HALOGENATED CONTAMINANTS IN THE TOP 5 FARMED FISH CONSUMED IN THE NETHERLANDS

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Introduction

The world production of aquaculture has grown in the last decades. The consumption of farmed fish in the Netherlands is also increasing, on the one hand by well-known species such as salmon and shrimp and on the other hand by introduction of new species such as pangasius, tilapia, sole and cod [1]. Several reports in recent years have shown that farmed salmon, trout and shrimps can be contaminated with a range of contaminants including polychlorinated dibenzo-p-dioxins and –furans (PCDD/Fs), polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) [2-5]. On contaminants in fish, two major knowledge gaps can be identified: (1) only few reports exist on the contamination of farmed fish with brominated flame retardants (BFRs) and perfluorinated compounds (PFCs) and (2) virtually no information is available on contamination of tilapia and pangasius. This study aims at filling these gaps.

Materials and methods

This survey focussed on the Top-5 aquaculture species relevant for Dutch consumers. This includes salmon, trout, shrimp, tilapia and pangasius. Details on the consumption volumes [1], contaminants and number of samples can be found in Table 1. Dioxins were not investigated in salmon and trout because literature data [6] showed that the levels commonly observed are below the EU MRL [7].

Table 1 Sample matrix

				Number of samples					
Species	Latin name	Consumption (tons/yr)*	Origin	PFCs	PBDEs	HBCDs	PCDD/Fs and dl-PCBs	PCBs	OCPs
Salmon	Salmo salar	8700	Norway, Scotland, Chili	7	7	7	-	7	7
Pangasius	Pangasius hypophthalmus	1700	Vietnam	7	7	7	5	7	7
Tilapia	Oreochromis mossambicus, Oreochromis niloticus	1200	China, Ecuador, Indonesia, Netherlands	7	7	7	5	7	7
Trout	Oncorhynchus mykiss, Salmo trutta	900	Denmark, Italy, Turkey	5	5	5	-	5	5
Shrimps	Penaeus monoden, Penaeus vannamei, Litopenaeus vannamei	1500	Bangladesh, Mixed-Asia**, Netherlands	6	6	6	5	6	6
Cod	Gadus Morhua	na	Norway	1	1	1	1	1	1
	Total	14000		33	33	33	16	33	33

* Dutch consumption of farmed fish in 2006 (ton/yr), representing approx. 18% of the total fish and shellfish consumption [1]

** Mixed origins were declared on the package label (Bangladesh/India, Indonesia/China or Thailand/Malaysia/Indonesia)

Samples were bought between October 2007 and January 2008 from supermarkets, fish stores, week markets and suppliers for restaurants. Samples were stored at -20C in their original packaging prior to further treatment. Each sample consisted of approx. 10 individual fillets (fish) or 1-1.5 kg (shrimps). Prior to analysis, the individuals of each sample were pooled and homogenised.
Dioxins and dl-PCBs – The WHO-selection of 17 tetra- to octa-substituted PCDD/Fs and 12 non-ortho and mono-ortho substituted PCBs were analysed by GC-HRMS. *PCBs and OCPs* – The target PCBs in this study included PCB 28, 52, 101, 118, 138, 153 and 180 and the target OCPs were HCBD, QCB, HCB, α-HCH, β-HCH, γ-HCH, heptachlor, trans-heptachlor epoxide, cis-heptachlor epoxide, aldrin, telodrin, isodrin, dieldrin, endrin, α-endosulfan, o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD, o,p'-DDT and p,p'-DDT. The samples were analysed by dual column GC-ECD. *BFRs* – Twenty-six BDEs (BDE 7, 15, 17, 28, 49, 71, 47, 66, 77, 100, 119, 99, 85, 126, 154+BB153, 153, 138, 156, 184, 183, 191, 197, 196, 208, 206, 209) were analysed by GC-ECNI-MS in all samples. Furthermore, HBCD (α-, β- and γ-diastereomers) were analysed by LC-ESI-MS/MS. The principles of the methods used can be found elsewhere (PCDD/Fs: [6]; PCBs and OCPs: [8]; PBDEs [9], HBCDs [10]). *PFCs* – The targeted compounds were perfluoalkylsulfonates (PFBS, PFHxS, PFOS, PFDS), perfluoroalkylcarboxylates (PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFTrA and PFTeA and perfluorooctanesulfonamide (PFOSA). Extraction and clean-up were performed similarly to the method first published by Powley et al. [11, 12], with an additional sample drying step prior to extraction. The extracts were analysed by LC-ESI-MS/MS. The quality of all analysis was assured routinely by analysis of procedural blanks, duplicate samples, internal reference materials, use of (mass labelled) internal standards and the participation in various interlaboratory studies with satisfactory results.

