LARGE DIFFERENCES IN DIOXIN AND PCB LEVELS IN HERRING AND SALMON DEPENDING ON TISSUE ANALYSED

Marie Aune[1], Rickard Bjerselius[1], Samuel Atuma[1], Lotta Larsson[1], Arpi Bergh[1], Per Ola Darnerud[1], Arne Andersson[1], Fredrik Arrhenius[2], Sture Bergek[3], Mats Tysklind[3] and Anders Glynn[1]

1 Swedish National Food Administration, P.O. Box 622, SE-751 26 Uppsala, Sweden
2 Institute of Marine Research, National Board of Fisheries, P.O. Box 4, SE-453 21 Lysekil, Sweden
3 Environmental Chemistry, Umeå University, SE-901 87 Umeå, Sweden

Introduction

Most organochlorine compounds (OCCs) used in agriculture and industry have been banned for decades in Sweden and many other countries and the release of dioxins (PCDD/DFs) have decreased. However, levels are still found in the surrounding biota as a result of the environmental persistence and high lipophilicity of the compounds. The levels of most OCCs have declined in food since the 1970s but the decline of PCDD/DFs and some PCBs in fish from some areas of the Baltic Sea appear to have ceased in the 1990s[1]. The maximum level for PCDD/DFs in fish and fishery products, 4 pg WHO-PCDD/DF-TEQ/g set by the European Council, is expressed on fresh weight basis due to the extremely large difference in fat content between lean (e.g. cod 0.7 %) and fatty fish (e.g. eel 30 %)[2]. In Sweden, some herring products are normally consumed with the fish skin and therefore the Swedish National Food Administration analyse muscle tissue of herring including the skin for OCCs. However, many products are sold without the fish skin and it is therefore of interest to both food authorities and industry to analyse different parts of the herring. In addition, we present results from a study on salmon indicating large differences in sum PCB depending on which part of the fish is analysed.

Methods and Materials

From individual herrings (*Clupea harengus*) caught 1998 in the Gulf of Bothnia (n=16) and on the Swedish West Coast (n=14), one sample of only muscle tissue and one sample of muscle tissue and skin were prepared from each fish (Table 1). For the muscle tissue sample the skin and the subcutaneous lipids were removed. The samples were then individually (n=60) analysed for PCB congeners (CBs 28, 31, 52, 101, 105, 110, 118, 128, 138, 149, 153, 156, 158, 167, 170 and 180) and organochlorine pesticides (results not presented). Pooled samples of so called “Rügen herring” caught at two different locations south west of Bornholm in year 2002 (n= 30 and 36, respectively) were then prepared (Table 1). One of the fillets from each fish was skinned and the muscle tissue used for one sample. The corresponding fillet was then sampled with the fish skin and pooled. The samples were then thoroughly homogenised and analysed for PCB congeners (CBs 28, 31, 52, 66, 74, 77, 81, 101, 105, 110, 114, 118, 126, 128, 138, 149, 153, 156, 157, 158, 167, 169, 170 and 180) and PCDD/DFs.

Two salmons (*Salmo salar*) caught in the Bothnian Bay and Baltic Proper in 1996 (weight 5.8 and 5.3 kg, respectively) were sampled from three different parts of the fish, the anterior muscle,

middle muscle and tail muscle. The samples were then individually analysed for the PCB congeners 28, 31, 52, 101, 105, 110, 118, 128, 138, 149, 153, 156, 158, 167 and 180.

For the non-ortho PCBs and PCDD/DFs extraction, clean-up and analysis were done according to validated methods at Umeå university, Sweden. HRGC/HRMS and isotopically labelled standards were used for the quantification. The mono- and di-ortho PCBs were analysed by HRGC/ECD according to validated methods at the Swedish National Food Administration. The results for the PCDD/DFs and the dioxin-like PCBs are expressed in WHO-TEQ according to the WHO TEFs for human risk assessment[3]. In calculating the WHO-TEQ values the medium-bound level has been used for non-quantified levels. For the individual samples the sum of the congeners CBs 28, 52, 101, 105, 110, 118, 138, 149, 153, 170 and 180 was used since there was a high number of non-quantified values for the other PCB congeners analysed.

Results and Discussion

The results show a highly significant difference of levels of sum PCB between herring muscle tissues analysed with or without skin (one-way ANOVA, $P<0.0001$ followed by paired students t-test $P<0.0001$) (Fig. 1a). The same degree of significance is shown for the fat content between herring muscle tissues analysed with or without skin indicating that almost 100 % of the sum PCBs are stored in the fat (Fig. 1b). The results also indicate that this relationship is relatively stable, since the analysed fish samples varied in both sum PCB levels and fat content (Table 1.) without causing any large changes in calculated ratios (expressed in % change in Figs. 1a, b). The same patterns of distribution and relative change are seen for the dioxins and dioxin like PCBs (Fig. 2). The dramatic decrease of dioxin and dioxin like PCBs when the skin is removed might have significant impact on species allowed for intra trade and export. It also addresses the need to have more distinct regulations as well as a need for more analyses of final food products, especially fish.

Table 1. Raw data with mean (min-max) of the herring analysed with or without the skin. See material and methods or further details.

