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ScienceDirect

Chemosphere 71 (2008) 1422–1431

CHEMOSPHERE

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Polybrominated diphenyl ethers (PBDEs) in farmed and wild salmon marketed in the Northeastern United States

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Received 11 July 2007; received in revised form 15 January 2008; accepted 16 January 2008
Available online 7 March 2008

Abstract

Recently, we reported on the analysis of polychlorinated biphenyls (PCBs) and chlorinated pesticides in farmed Atlantic salmon (*Salmo salar*) from Maine, eastern Canada, and Norway, and wild Alaskan Chinook salmon (*Oncorhynchus tshawytscha*). In this paper, we extend the analysis to polybrominated diphenyl ethers (PBDEs) in these samples. Total PBDE concentrations in the farmed salmon (0.4–1.4 ng/g, wet weight, ww) were not significantly different from those in the wild Alaskan Chinook samples (0.4–1.2 ng/g, ww), nor were significant differences found among regions. However, significant intra-regional variations in concentrations of total PBDEs and tetra-BDE 47 were observed in the salmon from the Canadian farms ($p < 0.01$). Congener profiles were dominated by BDE-47, followed by the penta-BDEs 99 and 100. PBDE concentrations in the Canadian samples were lower than those reported two years earlier. Removal of skin resulted in no overall reduction in PBDE concentrations in our farmed salmon, and in some cases, PBDE concentrations were higher in skin-off samples. PBDEs were correlated with lipids only in the skinned samples, suggesting that there is greater accumulation and retention of PBDEs in muscle lipids than in skin-associated fat. In skin-on samples, modest correlations were observed between concentrations of PBDEs and PCBs ($R^2 = 0.47$) and mono-*ortho* PCBs ($R^2 = 0.50$), whereas PBDEs were not correlated with non-*ortho* PCBs.

Keywords: Fish consumption; Brominated flame retardants; Human health risks; Persistent organic pollutants; Aquaculture

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are persistent and bioaccumulative flame retardant chemicals that are ubiquitous global contaminants. In the United States, human levels in blood, milk, and tissues are 10–20 times higher than in Europe or Asia and are increasing with a doubling time of ~5 years (Birnbaum and Staskal, 2004; Hites, 2004; Sjodin et al., 2004; Johnson-Restrepo et al., 2005a; Schecter et al., 2005a; Webster et al., 2005). These rising levels are of concern because, like the polychlorinated biphenyls (PCBs), PBDEs have been shown to cause reproductive toxicity, endocrine disruption, and neurodevelopmental effects in animals (Birnbaum and Staskal, 2004). Unlike PCBs and dioxins, PBDEs are indoor pollutants found in a variety of household products including textiles, furniture, clothing, electronics, and plastics (Schecter et al., 2005b; Stapleton et al., 2005). Of three commercial PBDE products, the penta-BDE and octa-BDE mixtures are no longer in use in the US, whereas the deca-BDE product is still manufactured and sold worldwide. While sources of human exposure to PBDEs can be diverse, food is recognized as an important exposure pathway. PBDE concentrations in US foods are reportedly

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0045-6535/$ - see front matter
doi:10.1016/j.chemosphere.2008.01.030

higher than levels in food from other countries, and the highest concentrations are found in fatty fish such as salmon as compared with other farm-raised animals (chicken, beef, pork) destined for human consumption (Schecter et al., 2006a). In addition to PBDEs, farmed salmon can be contaminated with complex mixtures of other persistent organic pollutants (POPs) including PCBs, polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs), and chlorinated pesticides due to dietary input from fish oil and meals in the commercial feed (WHO, 1999; Easton et al., 2002; Jacobs et al., 2002; Hites et al., 2004a; Carlson and Hites, 2005; Shaw et al., 2006).

Salmon consumption in the United States has increased by more than 26% annually since 1987; at present, more than half of the salmon consumed globally is farmed (FIGIS, 2004). An estimated 23.1 million US residents eat salmon (primarily Atlantic salmon, *Salmo salar*) more often than once a month, 1.3 million eat salmon at least once a week, and 180 000 eat salmon more often than twice a week (EWG, 2003). Many people eat salmon regularly because of the health and nutritional benefits associated with the intake of omega-3 fatty acids present in salmon. However, results of recent quantitative benefit–risk analyses for farmed and wild salmon (Foran et al., 2005a,b) suggest that these benefits, primarily the possible prevention of sudden cardiac death, may be outweighed by health risks of consuming contaminated farmed salmon, particularly for pregnant women and young children.