Results and discussion
Dioxins and dl-PCBs – The levels detected in pangasius, tilapia and shrimps were extremely low (lowerbound: <1 to 82 fg total-TEQ/g ww), and were much lower than those observed in an earlier study on Dutch wild caught marine and freshwater fish [6]. On upperbound basis, the levels were in the range of 0.2-0.3 pg total-TEQ/g ww, and were well below the EU-MRL [7].
PCBs and OCPs – Data on PCBs and a selection of OCPs is shown in Figure 1. The PCB levels in all fish met the MRLs for the 7 indicator PCBs as laid down in the Dutch 'warenwet" [13]. For OCPs, no MRLs are available, but generally, the sum-DDT levels were in the same range as the sum 7 PCB levels. Dieldrin and HCB were lower. The levels in the carnivorous fish (salmon, trout) were higher than those of the herbivorous fish and shellfish. Compared to the observations by Hites et al. [4], the levels in salmon are at the lower end of the ranges he observed. This suggest that the aquaculture industry nowadays uses lower contaminated feeds and possibly the salmon diet consist of a lower proportion of fish meal and fish oils.
BFRs – Data on a selection of BDEs is shown in Figure 1. The sum-BDE levels are approx. 10 times lower than the sum 7 PCB and the sum DDT levels in the same samples. Also for the BDEs, the levels are at the low end of the ranges reported by Hites et al. [3]. As compared to wild caught herring, the levels were much lower in aquaculture fish. Levels of HBCDs were detected in 16 samples, ranging from 6 pg/g to 1200 pg/g ww. In all cases, a-HBCD was the predominant diastereomer. In the other samples, the levels were <LOD.
PFCs – The levels of PFCs were also low. In most of the samples, PFCs were not detected at all.
Implications for human exposure - The Dutch consumption of (aquacultured) fish is increasing [1]. Within this group, the share of pangasius is growing rapidly. Considering the low contaminant levels observed in pangasius and tilapia, it is believed that the exposure from fish (as a whole) to the overall human exposure to PCBs, OCPs, BFRs and dioxins and dl-PCBs will decrease. Within fish as a group, wild caught fish (e.g. herring and mackerel) will presumably become more predominant contributors to the human exposure.
Herring has predominantly contributed to the fish-based human exposure to dioxins and dl-PCBs and BFRs.

Conclusions
This extensive study shows that contaminant levels in the Dutch consumption top five farmed fish (salmon, trout, tilapia, pangasius and shrimps) are very low (mostly < 1 ng/g ww), and far below the applicable MRLs. The contaminant levels decrease in the following order PCBs ≈ OCPs > BFRs > PCDD/Fs and dl-PCBs. It is likely that the Dutch consumer exposure to contaminants from (farmed) fish will change due to the low levels observed in farmed fish in this study as compared to e.g. wild caught herring.

