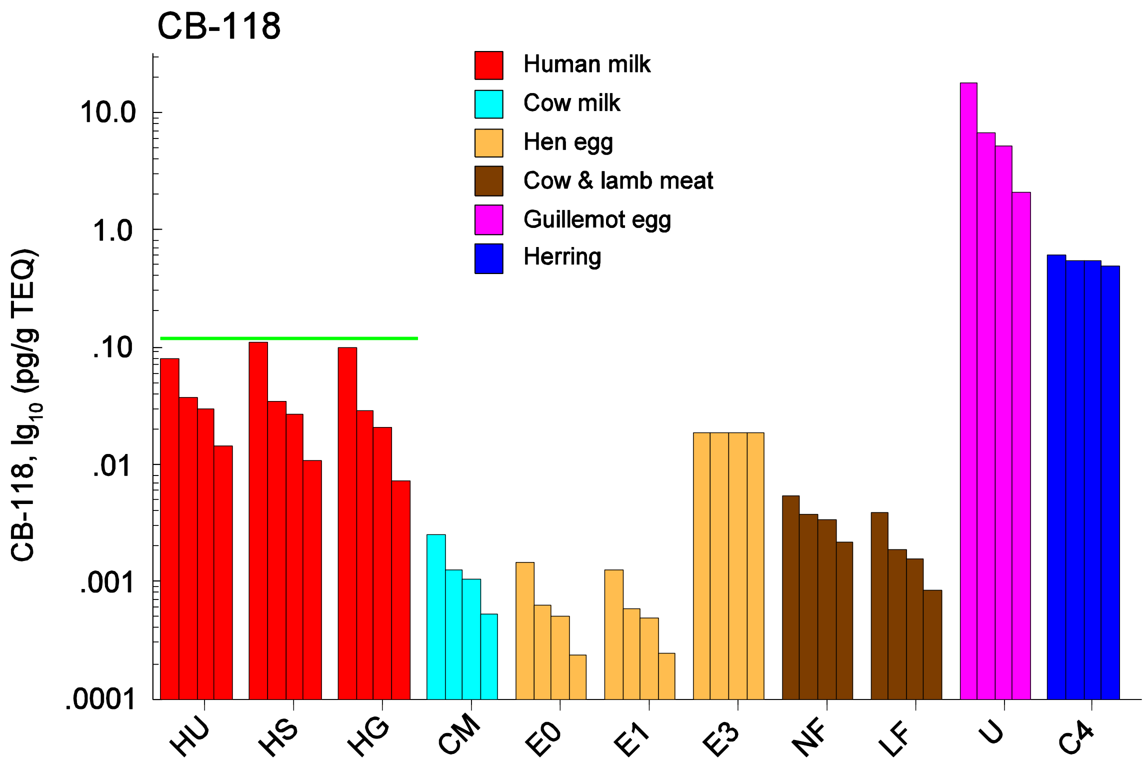


Figure 5.3. Concentrations of CB-118 (TEQ lw) for various kinds of biological matrices at year 2017, 2027, 2030 and 2040. H=Human milk (red), U=Uppsala, S=Stockholm, G=Göteborg, CM=Cow milk (light blue), E=hen eggs (light brown), 0= caged, 1= sputtering, 3="eco", NF=Cattle fat (brown), LF=Lamb fat (brown), U=Guillemot (purple), C4=Herring (blue) from s. Baltic Proper. The green line at 0.02 x 5.9 (0.12) pg TEQ/g lipid, indicate the concentration of CB-118, considered as "safe" at present, assuming relative contributions from dioxin-like substances remain unchanged.

CB-126

CB-126 is a non-ortho-PCB with a TEF-value of 0.1. It is the major contributor of the PCB to the sum of TEQ in human milk, about 33% of the sum.

