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Investigation of Selected Persistent Organic Pollutants in Farmed Atlantic Salmon (*Salmo salar*), Salmon Aquaculture Feed, and Fish Oil Components of the Feed

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There is extensive literature documenting the bioaccumulation of persistent organic pollutants in the marine environment, but relatively little data are available on contamination pathways in aquaculture systems such as that for farmed salmon. In recent years, the salmon industry has grown significantly in Europe. This study reports on the determination of a wide range of polychlorinated biphenyls (PCBs), organochlorine pesticides, and polybrominated diphenyl ethers (PBDEs) in farmed and wild European Atlantic salmon fish, aquaculture feeds, and fish oils used to supplement the feeds. The study confirms previous reports of relatively high concentrations of PCBs and indicates moderate concentrations of organochlorine pesticides and PBDEs in farmed Scottish and European salmon. Concentrations of the selected persistent organic pollutants varied among the samples: PCBs (salmon, 145–460 ng/g lipid; salmon feeds, 76–1153 ng/g lipid; fish oils, 9–253 ng/g lipid), S DDTs (salmon, 5–250 ng/g lipid; salmon feeds, 34–52 ng/g lipid; fish oils, 11–218 ng/g lipid), and PBDEs (salmon, 1–85 ng/g lipid; salmon feeds, 8–24 ng/g lipid; fish oils, ND–13 ng/g lipid). Comparison of the samples for all groups of contaminants, except for HCHs, showed an increase in concentration in the order fish oil < feed < salmon. Homologue profiles were similar, with an increase in contribution of hepta- and octa-PCBs in the fish, and profiles of DDTs were similar in all three types of samples. With a constant contribution to the total PCB content, the ICES 7 PCBs appear to be reliable predictors of the PCB contamination profile through all the samples. For PBDEs, BDE 47 dominated the profiles, with no significant difference in the PBDE profiles for the three matrixes. Samples with higher PCB contents generally showed higher levels of the pesticide residues, but this was not the case with the PBDEs, indicating the existence of different pollution sources.

Introduction

Although there is an extensive literature on the presence and fate of chlorinated organic compounds in the aquatic

environment and biota, most data relate to the natural environment, and little has been published on halogenated compounds in aquaculture systems. Data are available on the concentrations of polychlorinated biphenyls (PCBs) and other organochlorine contaminants such as polychlorinated dibenzo-*p*-dioxins (PCDDs) and furans (PCDFs) in fish from wild stocks, where exposure is associated with chronic contamination due to leaching of these chemicals from treated or contaminated soils into surface waters and the global distribution and deposition by atmospheric transport (1). With farmed fish, potential hazards associated with the ingredients and additives used in aquaculture feed were not considered significant until very recently (2–5).

Fish oil is a byproduct of the fish meal manufacturing industry and comes from many different parts of the world. As chlorinated compounds accumulate in the lipid compartment of the animal, oil extracted from fish caught in polluted waters may be contaminated with chlorinated hydrocarbons (6, 7). Existing data on the levels of PCBs, PCDDs, PCDFs, and organochlorine pesticides consumed in the United Kingdom have been mainly derived from total diet surveys and surveillance data for specific food types (8, 9). These data indicate that the presence of elevated organochlorine contamination of farmed fish, salmon, and trout samples tested in 1997 were found to contain residues of PCBs at concentrations between 23 and 620 ng/g lipid weight (10, 11). DDT and HCB were the predominant organochlorine pesticides detected in these samples.

The widely used flame retardants polybrominated diphenyl ethers (PBDEs) are also persistent and ubiquitous organic pollutants (POPs) that biomagnify and may have endocrine-disrupting effects (12–15). Concern with the risks to human health, particularly infants, is increasing due to observations of increasing PBDE concentrations in human breast milk (13, 16), although available data, including that reported here, suggest that current levels of PBDEs are an order of magnitude lower than those of PCBs. While PBDEs are in extensive production and use, existing data on the levels of PBDEs consumed in the United Kingdom are very limited, although surveys to determine the distribution and fate of PBDEs in marine life in waters around the United Kingdom are underway (17). However, there is little or no data in the public domain for aquaculture products, and data for farmed fish is very limited (18).

The determination of these contaminants in fish and identification of the original sources of the contamination is, therefore, important for dietary exposure assessment and the protection of public health, particularly in view of the increasing availability to the consumer of farmed salmon. The farmed-salmon industry is rapidly expanding; it has trebled in production from 1990 to 1998 (19). The three major producers are Norway, the United Kingdom, and Chile, although other countries, notably the United States, are also increasing the growth rate of their salmon farming industries. While the use of fish meal in aquaculture feeds for farmed fish and shrimp continues to increase (20), it should be noted that twice as much fish meal is used in poultry and pig feeds (19, 20). However, the feed costs to produce farmed salmon appear to be significantly greater on a live weight basis (21). The proportion of fish meal supplies used for farming fish rose from 10% in 1988 to 33% in 1997 (20, 22). The use of fish meal and fish oil of European origin are considered to be of critical concern when used in diets for farmed fish and other food-producing animals (4, 5). The aquaculture sectors most dependent on the use of fish oil (commonly herring oil) as a source of dietary lipids are the salmon and marine finfish

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TABLE 1. Sample Details for the Salmon, Feed, and Fish Oil^a

sample codes	sample type and collection	information	% lipid	sample weight (g) ^b	lipid weight (g)
M11	salmon: Scotland, Jan 1999	(f) 3+ years fresh	13.5	8.19	1.10
M12		(f) 2+ years fresh	14	8.13	1.14
M13		Norway frozen	13.7	8.39	1.15
M14		(w) 2+ years frozen	4.9	9.73	0.47
M24		(f) 3+ years fresh	19.9	9.69	1.93
M25		(f) 3+ years fresh	19.1	9.04	1.73
M28		(w)	12.3	8.19	1.17
M31		(f) <i>N</i> = 2, Smolt, fresh	9.3	8.13	0.85
M18	salmon: Belgian Market, March 2001	(f) sm, bio	3.9	4.60	0.18
M19		(f) fresh, bio	16.5	5.15	0.84
M20		(f) fresh	11.0	5.48	0.60
M21		(w) Ireland, sm	11.6	4.51	0.52
M22		(f) Ireland, sm	14.8	7.25	1.07
M01	S. feed, Jan 1999	fry 1–5 g (A)	18.1	3.00	0.54
M02		1000–2200 g (B)	30.1	3.00	0.90
M03		350–1000 g (B)	31.0	3.03	0.94
M04		1000–2200 g (C)	35.9	3.07	1.10
M05		fry (C)	19.8	2.26	0.45
M15		500–1300 g (D)	28.1	2.00	0.56
M16		1300–2200 g (D)	34.3	2.11	0.72
M17		500–1300 g (D)	32.7	1.97	0.64
M06	fish oils, Jan 1999	fish oil (C)	100	0.5	
M07		fish oil (C)	100	0.5	
M08		fish oil (C)	100	0.5	
M09		fish oil (C)	100	0.5	
M10		vegetable oil (C)	100	0.5	
MDEPA		fish oil (E), March 2001	100	0.5	

^a (w) = wild; (f) = farmed; sources A, B, C, D, and E denoted in brackets; S. feed = Scotland aquaculture salmon feed; sm = smoked; bio = bioculture. ^b Sample weight taken for analyses; ND = nondetects (treated as $1/2$ LOD for Σ calculations).

industries. Fish oil is now being strongly promoted by the aquaculture industry as an aquaculture additive in a “high energy diet” that encourages fast growth of fish while minimizing protein utilization as an energy source and improves the nutritional value to humans through raised omega-3 fatty acid levels. Its use has increased significantly since 1995 from less than 30% to 33% and even 36.3% of the total diet for salmon in recent years (23–26).

