PERGAMON

Chemosphere 47 (2002) 183–191

CHEMOSPHERE

www.elsevier.com/locate/chemosphere

Investigation of polychlorinated dibenzo-*p*-dioxins, dibenzo-*p*-furans and selected coplanar biphenyls in Scottish farmed Atlantic salmon (*Salmo salar*)

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Received 18 January 2001; received in revised form 15 August 2001; accepted 25 September 2001

Abstract

Farmed and wild Scottish Atlantic salmon were obtained from retail suppliers, producers, and Stirling University in Scotland during January, 1999, for determination of 17 2,3,7,8-Cl-substituted PCDDs and PCDFs, and seven non-ortho- and mono-ortho-PCBs. The study confirms previous reports of relatively high concentrations of PCDDs, PCDFs and, especially, PCBs in farmed Scottish salmon. The results indicate that high consumption of salmon, particularly by children under 5 years, could lead to intakes above the tolerable daily intake (TDI) and tolerable weekly intake (TWI) for these chemicals, especially the PCBs, when combined with mean or high level intakes from the typical UK diet. These results suggest further investigation of farmed salmon and salmon feed, including feed fortified with fish oil and feed fortified with selected vegetable oils, is warranted.

Keywords: Farmed salmon; Polychlorinated dibenzo-*p*-dioxins (PCDDs); Polychlorinated dibenzo-*p*-furans (PCDFs); Polychlorinated biphenyls (PCBs); Persistent organic pollutants

Abbreviations: PCDDs – polychlorinated dibenzo-*p*-dioxins; PCDFs – polychlorinated dibenzo-*p*-furans; PCBs – polychlorinated biphenyls; ICES – International Committee for the Exploration of the Sea; ND – not detected; NA – not analysed; LOD – limit of detection; mt – metric ton; TDI – tolerable daily intake; TEF – toxic equivalent factors; TEQ – sum of toxic equivalents; TWI – tolerable weekly intake

1. 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, until very recently, little has been published on chlorinated compounds in aquaculture systems or products from aquaculture. Data are available on the concentrations of polychlorinated biphenyls (PCBs), polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in fish from wild stocks, where exposure is associated with chronic contamination due to leaching of agricultural or industrial chemicals from treated or contaminated soils into surface waters and the global distribution and deposition by atmospheric transport (North Sea Task Force, 1993). However, with farmed fish, potential exposure hazards are associated with the contaminated fish oil (World Health Organisation, 1999).

Fish oil is a by-product of the fish meal manufacturing industry and comes from many different parts

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0045-6535/02/$ - see front matter
PII: S0045-6535(01)00201-6

of the world. Highly chlorinated compounds and chlorinated insecticides are lipophilic and bioaccumulate in the food chain. As they accumulate in the lipid compartment of the animal, the fish oil extracted from fish caught in polluted waters may be contaminated with chlorinated hydrocarbons (Jacobs et al., 1997, 1998; Ministry of Agriculture, Fisheries and Food, 1997). Existing data on the levels of PCDDs, PCDFs and PCBs consumed in the UK have been mainly derived from total diet surveys and surveillance data for specific food types (Wright and Startin, 1995; Ministry of Agriculture, Fisheries and Food, 1995, 1996, 1997; Food Standards Agency, 2000). These data indicate the presence of elevated organochlorine contamination of farmed fish. Of 161 salmon samples tested in 1997, all were found to contain residues of PCBs (ICES 7 set of PCBs: IUPAC numbers 28, 52, 101, 118, 138, 153, 180) at concentrations of 23–620 ng/g fat (Ministry of Agriculture, Fisheries and Food/Department of Health, 1998; Veterinary Medicines Directorate, 1998). The UK Ministry of Agriculture, Fisheries and Food (MAFF) recently reported on a survey of marine fish sampled in 1995–1996 (Ministry of Agriculture, Fisheries and Food, 1999). The mean concentrations of PCDDs, PCDFs and PCBs in 12 salmon samples were 25 pg/g TEQ fat. For both these reports the World Health Organisation toxic equivalent factors (WHO-TEFs) were not the recently revised values (Kutz et al., 1990; Van den Berg et al., 1998).

The determination of these contaminants in fish and identification of the original sources of the contamination are, 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 225,462 metric tons (mt) to 687,906 mt during the years from 1990 to 1998 (FAO, 1998) with the UK, one of the three major producers, generating 110,917 mt (FAO, 1998, 2000).

The salmon aquaculture industry is most dependent on the use of fish oil as a source of dietary lipids and herring oil is the standard oil fed to salmon. In recent years, fish oil use as a source of dietary fat has increased significantly since 1995 from less than 30% to 33% and even 36.3% of the total 'high-energy diet' for salmon (Trouw Aquaculture, no date; Royce, 1996; Bell et al., 1998; BOCM Pauls Fish Feed Group, 1998). Currently the aquaculture industry accounts for around 17% of the global production of fish meal and oil but this has been predicted to double in the next 15 years by the FAO in Rome (Bell, 1998).