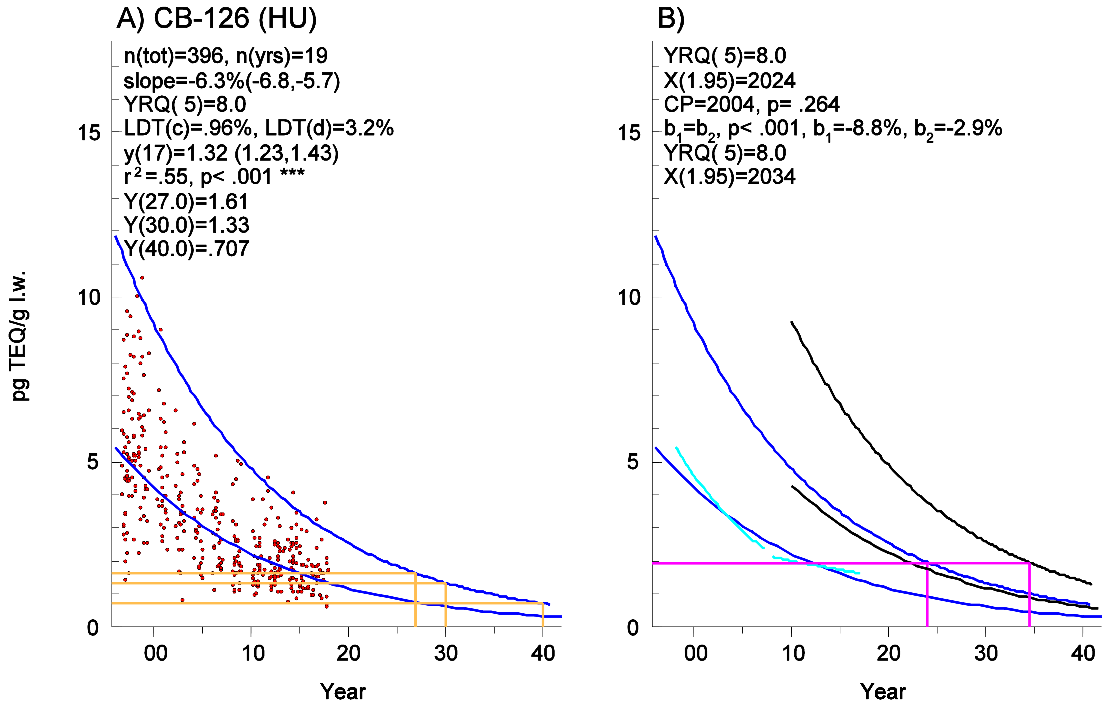


Figure 5.5. Human milk 1996-2017 from Uppsala (n=396). Average concentrations of CB-126 (adjusted for mother's age, BMI, weight gain during pregnancy and education) and upper 95% prediction (population) interval extrapolated to year 2040.

A) Concentrations were estimated from the upper population interval at year 2027 (1.6), 2030 (1.3) and 2040 (0.71 pg TEQ/g lipid) respectively.

B) Estimated year when approximately 97.5% of the population are exposed to target concentrations below 0.33 x 5.9 (=1.9) pg TEQ/g lipid, is during year 2024. Since the contribution from CB-126 to the sum of TEQ is approximately 33%, a target value of 1.947 was chosen. Below the target, exposure from dioxins, furans and dioxin-like PCBs would likely be safe provided that the relative contributions from the three groups remains similar.

Black lines simulate a "worst case scenario" from an imaginary population being exposed to twice as high PCDD TEQ concentrations as the mothers in Uppsala. In such a case 97.5% of the population was below the target in 2034.

Comparing the periods 1998 - 2006 (-8.8%) and 2007 – 2016 (-2.9%) a significant difference between the slopes was found (light blue lines). This may imply overoptimistic predicted years based on the latter decline.

No significant change-point was shown.