To our knowledge, this paper presents the first assessment of levels of organohalogenated compounds in the farmed-salmon food chain. Farmed European salmon, feed, and fish oil samples were analyzed for a wide range of PCBs, organochlorine pesticides, and PBDEs. The results of analyses for eight of the salmon samples for PCDDs and PCDFs and selected coplanar PCBs have already been presented (27, 28). The current paper builds on this initial study, including data on a further five salmon samples, eight aquaculture feed samples, and five fish oils. These values are compared with those for fish and feed data reported in the literature, and the potential contribution to the bioaccumulation of PCBs, organochlorine pesticides, and PBDEs in farmed salmon is discussed.

Materials and Methods

Sites and Sampling. Seven British salmon samples (of which six samples were from individual fish and one sample was a composite of two fish from the same source and of the same age and size to ensure similar sample quantities of tissue) and one Norwegian salmon (*Salmo salar*) sample that enters the European fish market were analyzed. The samples were of variable age, both farm-raised and wild, and were obtained from five different Scottish sites, as documented previously (27, 28). Whole body weights of the fish ranged from 400 g to 5.2 kg. In addition, five salmon samples, two originating from Ireland (one wild, one farmed) and three farmed samples for which no further information was

available, purchased from the Belgian market, were analyzed. The fresh and frozen samples were wrapped in polyethylene bags and frozen immediately at -20°C . Eight salmon feeds were analyzed (from four different Scottish sources), and five fish oils and one vegetable oil were analyzed (all but one of which were obtained from the same source, on the same date, but originally from widely varying sources). The fish oil and feed were not samples fed directly to the salmon collected, as these were not available, but were collected from Scottish sites in January 1999. Table 1 gives complete sample details.

Sample Preparation. The samples were thawed, filleted, skinned, and the epaxial muscle homogenized before being subdivided into smaller replicate portions of approximately 100 g. The portions were weighed, stored in tightly sealed polythene bags, and frozen at -20°C . The samples were sent packed in dry ice to the Toxicological Center, Antwerp, where they were logged-in and stored at -20°C prior to analysis.

All samples were analyzed for HCH isomers, HCB, DDT, and metabolites (six o,p and p,p isomers) (Table 2) and for nine PBDE congeners (IUPAC nos. 28, 47, 66, 71, 75, 99, 100, 153, and 154). The samples obtained from Scotland were analyzed for 59 PCB congeners (see Table 3), including mono-ortho PCBs with an assigned toxic equivalency value (TEF) (29). All dioxin-like PCB levels compared favorably with those found in duplicate salmon samples earlier by another laboratory (27, 28). The samples obtained in Belgium were analyzed for 23 PCB congeners. The procedures used to analyze the salmon samples were similar to those used to analyze organochlorine compounds in chicken and pork fat, as described elsewhere (30, 31). The methods used to analyze PBDEs are described in Covaci et al. (32). PCB 46, PCB 143, ϵ -HCH, and ^{13}C -BDE 47, 99, and 153 were used as internal standards, while 1,2,3,4-tetrachloronaphthalene (TCN) and bromobiphenyl (PBB 80) were used as recovery standards. All individual standards of PCBs and pesticides were obtained

TABLE 2. Summary of PCBs and Organochlorine Pesticides in European (Scottish, Irish, and Norwegian) Atlantic Salmon, Aquaculture Feed, and Fish Oil: Lipid Adjusted (ng/g, ppb)

sample codes	PCBs								HCB	HCHs				DDTs				
	Σ tri	Σ tetra	Σ penta	Σ hexa	Σ hepta	Σ octa	Σ PCBs ^a	Σseven markers ^b		α-HCH	β-HCH	γ-HCH	Σ HCHs	p,p-DDE	p,p-DDT	p,p-DDD	Σ DDTs	p,p-DDT/ΣDDT
Salmon																		
M11	13.5	50.3	113.4	151.8	42.5	3.8	375.5	162.8	19.5	6.8	ND ^e	16.1	23.0	71.4	10.5	22.7	104.7	0.10
M12	33.2	46.2	79.6	110.2	29.4	2.0	300.5	119.5	13.1	4.7	ND	10.3	15.1	50.1	8.9	18.4	77.5	0.11
M13	1.5	1.5	25.2	75.1	38.3	3.8	145.1	69.9	ND	1.1	ND	9.7	10.9	2.8	1.3	1.0	5.2	0.25
M14	20.6	76.1	145	167.2	47.3	3.6	459.9	183.0	43.5	4.0	2.0	12.4	18.5	123.0	40.1	46.0	249.5	0.16
M24	10.2	34.2	80.4	112.6	37.1	4.6	279.1	118.4	15.6	9.7	ND	ND	9.8	29.0	9.1	8.7	46.9	0.21
M25	10.2	34.5	79.5	111.3	37.0	4.6	277.1	116.6	15.7	10.8	ND	ND	10.9	29.6	10.3	8.5	48.5	0.26
M28	9.2	29.8	77.6	131.5	47.3	4.8	300.0	125.9	13.8	ND	ND	ND	ND	30.0	12.0	3.3	45.4	0.29
M31	19.5	33.1	71.4	105.2	29.7	2.5	261.4	100.4	12.6	ND	8.7	14.1	22.9	33.1	14.0	9.3	56.5	0.25
M18	7.2	8.3	71.5	112.1	26.9	0.6	226.6 ^c	140.4	ND	ND	ND	ND	ND	37.1	9.2	ND	46.4	0.20
M19	7.5	18.6	65.1	130.7	40.3	2.9	265.1 ^c	167.9	ND	ND	ND	ND	ND	42.2	19.1	ND	61.4	0.31
M20	4.4	21.4	63.2	103.1	26.5	1.2	219.8 ^c	141.0	ND	ND	ND	ND	ND	46.1	18.5	ND	64.7	0.29
M21	2.8	8.6	29.0	40.6	12.0	1.0	94.0 ^c	60.6	4.1	ND	ND	ND	ND	38.8	11.5	ND	50.5	0.23
M22	5.5	16.9	79.9	134.6	43.8	3.7	284.4 ^c	176.6	12.0	ND	ND	ND	ND	39.2	9.2	ND	48.5	0.19
Feed																		
M01	14.6	15.8	29.2	53.7	9.8	0.6	123.8	49.3	2.3	4.2	ND	11.2	15.5	22.1	7.6	5.9	35.7	0.21
M02	688.4	306.3	84.4	43.4	10.1	0.2	1153.2	258.7	4.2	31.0	ND	15.7	46.8	25.6	4.6	7.8	38.1	0.12
M03	412.6	210.4	65.4	52.8	8.0	0.1	749.4	168.6	4.9	15.0	ND	12.5	27.6	23.0	4.3	7.7	35.1	0.12
M04	3.3	18.1	63.5	81.0	14.4	0.9	181.3	68.5	5.9	4.6	1.8	36.6	43.1	24.4	4.7	9.6	38.8	0.12
M05	2.0	5.9	23.0	40.9	3.7	ND	75.6	31.7	0.6	2.3	ND	ND	2.4	21.4	5.4	6.9	33.8	0.16
M15	1.6	26.8	50.6	72.6	11.0	0.6	163.1	65.5	7.6	5.6	ND	15.7	21.4	33.5	6.0	12.2	51.7	0.12
M16	3.8	27.8	57.4	79.4	14.7	0.8	181.0	74.7	9.3	5.2	ND	9.5	14.8	35.6	2.8	12.8	51.3	0.05
M17	3.9	22.9	49.8	68.8	10.6	0.4	157.3	63.7	7.1	5.9	ND	12.7	18.7	30.4	6.0	11.4	47.9	0.13
Fish Oil																		
M06	1.4	11.5	24.3	30.9	3.7	ND	74.0	27.7	3.7	6.7	ND	5.1	11.9	19.3	3.4	7.2	30.0	0.11
M07	0.3	ND	1.5	1.3	ND	ND	8.8	2.2	1.5	1.3	ND	1.4	2.8	7.8	3.8	3.7	15.4	0.25
M08	0.2	15.2	51.5	132.6	46.3	4.4	252.5	101.8	0.2	4.7	ND	15.7	20.5	36.1	5.5	12.3	54.0	0.10
M09	0.5	ND	2.9	9.2	1.6	0.0	18.6	7.9	0.2	2.0	2.0	1.5	5.6	7.4	1.6	1.8	10.9	0.15
M10	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.1	ND	14.5	15.7	ND	ND	ND	ND	ND
MDEPA	0.6	31.5	130.4	225.6	57.5	4.7	450.3 ^c	220.6	2.3	4.2	0.9	4.4	9.6	152.4	21.4	44.4	218.3	0.10