This paper presents the results of analysis of farmed Scottish salmon samples for PCDDs, PCDFs and selected coplanar PCBs. These values are compared with those obtained from salmon reported in other studies. The potential contribution of fish oils to the bioaccumulation of PCDDs, PCDFs and PCBs in farmed fish is discussed, together with suggestions on how to reduce these contaminant levels whilst also achieving nutritional and environmental benefits in an economically viable way for both industry and the consumer.

2. Experimental

2.1. Sample collection

Nine British salmon samples (seven samples were from individual fish, and two samples were composites of two fish each) and one Norwegian salmon (*Salmo salar*) sample that enter the European fish market were analysed. Salmon fish samples of variable age, both farm raised and wild, were obtained from eight different sites in January 1999, and were essentially samples of opportunity. The timing of the sample collection was in accord with the seasonal variation in the oiliness of fish fed diets containing greater than 28% lipid, as the exudation of free oil affects the ease of handling and preparation of the fish samples. The sampling times to avoid are September to mid-November, and the month of April (Royce, 1996; Bell et al., 1998). The fresh samples were obtained from commercial farms, supplying the local and national markets and national supermarket chains, and the Glasgow fish market. They were wrapped in polyethylene bags and packed in ice (for less than 3 h) prior to being placed in a deep freeze. Upon collection, each sample was given a unique laboratory reference number, and the sample details logged into a database. Laboratory reference numbers and GC-MS data file codes are included in the results tables. When all samples were collected, and frozen, they were packed in ice and transported directly by M. Jacobs to the laboratory where they were stored at −20 °C. Random sampling was not possible, but unbiased representation of the situation of interest was achieved by obtaining fish from the Northwest Scottish Highlands, the Western Scottish Highlands, the lowlands surrounding Stirling, plus a wild fish from the Scottish border with England, two samples for which no reliable information was available (but one may have been wild), and a Norwegian sample for which no information was available.

Whole body weights of the fish ranged from 400 g to 5.2 kg, the fish ranged in age from 1 year to 3+ years. Table 1 gives sample details for the salmon and it is important to note that the 'wild' fish may not have been genuinely wild, but were probably farm escapees (S. George, personal communication).

Table 1
PCDDs, PCDFs and selected coplanar PCBs in Scottish Atlantic salmon: lipid adjusted (pg/g, ppt)