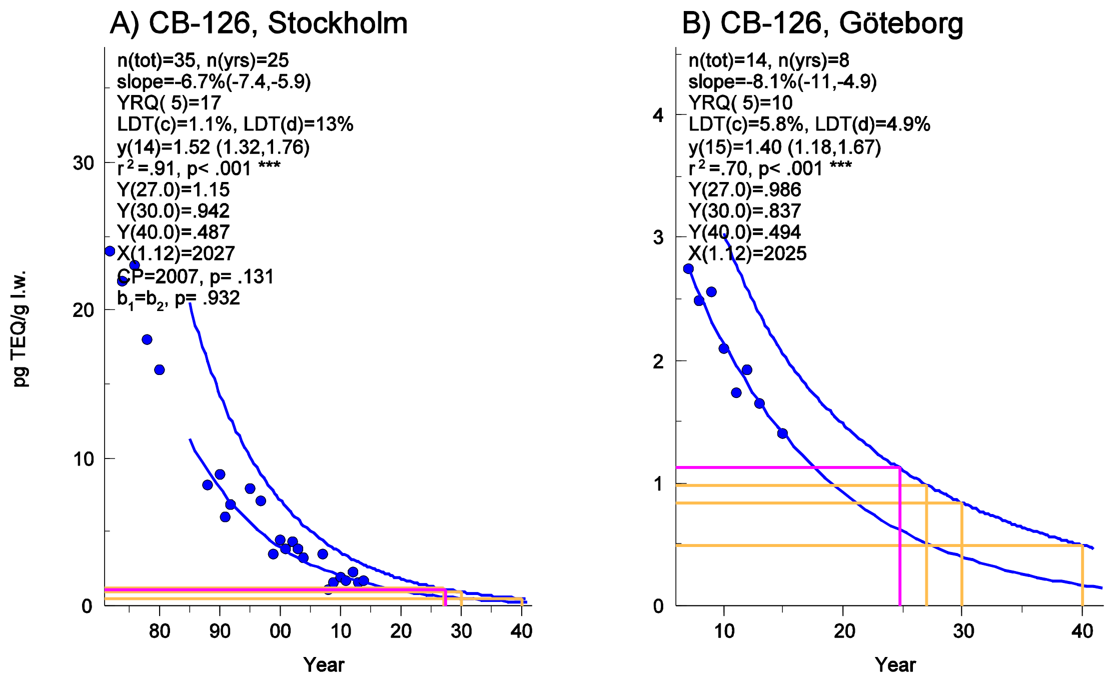


Figure 5.6. CB-126. Human milk. Average concentrations of PeCDD and upper 95% prediction (population) interval extrapolated to year 2040 (see statistics reported in the figure).

A) Stockholm 1972-2014 (n=35) Significant decreasing trend on average 6.7% per year.

There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016, and no change-point was identified.

B) Göteborg 2007-2015 (n=14). Significant decreasing trend on average 8.1% per year.

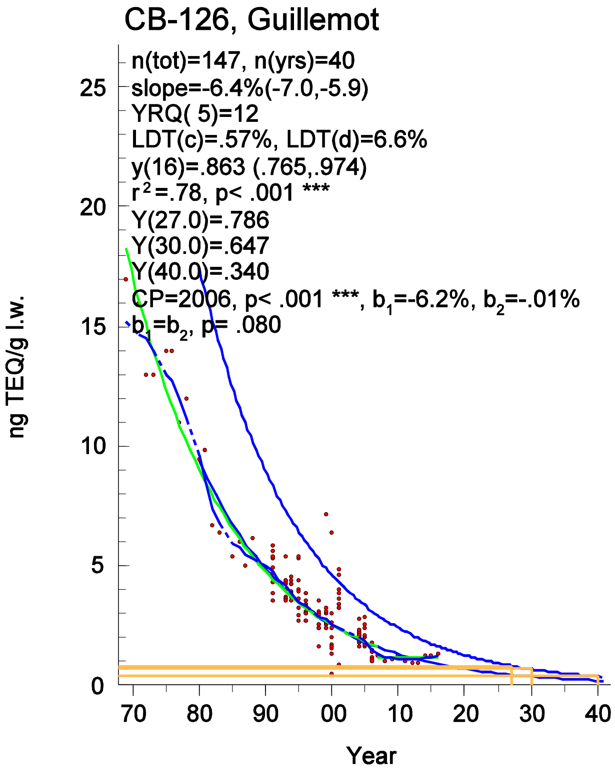


Figure 5.7. CB-126 in guillemot eggs from the Baltic Proper. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A significant change-point was detected, year 2006, indicating that the decrease of 6.2% up to 2006 has almost ceased (-0.01%) from that year (green lines).

Comparing the periods 1998 - 2006 and 2007 - 2016, show no significant difference in the slopes.