^a 59 PCB congeners. ^b ICES seven marker PCBs: IUPAC no's: 28, 52, 101, 118, 138, 153, 180. ^c 23 PCBs: 28 + 31, 52, 74, 101, 99, 110, 118, 105, 149, 153, 138 + 163, 128 + 167, 156, 187, 183, 180, 199, 170 + 190, 194. ^d ND = not detected ($1/2$ LOD).

from Dr. Ehrenstorfer Laboratories (Augsburg, Germany). PBDE native and labeled standards were available from Wellington Laboratories (Guelph, Canada). Acetone, hexane, dichloromethane, and isooctane were of pesticide-grade (Merck, Germany). Anhydrous sodium sulfate, basic aluminum oxide, and silica gel (Merck) were used after heating overnight at 120 °C. An accelerated Soxhlet extractor B-811 (Buchi, Switzerland) was used for the extraction of target compounds from fish tissues and feed.

Fish Tissue. The fish fillet was defrosted at room temperature, and approximately 10 g was precisely weighed and ground with 20 g anhydrous sodium sulfate and placed into an extraction thimble. Internal standards (20 ng of PCB 46 and PCB 143, 10 ng of ϵ -HCH, and 1.5 ng of a mixture of ^{13}C -BDE 47, 99, and 153) were added, and the mixture was extracted for 2 h with 75 mL of hexane/methylene chloride/acetone = 3:1:1 (v/v) into a hot Soxhlet manifold. After concentration and fat content determination, the extract was subjected to cleanup on two successive solid-phase extraction (SPE) cartridges containing 5 g of acid silica and acid silica (5 g)/neutral silica (1 g)/deactivated basic alumina (2 g) (from top to bottom), respectively. Fifty milliliters of hexane was used for the complete elution of PCBs, pesticides, and PBDEs in the same fraction. The final eluate was concentrated under nitrogen, and recovery standards were added prior to GC analysis.

Fish Oil. A total of 0.5 g of fish oil was precisely weighed and solubilized in 3 mL of hexane. Internal standards were added and the mixture equilibrated in an ultrasonic bath for 5 min, after which it was ready for application to the SPE cartridges.

Fish Feed. Approximately 3 g of fish feed were weighed accurately and placed into an extraction thimble. Internal

standards were added, and the feed was extracted for 2 h with 60 mL of hexane/methylene chloride/acetone = 3:1:1 (v/v) into a hot Soxhlet manifold. After concentration and fat content determination, the extract was ready to be applied to the SPE cartridges.

Fat Content Determination. The Soxhlet extract was concentrated and then transferred to a preweighed tube. The extract was completely dried under a gentle nitrogen stream and then kept at 60 °C until constant mass. The measured weight represented the fat content of the sample.

Organochlorine Analysis. One microliter was injected in pulsed splitless mode on a Hewlett-Packard 6890 GC connected via direct interface with a HP 5973 mass spectrometer. A 50 m \times 0.22 mm \times 0.25 mm, HT-8 capillary column (SGE, Zulte, Belgium) was used with helium as carrier gas at a constant flow of 0.7 mL/min. Injector and interface temperatures were set at 270 and 300 °C, respectively. The oven temperature program began at 90 °C, was kept there for 1 min, then was increased at 15 °C/min to 170 °C, was held for 3 min, then was increased at 4 °C/min to 270 °C, was held for 1 min, and was further increased at 10 °C/min to 290 °C and held for 15 min. The mass spectrometer was operated in electron impact ionization mode. Three most abundant ions were monitored for each level of chlorination for PCBs or for each pesticide. Method limits of detection (LOD) for individual PCB congeners ranged between 0.1 and 0.5 ng/g lipid. For HCHs and DDTs, the detection limit was 0.2 ng/g lipid for each isomer. Recoveries of target compounds ranged between 72% and 80%.

PBDE Analysis. The analysis, previously described in Covaci et al. (32), was performed on a Hewlett-Packard 6890 GC equipped with a 10 m \times 0.10 mm \times 0.10 μm AT-5 (5% phenyl polydimethyl siloxane) capillary column (Alltech,

TABLE 3. Congener-Specific PCBs in European (Scottish, Irish, and Norwegian) Atlantic Salmon, Aquaculture Feed, and Fish Oil: Lipid Adjusted (ng/g, ppb)