Samples	TEF	213-02	213-04	213-05	213-06	213-07	213-08	213-09	213-10	213-11	213-12
Source		L. Tain (f)	L. Tain (f)	L. Tain (f)	R. Allen (w)	L. Leven (f)	Norway (f)	S. uk	S. uk	S.L.A (f)	S.L.B (f)
Approximate age of fish		3+ yrs	3+ yrs	3+ yrs	2 yrs	2+ yrs	2+ yrs	2 yrs	2 yrs	1 yr	1 yr
% Lipid		12.13	14.7	13.69	7.71	9.3	13.73	12.14	3.43	4.04	5.39
Sample weight (g)		99.8	99.7	100	100.8	102.1	100.9	100.1	100.9	99	100.9
Whole weight – WWt (g)		59.88	49.85	50	98.78	81.68	50.45	60.06	98.88	97.02	98.88
Lipid adjusted – lwt (g)		7.26	7.33	6.84	7.62	7.6	6.93	7.29	3.39	3.92	5.33
Congener											
2,3,7,8-TCDD	1	0.68	0.64	1.09	0.61	0.53	0.63	0.87	1.39	0.61	0.51
1,2,3,7,8-PeCDD	1	1.27	1.32	2.06	1.06	1	1.28	1.42	4.16	1.2	0.8
1,2,3,4,7,8-HxCDD	0.1	−0.17	−0.17	−0.18	−0.16	−0.16	0.17	−0.17	0.85	−0.32	−0.23
1,2,3,6,7,8-HxCDD	0.1	0.26	0.28	0.43	−0.16	−0.16	0.3	0.22	1.39	−0.32	−0.23
1,2,3,7,8,9-HxCDD	0.1	−0.17	−0.17	−0.18	−0.16	−0.16	−0.18	−0.17	−0.37	−0.32	−0.23
1,2,3,4,6,7,8-HpCDD	0.01	−0.17	−0.17	−0.18	−0.16	−0.16	−0.18	−0.17	−0.37	−0.32	−0.23
OCDD	0.0001	0.69	0.68	0.73	0.66	0.66	0.72	0.69	1.47	1.28	0.94
2,3,7,8-TCDF	0.1	17.85	15.39	27.42	11.87	12.57	15.95	18.02	48.52	15.15	12.92
1,2,3,7,8-PeCDF	0.05	1.52	1.18	2.69	0.88	1.04	1.4	1.65	5.65	1.34	0.99
2,3,4,7,8-PeCDF	0.5	5.8	4.37	10.63	3.97	4.22	5.02	6.51	13.95	4	3.75
1,2,3,4,7,8-HxCDF	0.1	−0.17	−0.17	0.18	−0.16	−0.16	−0.18	−0.17	0.68	−0.32	−0.23
1,2,3,6,7,8-HxCDF	0.1	−0.17	−0.17	*0.16*	−0.16	−0.16	−0.18	0.13	0.78	−0.32	−0.23
2,3,4,6,7,8-HxCDF	0.1	0.16	0.19	0.29	−0.16	0.12	0.25	0.19	1	0.24	−0.23
1,2,3,7,8,9-HxCDF	0.1	−0.17	−0.17	−0.18	−0.16	−0.16	−0.18	−0.17	−0.37	−0.32	−0.23
1,2,3,4,6,7,8-HpCDF	0.01	−0.17	−0.17	−0.18	−0.16	−0.16	−0.18	−0.17	−0.37	−0.32	−0.23
1,2,3,4,7,8,9-HpCDF	0.01	−0.17	−0.17	−0.18	−0.16	−0.16	−0.18	−0.17	−0.37	−0.32	−0.23
OCDF	0.0001	0.69	0.68	0.73	0.66	0.66	0.72	0.69	1.47	1.28	0.94
Upper-bound TEQ fat (ND = 0.5 LOD)		6.83	6.06	11.51	5.01	5.06	6.23	7.56	18.21	5.62	4.7
Lower-bound TEQ fat (ND = 0 LOD)		6.75	5.97	11.45	4.89	4.96	6.16	7.48	18.13	5.42	4.53
PCB 77	0.0001	190.2	288.2	536.1	528.1	234.7	281.2	242.8	622.4	360.3	600.7
PCB 105	0.0001	5970.2	6359.7	7220.9	6957	4040.1	5659.7	6233.8	9240.3	4684	5089.3
PCB 118	0.0001	21 389.2	2440.6	22 991.4	24 207.5	14 811.1	17 092.1	25 091.3	31 873.9	16 377.2	17 152.4
PCB 126	0.1	107.5	129.8	203.3	125.9	64.7	141.4	113.1	183	81.2	84.1
PCB 156	0.0005	1889.4	2531.8	2262.6	2179.9	1029.2	1952.2	2327.6	2436.1	1283.8	1345
PCB 157	0.0005	535.4	678.7	657.4	598.5	337.1	444.2	677	689.4	358.4	398
PCB 169	0.01	22.3	20.8	28.5	16.7	15.2	20.9	24.4	30.1	15.1	12.4
TEQ of seven PCBs		14.67	17.36	24.82	17.02	9.05	17.63	15.88	23.99	11.06	11.49

Non-detects and LOD indicated by minus sign. Values in italics indicate that the values are not within the ion ratio range for QA/QC. (f) = Farmed; (w) = wild; S. uk = Stirling unknown; S.L.A = Stirling Loch A (f); S.L.B = Stirling Loch B (f).

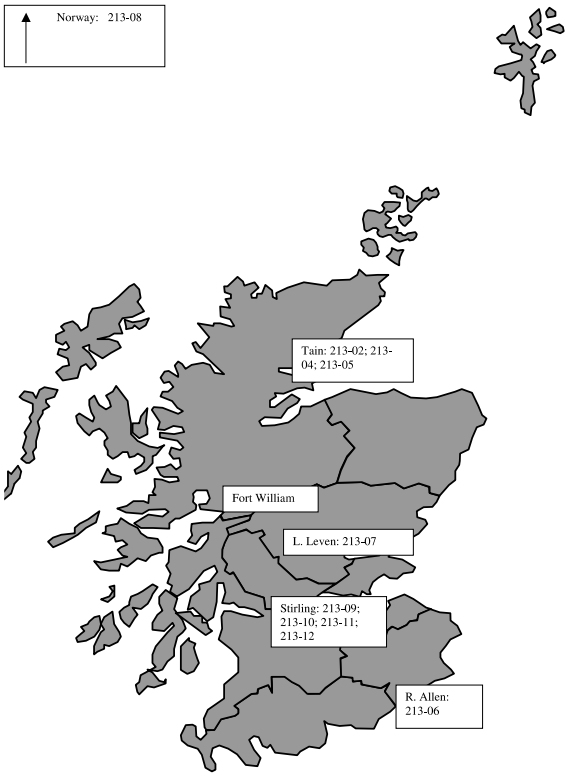

Fig. 1. Map of Scotland showing the geographical spread of sample sites.

Fig. 1 indicates the spread of the sample sites.

2.2. Sample preparation

The samples were thawed, filleted, skinned and the epaxial muscle homogenised 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.