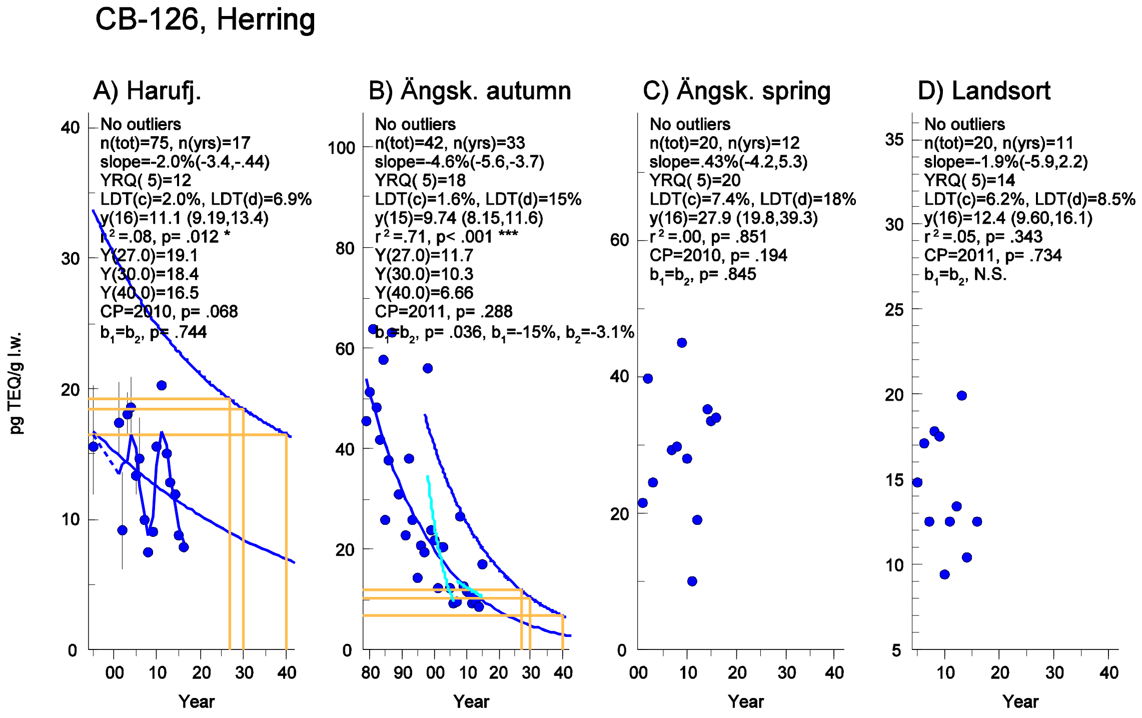


Figure 5.8. CB-126 in Baltic herring. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively.

A) Bothnian Bay (Harufjärden), significant trend decreasing trend, 2% a year.

B) Southern Bothnian Sea (Ängskärsklubb autumn), long time-series (33 year), significant decrease, on average 4.6% a year. There is a significant difference between the slopes calculated for the period 1998 - 2006 (-15%) and the period 2007 - 2016 (-3.1%) (light blue lines).

C) Southern Bothnian Sea (Ängskärsklubb spring), short time-series (12 years), no significant trends. Also for CB-126, the average concentrations are considerably higher in spring compared to autumn caught herring (almost 3 times for year 2016, in the example above).

D) Northern Baltic Proper (Landsort), short time-series (11 years), no significant trends.

There were no significant change-points for any of these sites.