PCB congener	salmon													feed							fish oil							
	M11	M12	M13	M14	M24	M25	M28	M31	M18	M19	M20	M21	M22	M01	M02	M03	M04	M05	M15	M16	M17	M06	M07	M08	M09	M10	MDEPA	
16	0.6	1.7	ND ^e	0.8	ND	0.4	0.4	1.2						1.4	37.8	23.0	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
17	0.6	2.5	ND	1.4	0.4	0.5	0.5	1.3						1.1	57.8	34.5	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
18	1.8	6.1	ND	3.6	1.3	1.3	1.3	3.2						3.1	138.3	81.2	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
28	4.8	9.1	0.4	6.2	3.9	3.6	3.1	5.8	7.2	7.5	4.4	2.8	5.5	2.8	153.5	91.2	0.8	0.8	ND	1.2	1.2	0.6	0.3	ND	ND	ND	0.4	
31	3.4	7.0	ND	5.0	2.9	2.9	2.5	4.5						2.1	119.5	71.6	0.6	0.6	ND	0.8	0.9	0.4	ND	ND	ND	ND	ND	
32	0.5	2.2	ND	1.0	0.4	0.5	0.4	0.9						0.8	44.7	26.8	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
33/20	1.9	4.6	ND	2.6	1.1	1.0	1.1	2.6						2.6	91.5	55.2	0.8	ND	ND	ND	ND	0.3	ND	ND	ND	ND	ND	
44	7.0	6.3	ND	9.6	4.8	4.8	4.0	5.2						3.3	41.9	28.4	2.6	3.7	4.7	4.5	3.3	1.7	ND	3.0	ND	ND	12.9	
47/48	3.5	3.8	ND	4.6	2.2	2.4	2.3	2.3						1.5	27.9	19.4	2.1	ND	1.8	2.1	1.7	1.0	ND	2.3	ND	ND	ND	
49	4.4	5.1	ND	8.4	3.1	3.3	3.5	3.3						1.7	34.9	23.8	2.0	ND	3.1	2.9	2.4	1.3	ND	2.7	ND	ND	ND	
52	13.9	10.9	ND	16.0	9.1	9.1	6.3	6.4	6.3	11.6	16.7	6.2	11.2	3.4	41.9	28.1	3.3	2.2	6.7	7.1	6.4	4.1	ND	7.2	ND	ND	4.6	
66	7.0	6.5	ND	10.6	4.3	4.2	3.8	4.0						1.6	39.5	26.4	2.4	ND	3.0	2.9	2.2	0.3	ND	ND	ND	ND	8.2	
70	9.5	9.0	ND	17.9	6.8	7.0	6.2	7.5						3.3	36.5	25.4	3.8	ND	5.4	5.4	4.9	2.9	ND	ND	ND	ND	ND	
74	5.1	4.6	ND	9.0	3.9	3.7	3.6	4.4	2.0	7.0	4.7	2.4	5.7	1.1	24.8	16.5	2.0	ND	2.2	2.9	2.1	ND	ND	ND	ND	ND	5.8	
84	1.7	1.3	0.6	2.1	1.6	1.6	1.6	1.5						0.7	1.6	1.1	1.3	0.7	0.8	0.9	0.7	0.4	ND	1.1	ND	ND	2.3	
85	2.1	1.5	1.0	2.9	2.1	2.1	1.8	1.6						1.1	2.8	2.4	1.3	0.6	1.1	1.1	1.1	0.4	ND	1.3	ND	ND	3.9	
87	5.1	3.2	0.5	7.8	4.5	4.6	4.3	4.2						2.0	5.7	4.3	3.2	1.6	2.8	3.0	2.5	2.9	ND	2.9	0.4	ND	7.0	
91	0.7	0.6	ND	1.0	0.7	0.7	0.6	0.6						ND	1.5	1.0	0.6	ND	ND	ND	ND	ND	0.6	ND	ND	ND	ND	
95	6.2	4.5	0.9	9.4	6.0	6.1	5.7	6.0						2.5	8.5	6.8	5.1	2.5	3.1	3.4	3.2	1.7	ND	4.6	0.4	ND	6.5	
97	2.3	1.7	ND	3.5	2.0	2.0	1.9	1.9						0.8	2.8	2.4	1.7	0.8	1.2	1.4	1.3	0.6	ND	1.6	ND	ND	3.5	
99	14.3	9.7	4.3	17.9	9.5	9.3	8.7	7.6						3.6	8.9	6.9	6.4	2.9	6.2	6.7	5.7	3.3	ND	10.5	0.3	ND	15.1	
101	26.5	19.1	0.8	32.4	18.8	18.8	18.5	16.9	26.5	23.4	24.2	11.6	26.3	4.3	16.7	12.9	12.2	3.0	10.3	12.7	10.6	4.1	0.5	15.1	0.5	ND	23.4	
105	7.3	4.5	3.4	9.0	5.4	5.1	4.8	4.4	6.4	6.1	5.8	1.9	6.2	0.9	5.1	3.9	2.7	ND	2.6	3.3	2.6	0.6	ND	ND	ND	ND	8.4	
110	19.5	14.9	1.6	24.0	11.2	11.3	11.7	11.6	16.7	14.3	13.0	5.2	16.0	4.0	15.6	12.0	17.2	4.1	9.5	11.1	10.0	4.3	0.4	12.9	0.4	ND	24.3	
114	1.0	0.9	ND	1.5	0.6	0.5	0.6	0.3						ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
118	24.5	16.7	11.5	31.6	16.5	16.1	15.9	13.7	13.1	11.1	10.6	5.9	18.6	8.0	13.8	10.4	10.3	6.1	11.1	12.4	10.5	5.3	0.6	0.9	0.9	ND	33.5	
123	2.2	1.1	0.7	1.8	1.5	1.4	1.4	1.1						0.7	1.0	0.9	1.1	ND	1.2	0.8	0.9	0.4	ND	ND	ND	ND	2.5	
128	3.2	2.0	2.0	3.8	3.0	2.6	2.5	2.3	5.3	6.5	4.9	2.4	6.6	1.4	0.7	0.8	1.8	1.4	1.7	1.9	1.7	0.9	ND	3.1	ND	ND	3.9	
130	1.4	1.2	0.7	1.6	1.0	1.0	1.3	1.1						0.6	0.6	0.6	1.0	ND	0.8	0.8	0.8	0.5	ND	1.6	ND	ND	2.1	
132	4.6	4.0	1.5	5.7	3.3	3.4	3.9	4.1						1.8	2.9	2.6	3.3	1.6	3.3	3.1	2.9	1.5	ND	5.4	ND	ND	5.2	
137	1.1	0.7	0.5	1.5	0.7	0.8	0.7	0.6						ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
138	33.1	22.6	19.8	33.9	24.4	24.1	27.9	20.5	41.8	47.8	38.4	14.6	48.5	10.2	12.4	9.6	15.0	6.3	13.1	15.1	12.9	3.9	0.3	26.1	2.0	ND	54.7	
141	3.9	2.9	1.6	5.5	3.0	3.0	3.8	2.7						1.6	1.8	1.6	1.9	1.8	2.4	2.3	2.2	1.0	ND	3.3	ND	ND	5.5	
144	3.1	2.0	0.8	3.3	2.3	2.1	2.4	2.2						0.8	1.9	1.3	1.7	0.8	1.6	1.5	1.5	0.8	ND	3.1	ND	ND	ND	
146	7.9	6.0	3.2	8.7	5.6	5.4	6.7	5.4						3.2	3.5	3.0	4.8	2.7	4.1	4.4	3.7	2.2	ND	8.5	0.9	ND	11.3	
149	21.8	17.4	6.2	26.7	15.7	15.9	18.6	17.4	26.9	22.0	20.6	8.0	25.2	7.1	11.4	9.7	13.7	6.5	11.9	13.2	11.6	5.9	0.5	26.6	0.9	ND	23.4	
151	6.4	4.6	2.4	8.0	5.1	5.0	5.8	5.1						2.1	2.8	2.5	3.5	2.0	3.3	3.7	2.9	1.6	ND	6.2	0.5	ND	8.2	
153	48.2	33.8	23.7	49.0	35.4	34.9	41.4	29.9	36.2	51.9	37.2	15.0	52.5	17.8	18.1	14.6	23.8	12.3	21.6	22.9	19.5	8.4	0.5	42.7	3.5	ND	83.5	
156	2.1	1.2	2.0	2.2	1.6	1.7	2.0	2.3	1.9	2.5	2.0	0.6	1.8	1.9	0.6	ND	1.3	0.7	0.7	1.6	1.3	0.5	ND	ND	ND	ND	4.4	
158	2.3	1.4	2.8	2.2	1.5	1.5	1.9	1.5						0.8	0.9	0.7	0.9	0.8	1.0	1.2	1.0	0.5	ND	1.7	ND	ND	3.4	
163	11.6	9.9	7.1	13.7	8.9	8.8	11.2	9.1						4.4	5.4	4.9	7.8	3.9	6.4	7.2	5.9	3.1	ND	4.3	1.4	ND	16.4	
167	1.1	0.4	0.7	1.4	1.2	1.1	1.4	1.0						ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	2.2	
170	3.7	2.0	6.3	3.5	3.7	3.4	4.8	2.7	5.6	6.4	4.1	1.6	5.9	1.0	1.0	ND	0.7	ND	0.8	1.4	0.9	0.5	ND	3.2	ND	ND	6.0	
171	1.0	0.7	1.3	1.0	0.9	0.9	1.1	0.7						ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.9	ND	ND	1.9	
172	0.7	0.5	0.9	0.8	0.6	0.7	0.9	ND						ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.8	ND	ND	1.0	
174	2.3	1.7	2.0	2.9	2.4	2.3	3.3	2.1						ND	0.6	ND	0.9	ND	0.6	0.9	0.7	0.4	ND	2.9	ND	ND	3.0	
176	0.6	0.5	0.4	0.9	0.5	0.5	0.7	0.6						ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.7	ND	ND	ND	
177	3.0	2.4	3.1	3.2	2.4	2.4	3.3	2.3						0.6	0.8	0.7	1.5	ND	0.7	1.1	0.8	0.5	ND	3.8	ND	ND	2.2	
178	1.5	1.1	0.9	1.7	1.2	1.2	1.5	0.9						ND	ND	ND	0.6	ND	ND	ND	ND	ND	ND	1.7	ND	ND	2.0	
179	2.1	1.6	0.4	2.6	1.7	1.7	2.0	1.6						ND	ND	ND	0.8	ND	ND	ND	0.7	0.6	0.2	ND	2.3	ND	ND	1.5

