TEMPORAL TRENDS AND PATTERNS OF PCBs IN THE SWEDISH MARINE AND FRESHWATER ENVIRONMENT

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Introduction

The Baltic is a 377,000 km² inland sea with brackish water and inhabited by a mixture of marine and freshwater species. The marine species are dependent of an inflow of water with higher salinity from the North Sea. The Baltic was found to be severely polluted by organic contaminants (e.g. PCB and DDT) in the 1960s[1, 2], and the high levels of these contaminants most probably caused major reproduction problems within the seal population in the Baltic[3, 4, 5]. The finding of PCB and DDT in the Swedish environment was the starting point of a continuous temporal monitoring of contaminants in biota in the Swedish marine, freshwater and terrestrial ecosystems. All new use of PCB was banned in Sweden in 1978 and PCB's have decreased significantly since the start of the monitoring, but the levels are still higher in the Baltic than in for example the North Sea. This study compares congener patterns of PCBs in the Swedish freshwater and marine environment and the rates of decrease for different congeners and temporal trends are also discussed.

Materials and methods

Fish Sampling

The samples within the Swedish marine and freshwater monitoring programme of contaminants in biota are all from locally unpolluted areas and collected annually. Herring is the most frequently used species in the marine programme (cod, eelpout, perch, blue mussels and guillemot eggs are also monitored) and perch in the freshwater programme (pike, arctic char and roach are also monitored). The earliest time-series in the marine environment are from the beginning of the 70-ies. Five sites of herring samples have been monitored since the beginning of the 80-ies: Bothnian Bay, s Bothnian Sea, n Baltic Proper, s Baltic Proper and the Kattegat. The present marine monitoring programme for herring and perch consists of 20 and two sampling sites respectively, along the Swedish coast. The earliest time-series in the freshwater environment are from the end of the 60-ies (pike from two lakes, one representing the arctic region in Sweden). Perch has only been analyzed for a few years, but is collected from 27 lakes within the freshwater programme. Results from a study of contaminants in perch, performed in cooperation with the costal county boards in northern Sweden in 2009, is also included in the analysis. The sampling sites within this study are selected for examination of local differences and are not necessarily un affected by local sources. The collection and sample preparation, both within the monitoring programme and in the study from 2009, follows internationally agreed guidelines[6, 7].

Chemical analysis

The samples were extracted with a mixture of polar and non-polar solvents. The lipid content of the organic phase was determined gravimetrically. After clean-up of the dissolved lipid extracts with concentrated sulphuric acid, the samples were analyzed on a gas chromatograph equipped with a μ-electron capture detector. Two 60 m columns with different polarity were used. [8, 9]

Results and discussion:

Temporal trends

The concentrations of PCBs (here illustrated by CB153 in Fig. 1) are generally decreasing with about 3-10 percent per year both in Swedish freshwater and marine fish species (e.g. pike, arctic char, herring and Baltic perch). The decrease is generally in the lower part of the range for freshwater fish. Decreasing trends for PCBs in biological samples of similar magnitudes are reported in studies from many other countries [10, 11, 12, 13]. The number of years to detect a significant annual change of 10 %, with an 80% statistical power for CB-153 in herring varies between 10-14 years (Fig. 1).

The ratios between the penta-PCBs and hexa-PCBs, illustrated by CB-101/CB-153 in Fig. 2, are decreasing over the whole time period for both the freshwater and the marine sampling sites. This may be expected due to higher degree of volatilization and degradation in lower chlorinated PCBs. However, during the last decade, this decrease seems to have leveled out at most monitored sites. It is important that further studies are conducted to monitor the cease of this decline, possibly caused by releases of PCBs from new sources.

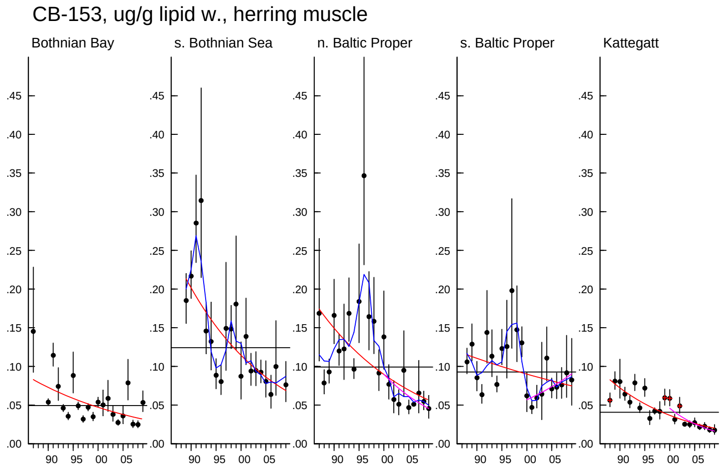

Figure 1. Log-linear trends of CB-153 (ug/g lipid weight) in herring muscle from the Bothnian Bay, southern Bothnian Sea, northern Baltic Proper, southern Baltic Proper and the Kattegatt (time series starting in 1987, 1989, 1987, 1988 and 1988 respectively). The red line shows a significant trend over the whole time period (the dotted red line, 0.05< p<0.1), the magenta colored line a significant trend for the last ten years, the blue line a non linear trend and the dotted line the mean concentration over the whole period.