CB-126, Herring

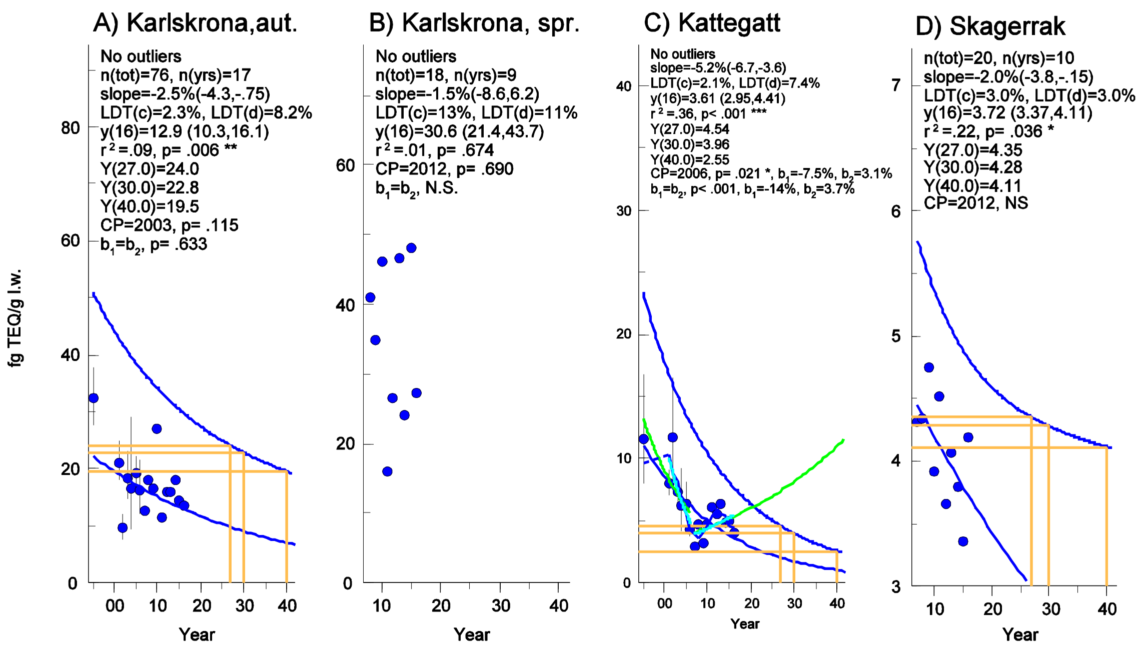


Figure 5.9. CB-126 in herring from the Baltic Proper and the Swedish west coast. Geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively. The estimated herring concentrations 2016, from the s. Baltic Proper are more than four times as high as the concentrations from the Swedish west coast.

A) Autumn caught herring from the s. Baltic Proper (Karlskrona), decreasing with 2.5% a year. There is no significant difference between the slopes calculated for the period 1998 - 2006 and the period 2007 - 2016.

B) s. Baltic Proper (Karlskrona), spring caught herring, show no significant trend. Estimated mean concentration for 2016 is more than twice as high as the one estimated for autumn caught herring from the same area.

C) Kattegat (Fladen), significant decreasing trend, on average 5.2% per year. A significant difference between the slopes calculated for the period 1998 - 2006 (-14%) and the period 2007 - 2016 (+3.7%) (light blue lines) as well as a change-point year 2006 (green lines), -7.5% before 2006 and +3.1% after.

D) Skagerrak (Väderöarna). Low concentrations. Significant decreasing trend 2% a year, after adjustment for age, weight and fat content (fat content variable in the Skagerrak).