180	11.8	7.3	13.7	13.9	10.3	10.0	12.8	7.2	9.3	14.6	9.5	4.5	14.0	2.9	2.2	1.8	3.0	1.0	2.3	3.4	2.7	1.3	ND	9.6	0.8	ND	20.5
183	3.3	2.0	2.9	3.4	2.8	2.8	3.5	2.2	2.8	4.0	2.5	1.4	4.5	0.6	ND	ND	0.9	ND	0.8	0.7	0.2	ND	3.4	ND	ND	5.2	
187	11.5	8.8	5.2	12.5	9.4	9.3	11.6	7.9	9.2	15.3	10.4	4.5	19.4	3.0	3.9	3.0	5.8	2.0	4.4	5.2	3.4	0.6	ND	17.1	0.4	ND	12.2
190	1.1	0.7	1.2	0.9	1.2	1.8	1.7	1.4	ND	0.9	ND	0.4	1.2	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	2.0	
194	0.9	0.5	1.2	0.9	1.8	1.8	1.1	0.5	ND	0.9	ND	0.4	1.2	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.8	ND	ND	1.4	
195	ND	ND	0.4	ND	ND	ND	0.4	ND	ND	0.9	ND	0.4	1.2	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.4	ND	ND	1.7	
196/203	1.4	0.7	1.2	1.3	1.2	1.1	1.4	0.7	0.4	2.0	1.0	0.6	2.5	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.5	ND	ND	1.7	
199	1.2	0.6	0.9	1.4	1.4	1.4	1.9	1.1	0.4	2.0	1.0	0.6	2.5	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.7	ND	ND	1.6	
ΣPCBs ^a	375.5	300.5	145.1	459.9	279.1	277.1	300.0	261.4	NA	NA	NA	NA	NA	124.0	1049.5	678.2	182.0	81.2	163.6	183.5	156.4	72.9	8.2	251.8	17.7	5.8	450.2
Σ seven markers ^b	162.8	119.5	70.1	183.0	118.4	116.6	125.9	100.4	140.4	167.9	141.0	60.6	176.6	49.4	258.6	168.6	68.4	31.7	65.4	74.8	63.8	27.7	2.4	101.7	7.9	0.7	220.6
% seven marker/Σ	43.4	39.8	48.3	39.8	42.4	42.1	42.0	38.4	38.4	24.6	29.7	24.6	39.9	39.9	24.6	24.9	37.6	39.1	40.0	40.8	40.8	38.0	39.3	40.4	44.6	12.1	49.0
ΣPCBs ^c	273.9	205.7	115.4	318.3	202.0	199.5	217.5	180.8	226.6	265.1	219.8	94.0	284.4	81.5	456.8	299.7	130.3	55.8	113.8	131.9	111.6	48.5	4.7	185.9	12.4	2.3	353.0
MO-PCB-TEQ	5.8	3.7	2.9	6.8	4.1	3.9	4.1	3.7	2.9	3.0	2.6	1.1	3.4	2.3	2.8	2.1	2.6	1.2	2.5	2.9	2.5	1.1	0.2	0.3	0.3	0.2	7.7
NO-PCB-TEQ ^d	20.7	6.7	14.4	18.7	11.0	13.2	12.8	8.3	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB-TEQ	26.5	10.4	17.3	25.5	15.0	17.2	16.9	12.0	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
diox-TEQ ^d	11.5	5.1	6.2	18.2	6.8	6.1	5.0	5.6	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
TEQ	38.1	15.4	23.5	43.7	21.9	23.2	21.9	17.6	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PCB/diox-TEQ	2.3	2.1	2.8	1.4	2.2	2.8	3.4	2.1	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

^a Sum of 59 congeners. ^b IUPAC nos. 28, 52, 101, 118, 138, 153, and 180. ^c Sum of 23 PCB congeners as measured in samples M18–M22. ^d Described in ref. 23. ^e NA = not available. ND = not detected (values below limit of detection were substituted with ¹/₂LOD, upper-bound method).

^a Sum of 59 congeners. ^b IUPAC nos. 28, 52, 101, 118, 138, 153, and 180. ^c Sum of 23 PCB congeners as measured in samples M18–M22. ^d Described in ref 23. ^e NA = not available. ND = not detected (values below limit of detection were substituted with ¹/₂LOD, upper-bound method).

Belgium) and connected via direct interface with a HP 5973 mass spectrometer. Helium was used as carrier gas at constant flow (0.4 mL/min). The low-resolution quadrupole mass spectrometer (EI) was operated at 70 eV in the selected ion monitoring (SIM) mode. The ion source and quadrupole temperatures were 230 and 250 °C, respectively. The electron multiplier voltage was set at 2300 V. Dwell times were set at 10 ms. The two most abundant ions (M^+ and $[M + 2]^+$ for the tri-, tetra-, and penta-congeners and $[M - 2Br]^+$ and $[M - 2Br + 2]^+$ for the hexa-congeners) were monitored for each level of bromination for native and labeled PBDEs. Recoveries of internal standards, ¹³C-labeled BDEs, ranged between 81% and 103% (RSD < 21%). Method limits of detection (LOD) for individual PBDE congeners ranged between 0.1 and 0.4 ng/g lipid.

Quality Control/Quality Assurance. Retention times, ion chromatograms, and intensity ratios of the monitored ions were used as identification criteria. A deviation of the ion intensity ratios within 20% of the mean values of the calibration standards was considered acceptable. The method performance was assessed through rigorous internal quality control, which included a daily check of calibration curves and regular analysis of procedural blanks and certified material CRM 350 (PCBs in mackerel oil). The method was tested by participation in an interlaboratory test organized by the Institute for Reference Measurements and Materials (IRMM, Geel, Belgium). Seven PCB congeners (nos. 28, 52, 101, 118, 138, 153, and 180) were determined in nonspiked and medium- and high-level spiked pork fat. The results of the individual PCB congeners deviated less than 10% from the target values. For PBDEs, the analysis of two samples of biota (eel and porpoise liver) used for the first interlaboratory test on PBDE (33) showed a variation of less than 10% from mean values obtained from 18 participating laboratories.

Results

All samples examined contained detectable residues of organochlorine contaminants. Summary results are presented in Table 2, with total sum (Σ) values given using a value equal to ¹/₂LOD where there were no detections, as recommended by the EC Scientific Committee on Animal Nutrition (5). Detailed results of PCB congeners are presented in Table 3. All concentrations were adjusted to the lipid content of the sample. The o,p' isomers of DDTs were not found in any sample. PBDEs were detected in all fish samples and in all feed samples but only in one of the fish oil samples (see Table 4). The total concentration of selected PBDEs is the sum of concentrations of the congeners measured and is an underestimate of the total BDE concentration, as other isomers present were not measured.

Salmon Samples. PCB concentrations in the farmed samples were between 145.1 and 459.9 ng/g lipid weight. Profiles and concentrations of PCBs were similar and of the same order of magnitude for all of the farmed samples but lower in a Norwegian sample (M13) and a magnitude lower in a wild Irish sample (M21). In the sample M13, the PCB profile is dominated by the hexa- and hepta-PCB congeners, while in the other samples, the lower chlorinated congeners (tri-, tetra- and some penta-PCB congeners) had a higher contribution to the ΣPCBs. The mean of the seven ICES marker PCBs was 129.5 ± 38.1 ng/g lipid, and they constituted 41.1% ± 1.8% of the total ΣPCB congeners for the Scottish salmon samples.