Two samples consisted of homogenised samples of two fish each (from the same source and of the same age and size) to ensure similar sample quantities of tissue. One set of the frozen samples remained at the laboratory for subsequent analysis. The other set of samples were packed in dry ice and taken directly by courier to the US EPA/OPP laboratory the following day, where they were inspected, logged in and stored at −60 °C prior to analysis.

3. Analytical procedures

One hundred grams of homogenised muscle tissue were extracted using a mixture of hexane/methylene chloride/2-isopropanol to obtain the lipid and an aliquot representing a 7–8 g subsample (when possible) taken for analysis. A separate aliquot was also removed to determine the lipid percentage. The crude lipid extract was fortified with ^{13}C-labelled analogues of the various 2,3,7,8-Cl-substituted congeners and selected coplanar PCBs. The crude extract was cleaned up using acid/base-modified silica gel, alumina, and graphitised carbon column chromatography. The eluent was then concentrated, fortified with ^{13}C-labelled internal standards, and analysed by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS). The samples were analysed for the 17 PCDDs/PCDFs which have toxic equivalency to 2,3,7,8-TCDD and the dioxin-like co-planar PCBs 77, 105, 118, 126, 156, 157, and 169. All analyses were performed on a Kratos Concept using isotope dilution in mass drift correction mode. The sample preparation procedures, analytical techniques, and quality control strategies described in this paper are defined in US EPA's Method 1613 and Ferrario et al. (1996, 1997).

4. Results

The concentrations and toxic equivalent contributions (TEQ) from the analyses of the fish tissue for the 2,3,7,8-Cl-substituted dioxins and furans and selected coplanar PCBs are listed in Table 1 on a lipid-adjusted basis together with summaries of the toxic equivalents (TEQ) results for each sample together with weight, age and lipid content data. All values were adjusted to the lipid content of the sample by dividing the whole weight concentration by the percentage lipid in each sample. The percentage lipid was determined according to Method 1613. The rounded result for each congener was multiplied by the appropriate international toxic equivalent factor (I-TEF) (Van den Berg et al., 1998) and summed (TEQ) for all 17 congeners. In cases where congeners were reported as non-detects, lower-bound TEQs were calculated by treating the result as if zero and upper-bound TEQs by treating the result as if present at half of the limit of detection (LOD). Quality control (QC) samples were included in batches of samples analysed. Data were rounded to three decimal places for all analytes and are reported on a lipid-adjusted basis. The whole weight detection limits may be calculated by dividing the detection limit by the appropriate weight given in Table 1.

TEQs were also calculated for non-ortho- and ortho-PCBs using internationally agreed TEF values by following the same procedures as above. All PCBs analysed for were detected. The summed concentration of selected PCBs is the sum of concentrations of the congeners measured and is an underestimate of the total PCB concentration as other isomers were present which were not measured.

The concentration of lipid found in the samples ranged from 3.43% to 14.7%. The immature fish are fed lower levels of oils, and tend to store lower levels of lipid in the flesh compared to mature fish and this is reflected in the lipid content of the samples, with one unusual exception (Sample 213-10). Whether this fish had a similar dietary lipid intake to the other fish sampled is not known, but it has been observed that there can be marked variation in flesh lipid content within fish fed the same dietary oil level (the percentage of lipid in flesh of fish fed 28% oil varied from 3% to 17%) such that certain individual fish utilise high-energy diets but deposit little lipid in their flesh while others tend toward greater adiposity (Bell, 1998).

Table 2 compares age-specific mean lipid-adjusted PCDD/F and selected coplanar PCB TEFs with the means of all samples in this study and the MAFF 1999 report (Parsley et al., 1999).

The data were disaggregated by both age and whether the fish was farmed or wild. Significant differences were observed between the 2+ year old fish and 1 year olds, shown in Table 2, with no statistically significant difference between the 3 and 2 year old fish (not shown). There was also no statistically significant difference between the farmed and wild fish (or farm escapees).

All fish contained detectable levels of PCDD/Fs and mono-ortho- and non-ortho-PCBs. Levels varied between samples, generally increasing with age, and showing a skewed distribution for the PCDD/Fs resulting from a minority of relatively high values. The main dioxins and furans detected were 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 2,3,7,8-TCDF, 2,3,4,7,8-PeCDF and to a lesser extent 1,2,3,7,8-PeCDF. Apart from one fish (sourced via Stirling University, 213-10) which had been frozen a year previously, the highest levels of PCDDs, PCDFs and PCBs were observed in the oldest fish, 3+ years, as expected. The 2,3,7,8-TCDD concentrations ranged from 0.51 pg/g lipid-adjusted in the post-smolt fish, around a year old (Sample 213-12) to 1.09 pg/g lipid-adjusted in the most mature fish, between 4 to 5 years old (Sample 213-05), and 1.39 pg/g lipid-adjusted in Sample 213-10. This is equivalent to 0.03 pg/g wet