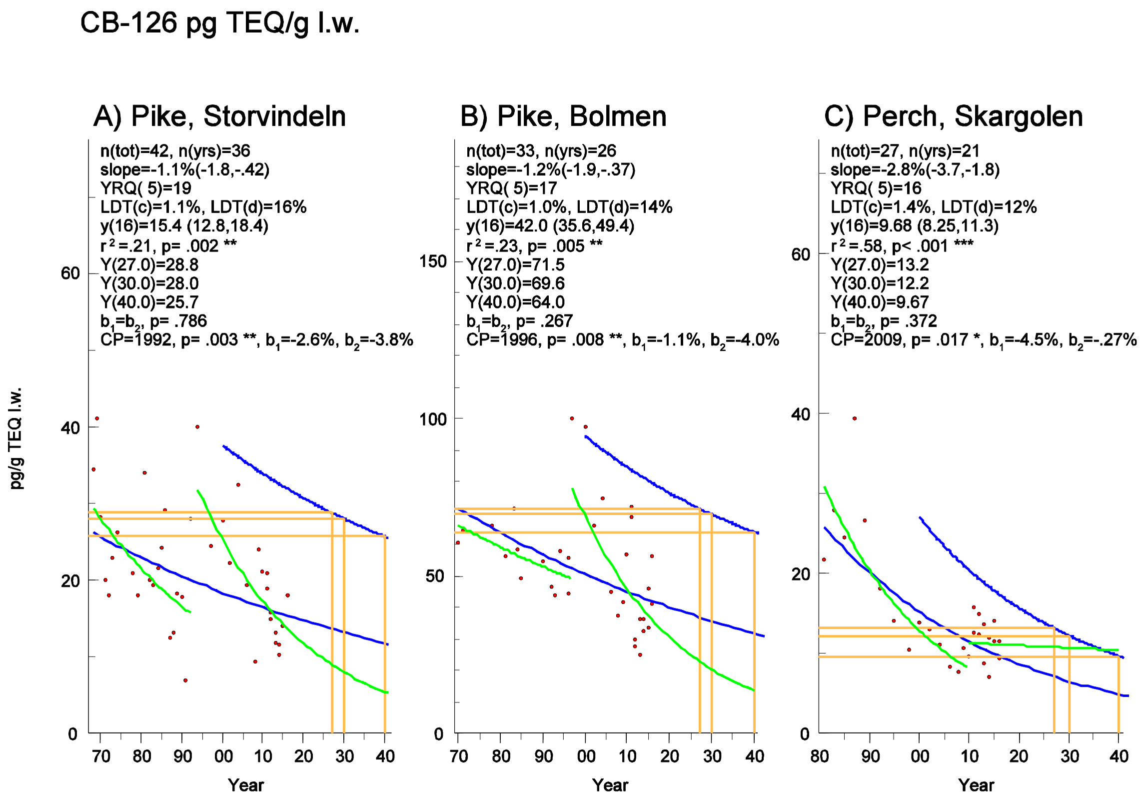


Figure 5.10. CB-126 in freshwater pike and perch.

A) Pike from n. Sweden (L. Storvindeln) showing a significant decrease, on average 1.1% a year. There is also a significant change-point with a gap 1992. We have no explanation of this gap and it might have occurred just by chance. The slopes of the two parts are decreasing in a similar way (-2.6%, -3.8% respectively).

B) Pike from s. Sweden (L. Bolmen), also here the concentrations are decreasing significantly on average with 1.2%, similar to the pike in northern Sweden and also here there is a significant change-point with a gap (-1.1%, -4.0% respectively). The change-point occur 1996, i.e. 4 years later than the northern pike (making it less likely that the gap is caused by contamination of the samples).

C) Perch from s. Sweden (L. Skärgölen) geometric mean and upper 95% prediction (population) interval extrapolated to year 2027, 2030 and 2040, respectively, showing an average decrease of 2.8% a year. After a significant change-point 2009, the initial decrease (-4.7%) seem to level out (-0.3%). The first part cover 12 years, the second only 6.

The TEQ values, expressed on a lipid weight basis, are quite high, both pike and perch are lean fish (lipid content < 1%) and therefore the fresh weight concentrations are substantially lower.

Summary tables for dioxin-like PCBs

*PCB	Matrix	N(tot)	N(years)	Trend(%)	***	Y(X=2027)	Y(X=2030)	Y(X=2040)
CB-118	HU	514	20	-7.86	---	0.07	0.05	0.02
	HS	35	25	-7.48	---	0.05	0.04	0.02
	HG	14	8	-8.52	---	0.05	0.04	0.02
	U8	319	29	-8.52	---	14.14	10.85	4.50
	C1	75	17	-3.07	---	0.63	0.58	0.47
	C2	42	33	-5.19	---	0.62	0.54	0.33
	V2	20	12	-3.24		0.02	0.03	0.04
	C3	20	11	-3.21		0.01	0.01	0.02
	C4	76	17	-3.49	---	1.04	0.96	0.75
	V4	19	10	2.32				
	C6	75	17	-3.30	---	0.24	0.22	0.18
	C7	20	10	-8.20	---	0.05	0.04	0.02
	ES	42	36	-3.31	---	0.53	0.48	0.35
	EB	33	26	-6.70	---	0.91	0.75	0.39
	PS	27	21	-3.16	---	0.23	0.21	0.16
CB-126	HU	396	19	-6.24	---	1.61	1.33	0.71
	HS	35	25	-6.69	---	1.15	0.94	0.49
	HG	14	8	-8.09	---	0.99	0.84	0.49
	U8	147	40	-6.42	---	0.79	0.65	0.34
	C1	75	17	-1.95	-	19.14	18.44	16.50
	C2	42	33	-4.64	---	11.75	10.29	6.66
	V2	20	12	0.43				
	C3	20	11	-1.90				
	C4	76	17	-2.54	--	24.03	22.82	19.52
	V4	19	10	14.26				
	C6	75	17	-5.20	---	4.54	3.96	2.55
	C7	20	10	-1.99	-	4.35	4.28	4.11
	ES	42	36	-1.11	--	28.78	28.02	25.70
	EB	33	26	-1.15	--	71.50	69.60	63.95
	PS	27	21	-2.77	---	13.16	12.24	9.67