The mean for HCB for all the salmon samples ($n = 13$) was 11.6 ± 11.9 ng/g lipid, but was much lower in sample M13 (under the detection limit of 0.2 ng/g lipid). The mean concentration for HCHs (Σα, β, γ-HCH) was 13.8 ± 7.6 ng/g lipid for the samples obtained in Scotland and 8.5 ± 9.1 ng/g lipid for all of the salmon samples ($n = 13$). γ-HCH was the predominant HCH isomer with a mean ratio γ-HCH/SHCHs

TABLE 4. Selected PBDEs in European Atlantic Salmon, Aquaculture Feed, and Fish Oils: Lipid Adjusted (ng/g, ppb)

sample codes	BDE 28	BDE 71	BDE 47	BDE 75	BDE 66	BDE 100	BDE 99	BDE 154	BDE 153	ΣBDEs	BDE47/Σ
M11	1.3	7.1	29.3	0.3	2.0	6.5	4.9	1.3	1.0	53.7	0.55
M12	0.9	4.4	14.7	ND ^a	ND	2.9	2.8	0.7	0.8	27.2	0.55
M13	ND	ND	0.4	ND	ND	ND	0.7	ND	ND	1.1	0.37
M14	0.8	1.7	12.9	ND	0.9	2.4	5.9	ND	ND	24.6	0.52
M24	1.5	7.1	28.9	0.2	2.2	6.4	5.0	1.6	ND	52.9	0.55
M25	1.5	7.1	28.0	0.4	1.8	6.1	4.4	1.0	ND	50.3	0.56
M28	2.0	7.9	43.0	0.3	3.2	9.9	14.0	3.6	1.3	85.2	0.50
M31	1.3	6.7	25.0	0.5	1.6	5.6	4.0	0.9	ND	45.6	0.55
M18	ND	ND	3.1	ND	ND	ND	ND	ND	ND	3.1	
M19	0.5	2.8	10.2	ND	0.8	2.8	1.6	0.2	ND	18.9	0.54
M20	0.5	2.4	9.4	ND	0.4	1.5	1.5	ND	ND	15.7	0.60
M21	0.2	0.7	5.0	ND	0.1	1.1	1.3	ND	ND	8.4	0.59
M22	1.0	8.1	25.8	0.7	1.7	7.9	5.6	1.3	ND	52.1	0.49
M01	0.3	1.4	7.0	ND	ND	ND	ND	ND	ND	8.7	0.81
M02	0.5	3.1	8.2	0.2	0.5	1.5	1.7	ND	ND	15.6	0.53
M03	0.5	3.1	8.2	ND	0.7	1.5	1.4	ND	ND	15.4	0.54
M04	0.7	4.3	12.3	0.3	0.7	1.8	2.7	ND	ND	22.8	0.54
M05	ND	1.4	5.1	ND	ND	0.9	0.7	ND	ND	8.1	0.63
M15	0.6	4.6	13.2	ND	ND	2.3	2.3	ND	ND	23.0	0.57
M16	0.6	4.4	13.0	0.3	0.9	2.4	2.2	ND	ND	23.9	0.54
M17	0.5	3.0	9.5	ND	ND	1.8	1.4	ND	ND	16.2	0.59
M06	0.3	2.7	7.1	ND	ND	1.4	1.2	ND	ND	12.7	0.56
M07, 08, 09, 10	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	

^a ND = not detected.

of 0.69 ± 0.01 . The mean concentration of DDTs ($\Sigma p,p$ -DDE, p,p -DDD, and p,p -DDT) was 69.6 ± 58.4 ng/g lipid for all the samples ($n = 13$), and p,p -DDE was the predominant contributor to the Σ DDTs. The ratio of p,p -DDT/ Σ DDTs ranged from 0.1 to 0.31 (mean 0.22). Concentration of DDTs in M13 was much lower (5.1 ng/g lipid).

The PCB, organochlorine pesticide, and PBDE contaminant loading of the bioculture farmed salmon ($n = 2$) appears to be of the same magnitude as that of the farmed salmon; however, the sample size is too small to draw any firm conclusions. Bioculture salmon diets are similar to those of farmed salmon with regard to fishmeal content, but the maximum fish oil content is 28% of the total diet (34).

All fish contained detectable levels of PBDEs; the sum of the BDEs (Σ BDEs) ranged from 1.1 to 85.2 ng/g lipid for the 13 salmon samples. The highest levels were found in a wild (or possible farm escapee) salmon sample. As expected, BDE 47 dominated the profiles, with a mean ratio BDE 47/ Σ BDEs of 0.53 ± 0.04 . The levels of PBDEs found the salmon analyzed in this study are comparable with those found in other cultured oily fish, particularly gray mullets and yellow tails reported recently in a Japanese study (18), and in wild fish (14).

Aquaculture Feeds. Concentrations of PCBs in the eight feed samples ranged between 75.6 and 1153.2 ng/g lipid (Table 2). Six feed samples presented the same profile of PCBs, with the penta- and hexa-PCB congeners representing up to approximately 80% of the total PCB content. The seven ICES marker PCBs constituted approximately 40% of the total SPCB congeners. As expected, the concentration of PCBs were high (147 ± 41 ng/g lipid), probably due to the high fish oil content of some samples (up to 35%). Two samples from the same manufacturer (M2 and M3) presented a totally different profile of PCB congeners when compared with the other six feed samples. Tri- and tetra-congeners (approximately 55% and 28%, respectively) dominated, and the profile was close to that of Aroclor 1242. This different profile correlated with the high PCB concentrations found for these two samples (the highest concentrations detected in all of the samples) and could point to the existence of local PCB sources or illegal dumping of used Aroclor mixtures in the

animal food chain (30). The mean for HCB was 9.2 ± 2.9 ng/g lipid, while for HCHs, the mean was 23.7 ± 14.9 ng/g lipid, with γ -HCH being the predominant HCH isomer (mean ratio γ -HCH/ Σ HCHs of 0.63 ± 0.16). The mean for the Σ DDTs was 42 ± 8 ng/g lipid, with p,p -DDE the predominant contributor to the Σ DDTs (Table 2). The mean ratio p,p -DDT/ Σ DDTs was 0.13 ± 0.04 . The levels of PBDEs ranged from 8.1 to 23.9 ng/g lipid for the eight feed samples. The fry feed samples, with a far lower fish oil content, had lower residue levels than the feeds designed for smolts and adult salmon with a fish oil content over 20%.

Fish Oils. The fish oil samples showed varying PCB contents between 8.8 and 252.5 ng/g lipid (ND = $1/2$ LOD). Hexa- and penta-PCB congeners dominated the PCB profile for all samples. The same profile was found in fish oil used for diet supplement (sample MDEPA) which contained higher PCB amounts (450 ng/g lipid). However, the tri- and tetra-PCB congeners were higher contributors to the total sum in the fish oil M06 than in the fish oils M08 and MDEPA. Furthermore, the hepta- and octa-congeners were more important for the last two oils. The difference in profiles can be due to different species used for collection of the oil and to different processing procedures. During the oil processing (usually a steam distillation), some of the low chlorinated PCB congeners can be lost through their increased volatility.

Mean HCB concentrations were 2.5 ± 1.5 ng/g lipid, while mean HCHs levels were 10 ± 7 ng/g lipid. γ -HCH was the predominant HCH isomer with a mean ratio γ -HCH/ Σ HCHs of 0.50 ± 0.21 . Mean Σ DDT concentrations were 66 ± 87 ng/g lipid, and p,p -DDE was the predominant contributor and the mean ratio p,p -DDT/ Σ DDTs was 0.15 ± 0.07 . The levels and ratios of the HCB, HCHs, and DDT are reflected in the aquaculture feed, and the DDT levels were less wide ranging in the feeds. PBDE concentrations ranged from ND to 12.7 ng/g lipid.