Table 2
Comparison of age-specific mean PCDD, PCDF and selected coplanar PCB detections with means of all samples in this study and the recent MAFF study (Parsley et al., 1999) (pg/g, ppt, lwt)

Concentration pg/g lipid	Mean of all samples: this study	SD	Mean for MAFF 99 study	SD	Mean 2 yr, $N = 5$: this study	SD	Mean 1 yr, $N = 2$ (four fish): this study	SD
Lipid (wt%)	9.63	±4.26	13.58	±3.32	9.26	±4.02	4.72	±0.95
2,3,7,8-TCDD	0.76	±0.28	0.84	±0.28	0.81	±0.35	0.56	±0.07
1,2,3,7,8-PeCDD	1.56	±0.97	1.44	±0.53	1.78	±1.34	1	±0.28
1,2,3,4,7,8-HxCDD	−0.03	±0.3	0.001	±0.21	0.16	±0.44	−0.14	±0.03
1,2,3,6,7,8-HxCDD	0.24	±0.45	0.56	±0.22	0.35	±0.61	−0.14	±0.03
1,2,3,7,8,9-HxCDD	−0.11	±0.04	−0.04	±0.21	−0.10	±0.05	−0.14	±0.03
1,2,3,4,6,7,8-HpCDD	−0.11	±0.04	−0.04	±0.4	−0.10	±0.05	−0.14	±0.03
OCDD	0.85	±0.29	0.05	±0.23	0.84	±0.35	1.11	±0.24
2,3,7,8-TCDF	19.57	±11.09	13.38	±4.75	21.39	±15.37	14.04	±1.58
1,2,3,7,8-PeCDF	1.83	±1.44	1.53	±0.58	2.12	±1.99	1.17	±0.25
2,3,4,7,8-PeCDF	6.22	±3.41	4.72	±1.54	6.73	±4.15	3.88	±0.18
1,2,3,4,7,8-HxCDF	0.009	±0.25	0.21	±0.17	0.07	±0.34	−0.14	±0.03
1,2,3,6,7,8-HxCDF	0.04	±0.28	0.21	±0.16	0.13	±0.37	−0.14	±0.03
2,3,4,6,7,8-HxCDF	0.22	±0.33	−0.13	±0.09	0.30	±0.41	0.06	±0.25
1,2,3,7,8,9-HxCDF	−0.11	±0.04	0.29	±0.22	−0.10	±0.05	−0.14	±0.03
1,2,3,4,6,7,8-HpCDF	−0.11	±0.04	−0.08	±0.19	−0.10	±0.05	−0.14	±0.03
1,2,3,4,7,8,9-HpCDF	−0.11	±0.04	−0.25	±0.23	−0.10	±0.05	−0.14	±0.03
OCDF	0.85	±0.29	−0.8	±0.81	0.84	±0.35	1.11	±0.24
PCB 77	388.4	±165.86	424.15	±160.63	381.84	±180.54	480.5	±169.99
PCB 105	6145.5	±1468.09	6265.83	±1792.87	6426.18	±1905.19	4886.65	±286.59
PCB 118	21 342.66	±5164.36	19 011.67	±5758.08	22 615.18	±0.68	16 764.8	±548.15
PCB 126	123.4	±44.02	137.63	±46.34	1256.62	±6815.4	82.65	±2.05
PCB 156	1923.76	±529.41	1841.67	±605.82	1985	±43.04	1314.4	±43.27
PCB 157	537.4	±141.80	550	±203.56	549.24	±564.24	378.2	±28
PCB 169	20.64	±5.89	32.12	±11.02	21.46	±153.65	13.75	±1.91
Sum of seven PCB TEFs	16.3		17.85		16.71		11.28	

For all means, ND = 0.5 LOD.

weight in the post-smolt to 0.15 pg/g in the most mature fish, and 0.05 pg/g in Sample 213-10. This fish had the lowest lipid content of all the samples including the post-smolts (see Table 1). 1,2,3,7,8-PeCDD concentrations ranged from 0.8 pg/g lipid adjusted to 4.16 pg/g lipid-adjusted in the Sample 213-10 fish.

All the selected coplanar PCBs were detected, and they were quantitatively the dominant contaminants. The consistent congener patterns were markedly higher than one would expect from the range of sources of the samples, unless they were fed on similarly sourced feeds. The highest PCB concentration detected in all samples was PCB 118 with a highest lipid-adjusted value of 31 900 pg/g (1093 pg/g wet weight: Sample 213-10) and lowest lipid-adjusted value of 14 800 pg/g (1380 pg/g wet weight: Sample 213-07).

The highest total lipid-adjusted PCB concentration was 45 100 pg/g (Sample 213-10), but the lipid-adjusted TEQ contribution for this fish was 24.0 pg/g, lower than the lipid-adjusted TEQ contribution for one of the largest fish (Sample 213-05) with a value of 24.8 pg/g. On a wet weight basis the largest fish PCB TEQ remained high at 3.40 pg/g, whilst the wet weight PCB TEQ value for Sample 213-10 fell to 0.82 pg/g due to the lower lipid content in the fish tissue.