Table 4. Trends and predicted concentrations of the dioxin-like CB-118 and CB-126 (pg TEQ/g lipid) at 2027, 2031 and 2040 from the upper 95% population interval, i.e. 97.5% of the population assumed to be below these concentrations. Matrix: H=Human milk, U=Uppsala, S=Stockholm, G=Göteborg, U8=guillemot egg, C=herring, V=spring caught herring, 1=Bothnian Bay, 2=Bothnian Sea, 3=n. Baltic Proper, 6=Kattegat, 7=Skagerrak, ES=pike from n. Sweden, EB=pike from s. Sweden, PS= fresh water perch from s. Sweden. --- = decreasing trend p<0.001, -- p<0.01, - p<0.05. Median trend = - 3.3 % per year i.e. lower than the PCDDs and approximately the same as for the PCDFs ***but*** the average trend for the three human milk series is almost -8% a year i.e. more than the PCDFs and about the same as for the PCDDs in human milk.

Conclusions

Generally, trends of dioxin concentrations in both human milk, food and wildlife are decreasing significantly. In some time series significant change-points were identified that showed a slower decrease during recent years. When the time series was divided in two periods, 1998-2006 and 2007-2016, in some cases a slower decrease was observed during the latter period. There are rather clear indications that the PCDFs (furans) have decreased at a slower rate or much slower rate than the PCDDs (dioxins) during recent time (e.g. Fig.1.3). If this slower rate of decrease persists in the future, the estimated concentrations for 2027, 2030 and 2040 maybe somewhat underestimated. On the other hand, to compensate for the uncertainty, the distance from the population interval to the regression line will steady increase as the year (x-value) diverges from the mean year (x-mean). This tendency will work in the other direction (i.e. "overestimate" the concentrations).

The dioxin concentrations in fat fish from the Baltic is still higher than on the west coast, probably too high for unlimited consumption, but this has to be determined in a new risk assessment by the Livsmedelsverket. Note that there are considerable both geographical (increasing levels to the south, fresh water fish) and seasonal (higher levels in spring compared to autumn caught, herring) variation in fish. Dioxin concentrations are also expected to be higher in older fish (other factors kept constant). Saltwater fish, like herring, in the northern Baltic with low salinity grow slower than in the southern, more saline, Baltic. Therefore herring from the northern Baltic have higher dioxin concentrations than herring from the southern Baltic when comparing fish of the same size.

The dioxin concentration in humans are affected by the concentrations in the food that may be produced in areas far away from Sweden and the Baltic Sea area. Therefore the link today between the concentration in human milk and the Swedish environment is strongest for milk and other dairy products, eggs, meat, etc. produced in Sweden, and especially fat fish from the greater lakes in Sweden and the Baltic. The consumption pattern has changed and may continue to vary over time. Dioxin levels in imported food is also decreasing.

A reasonable target for daily intake at TWI is 17 pg per day, as determined for young women weighing 60 kg. The food basket data indicated that for the average young woman in Sweden this level is reached in year 2053, for the upper population interval, not until 2145, provided the trend continue over the years.