Industry efforts are under development to reduce contaminant levels in salmon feed by substituting vegetable oils for contaminated fish oils in commercial feed (Bell et al., 2005; Bethune et al., 2006). Since lipophilic contaminants are stored in fat, removal of skin and subcutaneous fat from the edible portion of the flesh and some cooking procedures have also been proposed as a means to reduce contaminant exposure, although studies to date (Skea et al., 1979; Hora, 1981; Voiland et al., 1991; Zabik et al., 1995; Schecter et al., 2006b) have generated inconsistent results.

In a previous study, we reported on concentrations of PCBs, dioxin-like PCBs, and chlorinated pesticides in salmon marketed to consumers in the northeastern United States, including farmed Atlantic salmon from Maine, eastern Canada, and Norway and wild-caught Chinook salmon (*Oncorhynchus tshawytscha*) from the Aleutian Islands, Alaska (Shaw et al., 2006). Here we provide results on the analysis of PBDEs in these samples. Whereas other studies have compared contaminant loads in farmed salmon by region, this study examined intra-regional differences in salmon from individual farms. In addition, we analyzed skin-on and skin-off fillets to determine to what extent the removal of skin contributes to reductions in PBDE concentrations. It has been proposed that lipid content in muscle tissue of salmonids is a major determinant of the distribution of lipophilic PCBs (Persson et al., 2007); however, the mechanisms of PBDE accumulation in salmon lipids are not well understood. Thus, we examined relationships between PBDEs and muscle lipids as well as correlations between PBDEs and PCBs in salmon fillets.

2. Materials and methods

2.1. Samples

Details of the sample collection and processing have been previously described (Shaw et al., 2006). Briefly, a total of 70 farmed and wild salmon were collected from wholesale and retail outlets in Maine between August 2003 and May 2004. Suppliers provided information on the origin (region and farm) of the fish. Wild-caught Chinook salmon from the Aleutian Islands, Alaska (AK), were purchased from a wholesale supplier. The farmed salmon represented six locations in three regions, including two farms in eastern Maine (ME1 and 2), three in eastern Canada (CAN1, 2, and 3), and a farm in Norway (NOR).

Of 10 whole salmon obtained from each farm, nine were randomly selected for analysis. Length and weight metrics are shown in Table 1. All farmed salmon were 2.5–3-years old; the ages of the wild salmon were unknown. The whole fish were thawed, weighed, measured, and filleted to yield two boneless fillets per fish. One fillet from each fish was left intact, and from the other, we removed the skin and associated fat, belly flap, and lateral line. Preparation methods as previously described (Shaw et al., 2006) followed procedures used by retail suppliers to ensure that samples were replicas of those sold to consumers.

The farmed Norwegian salmon samples analyzed in this study were purchased in 2004 from a retail distributor in the northeastern US who provided documentation of the protocol under which the salmon were raised. The documents indicated that the salmon were organically-farmed, free of antibiotics and artificial color, raised on 100% organic feed certified by the Soil Association of the UK, virtually free of dioxins, and containing PCBs and trace elements dramatically lower than ordinary farm-raised salmon. The feed was reported to contain less than 28% marine oils. It was later discovered that non-organic salmon were being exported to the US from Norway and marketed as organic around the same time that we purchased these Norwegian fish (Shaw et al., 2007). Since it is likely that

Table 1
Biometrics of salmon sampled from each region and producer

Producer	Type	*N*	Age (years)	Mean ± SD		
				Weight (kg)	Length (cm)	Lipid (%)
Norway	F	3	2.5–3	4.1 ± 0.05	76 ± 1.0	18 ± 1.6
Maine 1	F	3	2.5–3	4.8 ± 0.14	77 ± 1.8	18 ± 0.71
Maine 2	F	3	2.5–3	1.2 ± 0.13	56 ± 1.0	7.7 ± 2.0
Canada 1	F	3	2.5–3	3.8 ± 0.28	80 ± 0.25	13 ± 1.0
Canada 2	F	3	2.5–3	4.0 ± 0.63	80 ± 1.8	14 ± 1.6
Canada 3	F	3	2.5–3	4.6 ± 0.22	81 ± 0.84	14 ± 1.2
Alaska	W	3	Unk	5.2 ± 2.1	80 ± 3.8	7.6 ± 2.2

F, farmed; W, wild; Unk, unknown.