5. Discussion

The results from this study are comparable and of a similar order of magnitude to the values reported in the recent MAFF study (Parsley et al., 1999), see Table 2. The results presented here indicate a contamination problem, especially in the case of the selected PCBs. Comparisons with background data from the MAFF study (using the pre-1998 TEF values, Kutz et al., 1990) suggest that, for wild species such as cod, the concentrations on a lipid-adjusted basis are similar. However due to the higher fat content of salmon the concentrations reported here are comparatively higher on a whole weight basis. Species with a higher fat content in the body tissues, such as herring, are reported to have consistently higher concentrations both on a lipid-adjusted (mean 83 pg/g WHO-TEQ fat) and whole weight basis, but mackerel appears to be an exception (mean 17 pg/g WHO-TEQ fat). Flat fish with a low fat content, such as plaice, approach the concentrations seen in herring on a lipid-adjusted basis (mean 67 pg/g WHO-TEQ fat). TEQ levels reported for UK farmed trout are in very good agreement with the salmon concentrations on a lipid-adjusted basis (Ministry of Agriculture, Fisheries and Food/Department of Health, 1998).

More detailed comparison with other existing data sets is confounded by the fact that many previous studies quote only means and ranges, or they use a different TEF system or the WHO-TEF system prior to 1998.

Thus direct comparison with Norwegian data for Atlantic salmon was not possible as the TEQ values reported in the literature use the Nordic TEFs, and not the revised WHO-TEFs (Vuorinen et al., 1997; Becher et al., 1998). As the values for the individual congeners are not given, a corrected TEQ value cannot be recalculated, as undertaken in this report with the UK MAFF study. Table 3 provides comparative data for PCDD/Fs and coplanar PCBs in salmon and herring fish from studies where individual congener levels were reported.

Both this study and the MAFF study report surprisingly high detections of PCB congeners. From the literature one might speculate that the source of contamination was from the herring- and fish-oil-based aquaculture feed consumed by the salmon (Bergqvist et al., 1990). A fish oil study found relatively high levels of PCBs 52, 101, 118, 138, 149, 153, 170, 180 in both the herring oil sample from Germany and salmon oil sample, e.g. PCB 118 detection of 60 pg/g lwt in salmon oil and 158 pg/g lwt in herring oil (Jacobs et al., 1998). (The coplanar PCBs, TCDDs and TCDFs were not analysed in this fish oil study.) Preliminary data on the analyses of salmon aquaculture feeds and fish oil components of the feeds for organochlorine pesticides and selected PCBs indicate that this is a probable route of contamination (Jacobs and Covaci, unpublished data).

Previous reports have detected significant levels of PCDDs, PCDFs and PCBs in fatty fish such as herring and salmon, particularly from the Baltic Sea, see Table 3 (Rappe et al., 1989; Bergqvist et al., 1990; Svensson et al., 1991; De Boer et al., 1992; Liem and Theelen, 1997; Vuorinen et al., 1997; Atuma et al., 1998).

The potential contribution to the human diet of PCBs, PCDDs and PCDFs from Scottish salmon will vary according to the age of the fish, whether the individual fish had a predisposition to adiposity or not, and the frequency of consumption, portion size, cooking practises (it is more usual to retain the oils when cooking salmon, for their nutritional fatty acid benefits), and age of the consumer. The UK Ministry of Agriculture, Fisheries and Food (1999), using pre-1998 WHO-TEF values, give an estimate of upper-bound dietary intake for adults of PCDD/Fs and PCBs from salmon of 2.3 pg TEQ/kg bw/day. Upper-bound adult dietary exposure for high-level consumers is 5.6 pg TEQ/kg bw/day; the average consumer intake is estimated at 2.6 pg TEQ/kg bw/day. However young children are particularly susceptible to higher intakes as they have a more limited diet than older children and adults, together with a low body weight. This leads to higher consumption:body-weight ratio than that of the average consumer. Currently the World Health Organisation recommends a tolerable daily intake (TDI) of 1–4 pg TEQ/kg bw/day (van Leeuwen and Younes, 1998) and the European Commission Scientific Committee on Food recommend

Table 3
Examples of congener-specific concentrations of PCDDs, PCDFs and coplanar PCBs in salmon and herring from other studies (pg/g wet weight, wwt) unless otherwise indicated