Forecasts based on extrapolations are *uncertain*. Dioxins are produced unintentionally and the sources are diverse. Measures have been taken to reduce known sources, the effect of these measures are shown in the decreasing trends. The efforts to reduce discharges further than is done today can be expected to become more difficult and expensive than today. Contributions from sources difficult to control e.g. increased combustion of biomass from the forestry industry.

The calculated TEQ-values are based on the TEF-values from 2005, and will change at each reevaluation of the TEF-values.

References

Assefa A, Tysklind M, Bignert A, Wiberg K (2019) Sources of polychlorinated dibenzo-p-dioxins and dibenzofurans to Baltic Sea herring. *Chemosphere 218 (493-500)*

Bignert A, Danielsson S, Ek C, Faxneld S, Nyberg E (2017) Comments Concerning the National Swedish Contaminant Monitoring Programme in Marine Biota, 2017 (2016 years data), *Rapport till Naturvårdsverket. Naturhistoriska riksmuseet, Rapport nr 10:2017, 341 pp.*

Bignert A (2013) The PIA application for statistical analysis of time-series data. Available: http://www.amap.no/documents/index.cfm?dirsub=/PIA%20Trend%20Analysis%20.

Glynn A, *et al.* (2020)

Glynn A, Aune M, Fridén U, Lignell S, Darnerud PO, Bignert A (2016) Temporal trends of PCDD/Fs and PCBs in mother´s milk in Sweden are the current maternal body burdens safe for the fetus? *Organohalogen Compounds Vol. 78, (2016)*

Gyllenhammar I, Glynn A, Fridén U, Cantillana T, Aune M, Lignell S, Bignert A (2017) Levels of persistent halogenated organic pollutants (POP) in mother's milk from first-time mothers in Uppsala, Sweden: results from year 2015 and 2016, and temporal trends for the time period 1996-2016. *Rapport till Naturvårdsverkets hälsorelaterade miljöövervakning, 17 pp.*

Haglund P, Malmvärn A, Bergek S, Bignert A, Kautsky L, Nakano T, Wiberg K, Asplund L (2007) Brominated Dibenzo-p-Dioxins - A New Class of Marine Biotoxins. *Environ. Sci. Technol., 41(9), 3069-3074.*

Nyberg E, Faxneld S, Danielsson S, Bignert A (2018) The National Swedish Contaminant Monitoring Programme for Freshwater Biota, 2018. *Rapport till Naturvårdsverket. Naturhistoriska riksmuseet, Rapport nr 3:2018, 173 pp.*

Nyberg E, Aune M, Awas R, Benskin J, Bergh A, Bignert A, Dahlgren H, Danielsson S, de Wit C, Egebäck A-L (2017) Monitoring of POPs in human milk from Stockholm and Gothenburg, 1972-2015**.** *Rapport till Naturvårdsverket. Naturhistoriska riksmuseet, Rapport nr 9:2017, 63 pp.*

Sturludottir E, Gunnlaugsdottir H, Nielsen OK, Stefansson G (2017) Detection of a change-point, a mean - shift accompanied with a trend change, in short time - series with autocorrelation. *Commun Stat Simul Comput 46:5808–5818.*

Sundqvist KL, Tysklind M, Cato I, Bignert A, Wiberg K (2009) Levels and homologue profiles of PCDD/Fs in sediments along the Swedish coast of the Baltic Sea. *Environ Sci Pollut Res Int. 16(4):396-409.*

U.S. EPA (2010) Recommended toxicity equivalence factors (TEFs) for human health risk assessments of 2,3,7,8-tetrachlorodibenzo-p-dioxin and dioxin-like compounds. *EPA/100/R-10/005. https://archive.epa.gov/raf/web/pdf/tefs-for- dioxin-epa-00-r-10-005-final.pdf*

Wiberg K, Assefa AT, Sundqvist K L, Cousins IT, Johansson J, McLachlan MS, Sobek A, Cornelissen G, Miller A, Hedman J, Bignert A, Peltonen H, Kiljunen M, Shatalov V, Cato I, (2013) Managing the dioxin problem in the Baltic region with focus on sources to air and fish - Final report from the research project BalticPOPs. *The Swedish Environmental Protection Agency. Report 6566 • May 2013. ISBN 978-91-620-6566-9. 133 pp.*