Comparison between the Three Types of Samples. The three types of samples were compared using median values for all groups of compounds (Figure 1). Except for HCHs, all other groups of contaminants show an increase in concentration in the order fish oil < feed < salmon. Samples with concentrations close to the median value can be considered

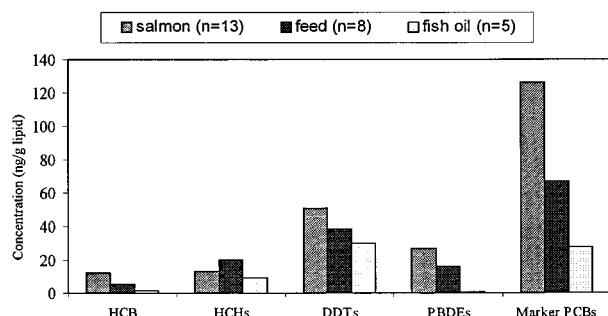


FIGURE 1. Median concentrations of HCB, HCHs, DDTs, PBDEs, and marker PCBs in salmon, feed, and fish oil samples.

as representative for each type of sample: M06 for fish oil, M04 for feed, and M24 for salmon.

For individual congeners (Figure 2), a higher contribution of PCB 18, 28, 31 (tri-PCBs); 66 (tetra-PCB); 101, 105 (penta-PCBs); 138 (hexa-PCBs); and all hepta- and octa-PCB congeners was found for the salmon, together with a lower contribution of PCB 47/48, 49, 52 (tetra-PCBs); 87, 110 (penta-PCBs); and 132, 146, and 149 (hexa-PCBs). The profiles of homologues were similar, with an increase of contribution of hepta- and octa-PCBs in the fish. Profiles of DDTs were similar in all three types of samples. For HCHs, the α -HCH isomer had a higher contribution in the fish oil, while the γ -HCH isomer had a lower contribution. With a constant contribution to the total PCB content, the ICES seven PCBs appear to be reliable predictors of the PCB contamination profile through all of the samples. For PBDEs, BDE 47 dominated the profiles, as expected, ranging between 50% and 100% of the Σ BDEs included in the analyses. There was no significant difference in the PBDE profiles for the three matrixes. Samples with higher PCB contents generally showed higher levels of the pesticide residues, but this was not the case with the PBDEs, indicating the existence of different pollution sources.

Discussion

The PCB results from this study are in good agreement and of a similar order of magnitude to the values for salmon reported in recent studies (35, 36) (see Table 5). The comparability of the PCB data suggests that both the data sets can be considered fairly representative of PCB concentrations in British farmed salmon. Previous reports have detected significant levels of PCBs and organochlorine pesticides in fatty fish such as herring and salmon (Table 5) (35–39). The seven marker PCB concentrations in the farmed-salmon samples from this study and a Dutch study (40) were very similar. The Dutch study found PCB concentrations of 110 and 170 ng/g lipid in farmed salmon from Norway, while farmed salmon from Scotland contained 135 and 210 ng/g lipid ranging from 61 to 183 ng/g lipid. The Dutch study also found that farmed salmon from Norway or Scotland had relatively low TEQ values (<4 pg TEQ/g whole weight). Assuming a mean value of 20% of the lipid content (40), the TEQ value becomes approximately 20 pg of TEQ/g of lipid, similar with or somehow lower than the values found in our study (see Table 3). Wild salmon from polluted waters contain higher concentrations of PCBs. Up to 3500 ng/g lipid for the sum of six PCB congeners are reported for Baltic salmon (41). In Lake Michigan Coho and Chinook salmon (*Oncorhynchus kisutch* and *Oncorhynchus tshawytscha*), mean concentrations of PCBs (78 congeners) were 39 870 and 53 638 ng/g lipid (42), much higher than that in the farmed salmon analyzed in this study.

PCB profiles are more species-dependent for fish and birds than for mammals due to selective metabolism depending upon the degree of chlorination. Fish species can accumulate

lower chlorinated PCB congeners (such as tetra- and penta-PCBs), some of which have the highest TEF values among dioxin-like PCBs (29). Indeed, levels of PCBs are higher in fish as compared with other farm-raised animals (such as chicken, beef, or pork) intended for human consumption. The concentrations of organochlorine pesticides in salmon compare favorably with those reported for farmed salmon obtained during 1997 in the United Kingdom. Σ DDT concentrations ranged from 10 to 40 ng/g whole weight ($n = 14$), γ -HCH concentrations ranged from ND in 11 samples to 2 ng/g whole weight in 3 samples, and HCB ranged from 2 to 5 ng/g whole weight ($n = 14$) (42). The United Kingdom does not have statutory maximum residue levels (MRLs) for these pesticides in fish, although they exist for other food products (43).

The average concentration across all salmon samples of the Σ BDE congeners was 33.7 ng/g lipid. The results from this study are an order of magnitude lower than the respective PBDE values for salmon reported in recent studies from polluted open waters. Previous reports have detected significant levels of PBDEs in fatty fish such as sprats, herring, and salmon from the Atlantic and more polluted waters such as the Baltic (12, 13), showing age-related accumulation and biomagnification. The concentrations of PBDEs in the Atlantic salmon were markedly lower than that in Lake Michigan Coho and Chinook salmon (44) (mean 2440 ng/g lipid, range 773–8120 ng/g lipid). Salmon from Lake Michigan tributaries in the United States appear to have the highest PBDE concentrations so far reported worldwide, several orders of magnitude greater than the concentrations reported here, but with a similar congener distribution (44).

PCB contamination for the farm-raised salmon comes via aquaculture feed, while for the wild salmon sources, are more diffuse but clearly biomagnify through trophic levels (42). One of the routes of food chain contamination may be partly as a consequence of ingestion of plastic resin pellets (45) by the marine wild fish, used in the manufacture of fish meal. There is very little literature available for PCB and organochlorine pesticide comparison in fish feed. Data from the 1970s exists for Aroclor 1254 in nine feed samples (range 0.05–0.23 mg/g whole weight) and p,p' -DDE (range 0.05–0.34 mg/g whole weight) (46). Fish meal, a principal component of fish feed, shows a much higher degree of contamination with organochlorines if sourced from North Atlantic stocks than those from South Pacific (5). Thus, fish meal originating from Denmark and the Faroe Islands contained 120 and 250 ng/g lipid for the seven marker PCBs, respectively, while fish meal from Peru had a concentration of 10 ng/g lipid (30).

The PCB concentrations in fish oils from this study are consistent with the study reported by the United Kingdom Ministry of Agriculture, Fisheries and Food (now the Food Standards Agency) (7), with surprisingly high levels of PCB congeners. A previous study (6) has found relatively high levels of PCBs 101, 118, 138, 149, 153, 170, and 180 in both a herring oil sample from Germany and a salmon oil sample. For example, the concentration of PCB 118 was 158 μ g/L lipid in the herring oil and 60 μ g/L lipid in the salmon oil.

The average concentration of the Σ BDE congeners in the analyzed feed samples was 16.6 ng/g lipid. At the present time, comparative data are not available in the public domain. The target PBDEs were not detected in all but one of the oil samples, a fish oil from the Northern Hemisphere, with a Σ PBDE concentration of 12.7 ng/g lipid. The data available in the literature concerning organohalogen contamination of fish oils varies widely, with higher contamination levels observed from more polluted Northern hemisphere waters (5, 6, 47).