Fish	Salmon	Farmed salmon	Farmed salmon	Herring	Herring	Herring	Herring	Wild salmon[a]	Herring[b]	Fatty seafish[a]
Sample size	$N = 10$							$N = 8$	$N = 10$	$N = 2$
Region	Scotland, Norway	Gulf of Bothnia	Gulf of Bothnia	Väderöarna	Landsort	Skagerack	Central North Sea	Swedish Baltic	North Sea	Netherlands
Reference	This study[c]	Rappe et al. (1989)	Rappe et al. (1989)	Rappe et al. (1989)	Rappe et al. (1989)	De Boer et al. (1992)	De Boer et al. (1992)	Atuma et al. (1998)	Parsley et al. (1999)	Liem and Theelen (1997)[d]
Congener										
2,3,7,8-TCDD	0.072	0.3	0.2	<0.1	<0.1				0.2	0.68
1,2,3,7,8-PeCDD	0.14	1.1	0.7	0.6	2.8				0.83	1.6
1,2,3,4,7,8-HxCDD	0.011	0.2	<0.1	<0.2	0.3				0.13	0.25
1,2,3,6,7,8-HxCDD	0.027	0.4	0.7	<0.2	2.4				0.38	0.86
1,2,3,7,8,9-HxCDD	0.0077	<0.1	<0.1	<0.2	<0.2				0.14	0.18
1,2,3,4,6,7,8-HpCDD	0.0077	1.2	0.5	<0.2	0.6				0.37	0.63
OCDD	0.07	1.9	0.8	1.1	0.7				4.6	6.2
2,3,7,8-TCDF	1.76	9.0	7.8	1.7	5.3				4.9	9.7
1,2,3,7,8-PeCDF	0.15	2.1	1.7	0.4	2.5				1.1	1.5
2,3,4,7,8-PeCDF	0.58	3.6	2.8	3.0	19.0				2.2	8.0
1,2,3,4,7,8-HxCDF	0.01	0.2	<0.1	0.2	0.7				0.21	0.37
1,2,3,6,7,8-HxCDF	0.01	0.1	<0.1	0.1	0.8				0.21	0.41
2,3,4,6,7,8-HxCDF	0.019	0.7	0.2	<0.2	0.8				ND	0.63
1,2,3,7,8,9-HxCDF	0.0077	NA	NA	NA	NA				0.3	ND
1,2,3,4,6,7,8-HpCDF	0.0077	1.6	<0.7	<0.2	1.2				0.12	0.19
1,2,3,4,7,8,9-HpCDF	0.0077	NA	NA	NA	NA				ND	ND
OCDF	0.077	<0.5	<0.3	<0.2	0.3				ND	ND
PCB 77	33.77					41	20	0.87–2.57	1093.9	260
PCB 105	12.29					920	NA	45.3–82.6	20.26	NA
PCB 118	2.05					28 100	NA	134–249	60.26	NA
PCB 126	587.38					11	8.9	0.28–4.01	429.33	100
PCB 156	2035.65					NA	NA	15.4–38.1	5.23	NA
PCB 157	193.34					NA	NA	NA	1.76	NA
PCB 169	53.50					2.5	1.8	0.14–0.53	131.13	28

ND = not detected, NA = not analysed.
[a] In pg/g lipid.
[b] In ng/g lipid.
[c] ND = 0.5 LOD.
[d] The NL study uses the earlier TEF scale where the differences are: 1,2,3,7,8-PeCDD 0.5, OCDD/F 0.001; ND = 0.03 LOD.

a tolerable weekly intake (TWI) of 14 pg WHO-TEQ/kg bw (Scientific Committee on Food, 2001), a revision of the temporary TWI (tTWI) of 7 pg WHO-TEQ/kg bw recommended in November 2000 (Scientific Committee on Food, 2000). This recommendation has not been adopted universally. The UK Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT) recommends a TDI of 10 pg/kg bw/day for 2,3,7,8-TCDD, based on lifetime averages. This is equivalent to an intake of 600 pg TEQ/day for a 60 kg person, and is based upon an expert group recommendation convened by WHO/EURO in 1991, rather than the more recent WHO recommendation; however COT recommendations are under revision. The COT has acknowledged that "for people in the UK and many other countries, the daily intake of dioxins could be close to or even above the guideline value" (COT, 2000; Food Standards Agency, 2000). One gram of Sample 213-05, which was both the largest fish and the fish that contained the greatest concentrations, would give a contribution of 4.98 pg of the congeners analysed towards the total TEQ/day. The possible contribution to dietary intakes of organochlorines from farmed fish could be significant for high consumers, and may be congener specific, similar to the species being consumed, as recently observed in a Finnish study (Kiviranta et al., 2000).

Whilst diets based on marine fish oils are currently favoured by the aquaculture industry, it is likely that these oils are contributing greatly to the contamination of farmed salmon by PCDDs, PCDFs and PCBs. There is experimental evidence that suggests aquaculture diets utilising 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 parr, could be more beneficial in accommodating successful seawater adaptation than diets based on marine fish oils (Bell, 1998). Vegetable-oil-based diets could also facilitate the requirement for high-energy aquaculture feeds on an economically competitive basis, whilst reducing the problems of organochlorine contamination. Contamination problems do exist with vegetable oils, but to a far lesser degree than fish oils (Jacobs et al., 1998). This would reduce the reliance on fish meal and oils from non-sustainable natural resources and accommodate the salmon farming industry expansion. It would also demonstrate sensitivity to public confidence after the recent PCB and dioxin food crises, producing a nutritious food using environmentally friendly methods, delivering easily utilised essential $n-3$ fatty acids to the human consumer.