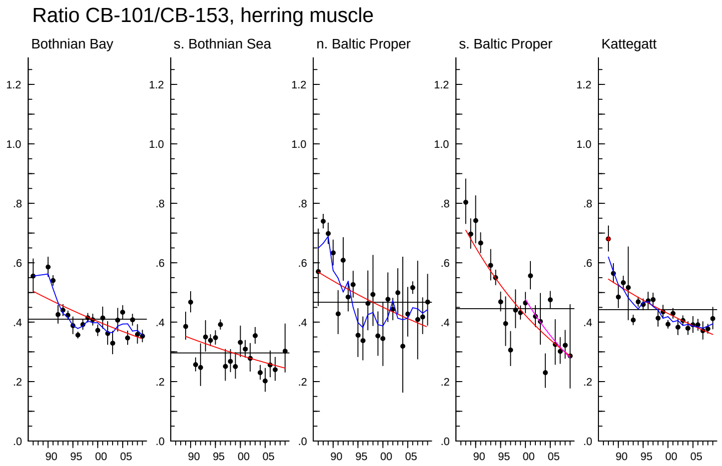

Figure 2. Log-linear trends of the CB-101/CB-153-ratio in herring muscle from the Bothnian Bay, southern Bothnian Sea, northern Baltic Proper, southern Baltic Proper and the Kattegatt (time series starting in 1987, 1989, 1987, 1988 and 1988 respectively). The red line shows a significant trend over the whole time period, the

magenta colored line a significant trend for the last ten years, the blue line a non linear trend and the dotted line the mean ratio over the whole period.

Spatial Patterns

The concentrations of PCBs, illustrated by the mono-ortho CB-118 in Fig. 3, are generally at the same level in marine and freshwater perch. The figure shows that higher concentrations are found in perch from a few of the lakes and in perch from one of the marine sampling sites within the study from 2009. The marine sampling site could be considered to be polluted by a local source since a large number of other contaminants also show elevated levels at that specific site compared to the marine reference sites. The concentration of PCBs in herring muscle is in general significantly higher in the Baltic than in Skagerrak and Kattegatt, but no such pattern with lower concentrations towards the western parts of Sweden is indicated for the freshwater sampling sites (perch).

Differences in congener pattern were seen between marine and freshwater perch (Fig. 4). The freshwater perch has in general higher relative concentrations of CB-180 than the marine perch. This may not only be explained by the source of the PCBs, but could also be related to differences in diet between the marine and freshwater perch. Clear differences in congener pattern were also seen between arctic char and pike from the northern parts of Sweden (Fig. 4). Pike has in general higher relative concentrations of CB-180 and arctic char relatively higher concentrations of CB-101. Perch from northern Sweden has the same higher relative concentration of CB-180 as pike. The dissimilarities between arctic char and pike might be explained by differences in diet, uptake of contaminants and metabolism.