	Bothnian Bay	Swedish West Coast	Rügen, SW Bornholm
No of analysis (individuals)	16 (16)	14 (14)	2 (66)
Weight (g)	28.2 (22.7-35.3)	63.9 (57.4-72.2)	49.2 (28.0-76.1)
Fat content incl skin (%)	5.80 (1.01-11.0)	12.7 (6.65-18.4)	7.10 (6.96-7.23)
Fat content excl skin (%)	2.41 (0.81-4.33)	6.08 (3.87-8.62)	3.51 (3.51-3.52)
Sum PCB incl skin (μg/kg fw)[a]	21.1 (1.99-47.2)	13.2 (8.18-24.2)	22.3 (21.8-22.8)
Sum PCB excl skin (μg/kg fw)	7.52 (1.05-20.6)	5.92 (3.95-11.2)	10.1 (9.27-10.9)
PCB-TEQ incl skin (pg/g fw)[b]			2.00 (1.95-2.04)
PCB-TEQ excl skin (pg/g fw)			0.92 (0.88-0.97)
PCDD/DF-TEQ incl skin (pg/g fw)			1.71 (1.64-1.78)
PCDD/DF-TEQ excl skin (pg/g fw)			0.81 (0.76-0.85)

[a] The sum of the PCB congeners 28, 52, 101, 105, 110, 118, 138, 149, 153, 170 and 180.
[b] The sum TEQ for the dioxinlike PCB congeners analysed.

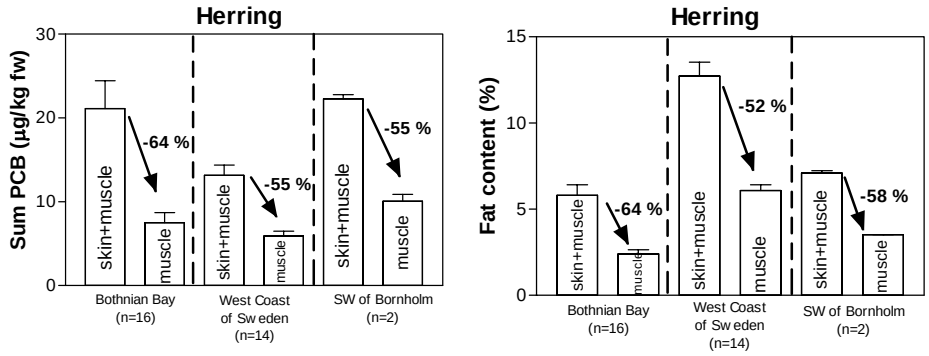

Figure 1a, b. Herring from three different locations were sampled as muscle tissue and muscle tissue with fish skin and then analysed for PCB congeners. The sum PCB mean concentrations (µg/kg fresh weight) and fat contents (%) for the different samples and locations are shown.Numbers at arrows represent the decrease in percent.

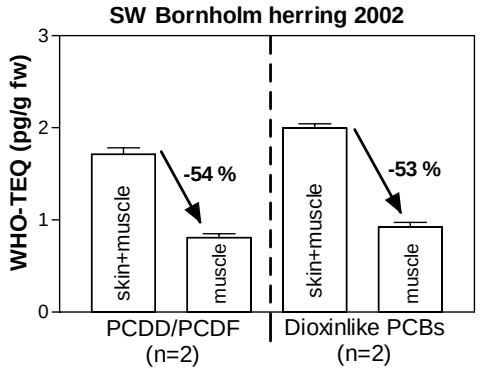

Figure 2. The difference in mean WHO-PCDD/DF-TEQ (pg/g fresh weight) and mean WHO-PCB-TEQ (pg/g fresh weight) is shown for two pooled samples of herring analysed as muscle tissue or muscle tissue including skin. The herrings were caught south west of Bornholm in 2002. Numbers at arrows represent the decrease in percent.

In addition, the preliminary results from the salmon study shows that there can be significant differences in reported levels of sum PCB depending on which part of the fish is analysed (Fig 3). In accordance with the sum PCB - fat content relationship described for herring, the fat contents for the anterior, middle and tail tissues are 22, 17 and 4 %, respectively explaining the large variation in the same fish. Indeed, a 4.5-fold difference is seen between the anterior muscle tissue compared with the tissue at the tail.

Again, this stresses the importance of specific and clear instructions for sample preparation to avoid misleading results and conclusions based on non-representative values.

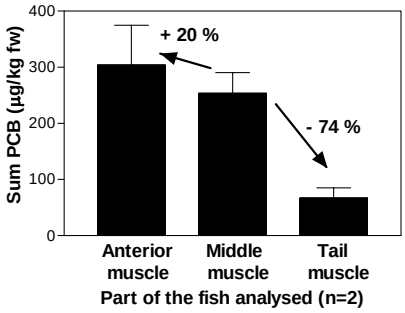

Figure 3. Different parts of two salmons from the Bothnian Bay and Baltic Proper were analysed for PCB congeners. The sum PCB mean levels (µg/kg fresh weight) for the analysed parts as well as the percentage differences between the parts are shown.

Acknowledgements
We thank Elvy Netzel and Ingalill Gadhasson at the NFA for technical assistance.

References

1. Bignert A. (2002) Comments concerning the National Swedish Contaminant Monitoring Programme in Marine Biota. www.nrm.se/mg/mcom
2. Council Regulation 2375/01/EC amending Commission Regulation (EC) No 466/2001
3. Van den Berg et al. (1998) Env. Health Persp. 106, 775