Wild salmon from Lake Michigan have been shown to have a significant relationship with the congener concentra-

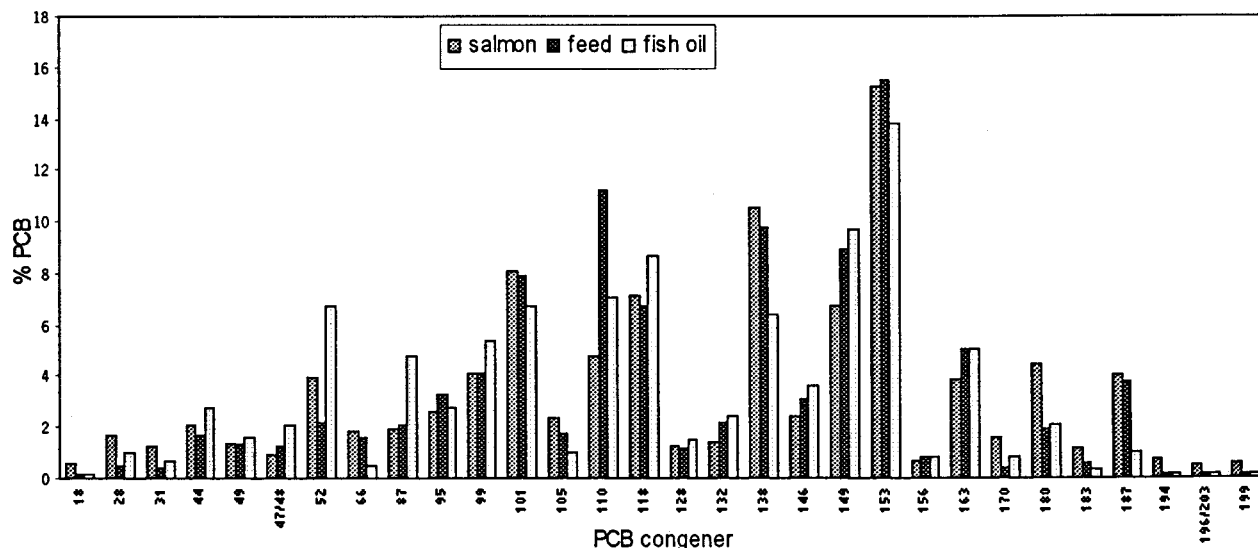


FIGURE 2. Profiles of individual PCB congeners in salmon, feed, and fish oil.

TABLE 5. Comparison of Mean Congener-Specific PCB (ng/g, ppb, lipid weight) with Means of Salmon Samples from This Study ($n = 8$) and Parsley et al. ($n = 12$) (35)

PCB congener	salmon, mean \pm SD (ng/g lipid weight)		herring, mean \pm SD (ng/g lipid weight) Parsley et al. ($n = 10$)
	this study ($n = 8$)	Parsley et al. ($n = 12$)	
18	2.8 ± 1.8	1.3 ± 0.6	1.2 ± 0.5
28	7.4 ± 8.7	4.3 ± 1.3	8.0 ± 3.6
31	4.0 ± 1.6	2.7 ± 0.8	3.5 ± 1.5
47	3.0 ± 1.0^a	2.1 ± 0.6	4.4 ± 2.2
49	4.4 ± 1.9	4.0 ± 1.2	4.4 ± 2.1
52	10.2 ± 3.7	10.2 ± 3.4	17.8 ± 8.7
99	10.2 ± 4.2	9.6 ± 2.9	29.8 ± 14.2
101	19.0 ± 9.0	22.8 ± 6.8	55.7 ± 25.9
105	5.5 ± 1.8	6.3 ± 1.8	20.3 ± 10.5
114	0.8 ± 0.4	0.4 ± 0.1	0.9 ± 0.5
118	18.3 ± 6.5	19.0 ± 5.8	60.3 ± 28.7
123	1.4 ± 0.5	0.9 ± 0.3	2.4 ± 1.2
128	2.7 ± 0.6	4.6 ± 1.4	14.7 ± 7.2
138	25.8 ± 5.4	34.3 ± 10.0	110.5 ± 46.8
153	37.0 ± 8.7	37.2 ± 11.1	129.7 ± 70.0
156	1.9 ± 0.4	1.8 ± 0.6	5.3 ± 3.1
167	1.0 ± 0.3	1.1 ± 0.3	2.5 ± 1.3
180	10.9 ± 2.7	11.6 ± 4.5	40.7 ± 20.4
lipid (%)	13.3 ± 4.9	13.6 ± 3.3	9.6 ± 2.9

^a PCB 47/48.

tions in lower trophic levels of invertebrates. PCB congeners biomagnify 20–30-fold, with a more efficient transfer of penta- and hexa-PCBs through the pelagic food web (42).

The risks of exposure to organochlorine contaminants from eating relatively contaminated fish have been known for many years, but how to quantify the risks to human health is frequently disputed. While PCBs and *p,p'*-DDE provide the bulk of organochlorine residues in human tissues (48), there is a lot of variability in regulations for PCB and pesticide concentrations in food worldwide. Many E.U. member states do not have national maximum limits for concentrations of PCBs in food (3). However, in Holland, the current norm for fish designated for human consumption is 620 ng/g lipid for the sum of the seven marker PCBs (40), a value lower than the concentrations observed in Baltic or Lake Michigan salmon. Because of comparatively lower toxicity, the risk assessment of nondioxin-like PCBs have been given a lower priority than dioxin-like PCBs but with the proviso that the

PCB risk assessment (49) should be updated on a congener-specific toxicological evaluation to provide an adequate risk assessment soon (5). In terms of risk assessment, the potential contribution to the human diet of PCBs, organochlorine pesticides, and PBDEs from farmed European salmon will vary according to the diet and age of the fish, the frequency of consumption, portion size, cooking practices, and the age of the consumer. The possible contribution to dietary intakes of organohalogen compounds from farmed salmon could clearly be significant for high consumers, particularly if pregnant or breast feeding. As the PCB data is corroborated by Parsley et al. (35), national extrapolations could be suggested on the basis of this study but not for the other target contaminants because of the relatively small sample size and lack of available comparative data.

Further investigation of farmed salmon and salmon feed, including feed fortified with fish oil and feed fortified with selected vegetable oils, are warranted, together with consistent regular vigilance by health authorities for PCBs, organochlorine pesticides, and PBDEs to monitor and reduce current human dietary exposure. While PCBs and many organochlorine pesticides have been banned in most of the world, they are still being delivered in the diet, and European farmed salmon can be a significant source. In addition, in some cases, there are suggestions of recent usage, particularly of DDT.

While diets based on marine fish oils are currently favored by the aquaculture industry, it is likely that these oils are contributing greatly to the contamination of farmed salmon by POPs. The degree of variations in levels of contaminants for the feed and fish oil data indicate the significance of changes in feedstock and variations in oil sources and processing procedures that can result in batch-to-batch differences in contaminant levels, even in the final product from a single manufacturer. The generation of additional data is needed to further assess the importance of batch variation in animal feeds.

There is experimental evidence that suggests aquaculture diets utilizing vegetable oils (with both n-3 and n-6 fatty acids), and having fatty acid compositions which resemble those of invertebrates that comprise the natural diet of salmon, could be more beneficial in accommodating successful seawater adaptation than diets based on marine fish oils (50). Vegetable oil-based diets could also facilitate the requirement for high-energy aquaculture feeds on an economically competitive basis, while reducing the problems of organochlorine contamination. Contamination problems do

exist with vegetable oils but to a far lesser degree than fish oils (6). This would reduce the reliance on fish meal and oils from unsustainable natural resources and demonstrate sensitivity to public confidence after the recent PCB and dioxin food crises.

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