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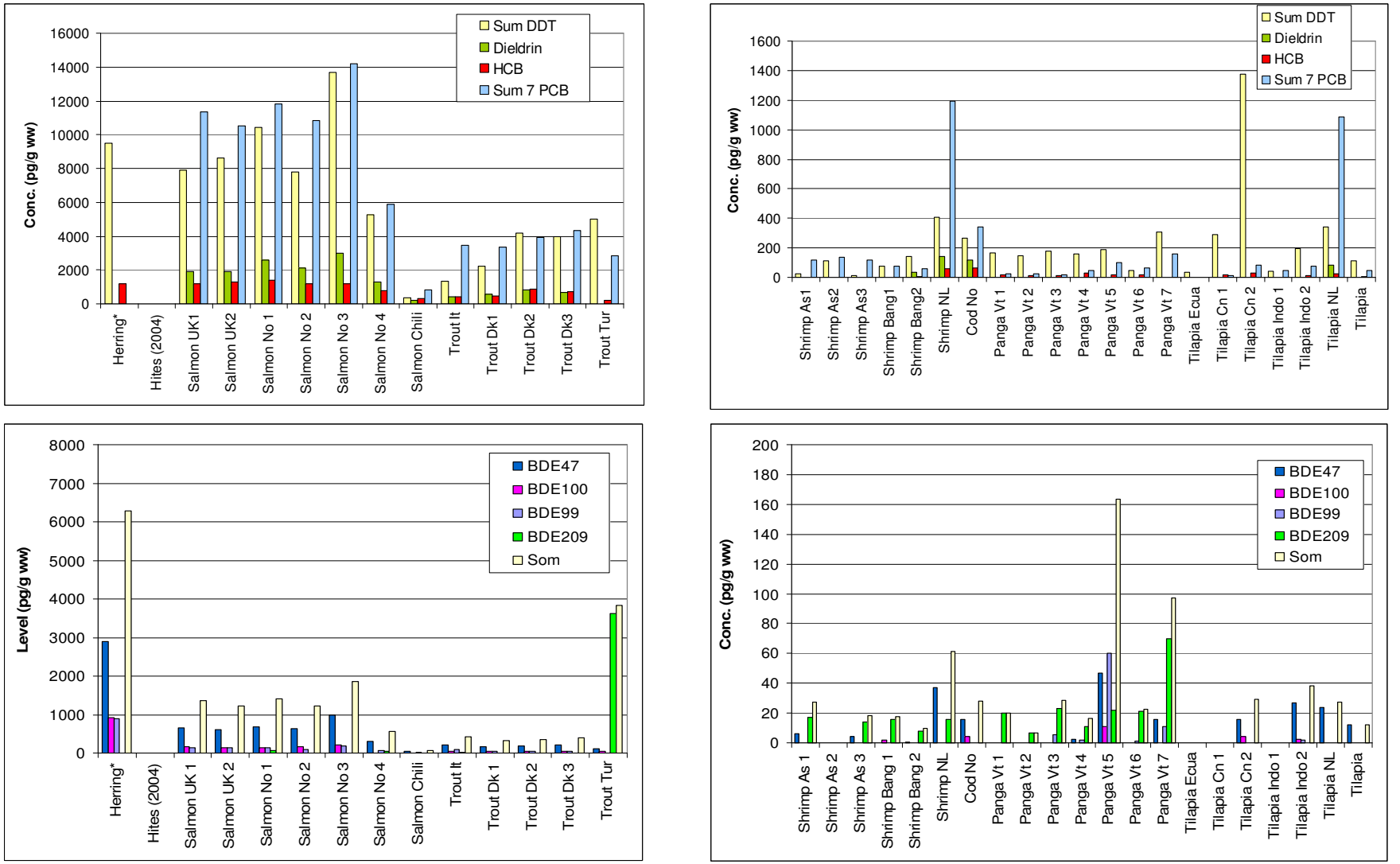

Figure 1. Contaminant concentrations in Top 5 consumed aquaculture species in the Netherlands. Top: PCBs and OCPs; bottom: PBDEs. In the left graphs, data from two other studies are shown for comparison reasons (wild caught herring (*) from North-west Europe, 2003 and salmon samples from the Hites et al. studies [3, 4]. Country codes (see also Table 1): UK = Scotland, No = Norway, It = Italy, Dk = Denmark, Tur = Turkey, As = Asia mixed origins, Bang = Bangladesh, NL = Netherlands, Vt = Vietnam, Ecua = Ecuador, Cn = China and Indo = Indonesia.