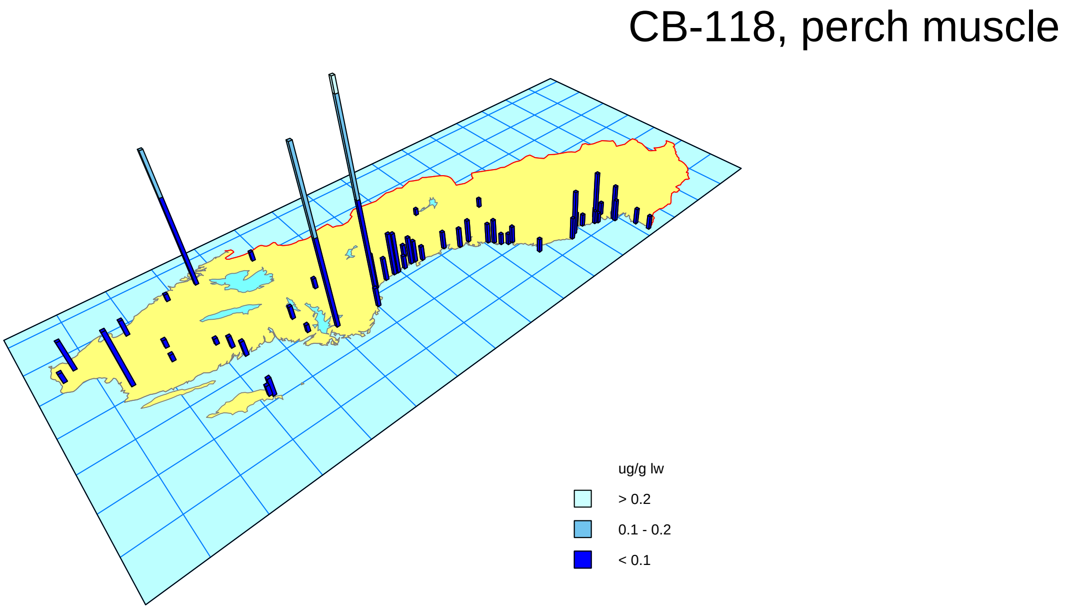

Figure 3. Spatial variation in concentration (ug/g lipid weight) of CB-118 in marine and freshwater perch muscle. Arithmetic mean values 2007-2009 for the freshwater and marine reference sites and values from 2009 for the other marine sites.

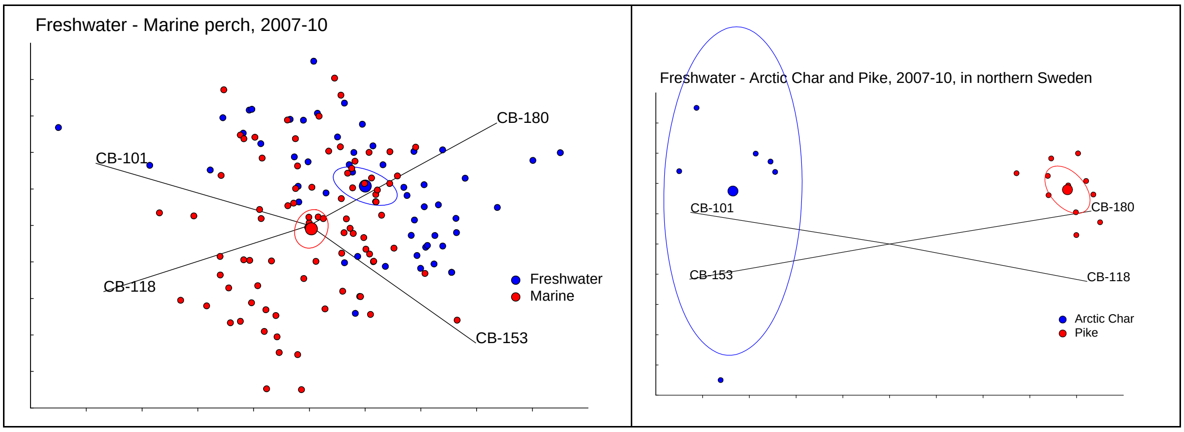

Figure 4. PCA analysis, biplot and Hotelling's 95% confidence ellipses for centre of gravity for the groups. The figure on the left show marine and freshwater perch and the figure on the right show arctic char and pike from the northern parts of Sweden.

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References:

1. Jensen S, Johnels A.G, Olsson M, Otterlind G. (1969); *Nature* 224: 247-50
2. Jensen S, Johnels A.G, Olsson M, Otterlind G. (1972); *Ambio Special report* No 1: 71-85
3. Bredhult C, Bäcklin B-M, Bignert A, Olovsson M. (2008); *Reproductive Toxicology* 25: 247–55
4. Helle E, Olsson M, Jensen S. (1976); *Ambio* 5: 188-9
5. Helle E, Olsson M, Jensen S. (1976); *Ambio* 5, 261-3.
6. OSPAR. JAMP Guidelines for Monitoring Contaminants in Biota. (1999); *OSPAR Agreement*: 1999-2, OSPAR Commission
7. EC. Common implementation strategy for the Water Framework Directive (2000/60/EC): Guidance on chemical monitoring of sediment and biota under the Water Framework Directive. (EC); *Technical report* 2010-041
8. Eriksson U, Häggberg L, Kärsrud A-S, Litzén K, Asplund L. (1997); Institute of Applied Environmental Research, Stockholm University, *ITM Report* no.59
9. Jensen S, Reutergårdh L, Jansson, B. (1983); *FAO Fisheries Technical Paper* No 212(9): 21-33
10. Braune B.M, Outridge P.M, Fisk A.T, Muir D.C, Helm P.A, Hobbs K, Hoekstra P.F, Kuzyk Z.A, Kwan M, Letcher R.J, Lockhart W.L, Norstrom R.J, Stern G.A, Stirling I. (2005); *Sci. Total Environ* 351-352, 4-56
11. Helgason L.B, Barrett R, Lie E, Polder A, Skaare J.U, Gabrielsen G.W. (2008); *Environ Pollut.* 155, 190-8
12. Ryan M.J, Stern G.A, Diamon M, Croft M.V, Roach P, Kidd K. (2005); *Sci. Tot. Environ.* 351-352, 501-22
13. Rigét F, Vorkamp K, Dietz R, Rastogi S.C. (2006); *J. Environ. Monit.* 8,1000-5