Acknowledgements

This paper has not been subject to the USEPA publication review process and, therefore, does not necessarily reflect the views of the Agency and no official endorsement should be inferred. Nor does the mention of trade names of commercial products constitute endorsement or recommendation of use.

References

Atuma, S.S., Aune, M., Bergh, A., Wicklund-Glynn, A., Darnelund, P.O., Larsson, L., Olsson, M., Sandström, O., 1998. Polychlorinated biphenyls in salmon (*Salmo salar*) from the Swedish east coast. Organohalogen Compounds 39, 153–156.

Becher, G., Jensen, A.J., Zubchenko, A., Haug, L.S., Hvidsen, N.A., Johnsen, B.O., Kashin, E., 1998. Dioxins and non-ortho PCBs in Atlantic salmon, *Salmo salar*, from major Norwegian and Russian salmon rivers. Organohalogen Compounds 39, 427–430.

Bell, J.G., 1998. Current aspects of lipid nutrition in fish farming. In: Black, K.D., Pickering, A.D. (Eds.), Biology of Farmed Fish. Sheffield Academic Press, UK, pp. 114–145.

Bell, J.G., McEvoy, J., Webster, J.L., McGhee, F., Millar, R.M., Sargent, J.R., 1998. Flesh lipid and carotenoid composition of Scottish farmed Atlantic salmon (*Salmo salar*). Journal of Agriculture and Food Chemistry 46, 119–127.

Bergqvist, P.-A., Hjelt, M., Rappe, C., 1990. Bioaccumulation of PCDDs and PCDFs in farmed salmon with natural herring oil added to the feed. In: Wahlberg, C., Minderhovd, A., Angeletti, G. (Eds.), Transport of Organic Micropollutants in Estuaries, Marine and Brackish Waters. Proceedings of a Workshop "Organic Micropollutants in the Aquatic Environment", October 4–5, 1990. Water Pollution Research Report, 24. Commission of the European Communities, Directorate-General for Science, Research and Development, Brussels, pp. 111–119.

BOCM Pauls Fish Feed Group, 1998. Fulmar Salmon Feed. Rising To the Challenge. Renfrew Mill, Renfrew, Scotland.

De Boer, J., Stronck, C.J.N., van der Valk, F., Wester, P.G., Daudt, M.J.M., 1992. Method for the analysis of non-ortho substituted chlorobiphenyls in fish and marine mammals. Chemosphere 25 (7), 1277–1283.

FAO Fishery Information, Data and Statistics Unit, 1998. Aquaculture production statistics 1987–1996. Fisheries Circular No. 815, Rev. 10. Rome, FAO.

FAO Globefish, Fisheries Industries Division, 2000. Available from http://www.globefish.org (cited September 2000).

Ferrario, J., Byrne, C., Dupuy, A., 1997. Background contamination by coplanar PCBs in trace level high resolution gas chromatography/high resolution mass spectrometry (HRMS/MRGC) analytical procedures. Chemosphere 34 (11), 2351–2466.

Ferrario, J., Byrne, C., McDaniel, D., Dupuy, A., Harless, R., 1996. Determination of 2,3,7,8-chlorine-substituted dibenzo-*p*-dioxins and -furans at the parts per trillion level in United States beef fat using high-resolution gas chromatography/high resolution mass spectrometry. Analytical Chemistry 68 (4), 647–652.

Food Standards Agency, 2000. Food Standards Agency UK – dioxins and PCBs in the UK diet: 1997 total diet study

samples. Food Surveillance Information Sheet 4/00, FSA, London, UK.

Jacobs, M.N., Johnston, P.A., Santillo, D., Wyatt, C.L., 1997. Organo-chlorine pesticide and PCB residues in pharmaceutical and industrial grade fish oils. International Journal of Environment and Pollution 8, 74–93.

Jacobs, M.N., Johnston, P.A., Santillo, D., Wyatt, C.L., 1998. Organo-chlorine residues in fish oil dietary supplements: comparison with industrial grade oils. Chemosphere 37, 1709–1731.

Kiviranta, H., Vartianinen, T., Verta, M., Tuomisto, J.T., Tuomisto, J., 2000. High fish specific dioxin concentrations in Finland. Lancet 355, 1883–1885.

Kutz, F.W., Barnes, D.G., Bottimore, D.P., Greim, H., Bretthauer, E.W., 1990. The international toxicity equivalency factor (I-TEF) method of risk assessment of complex mixtures of dioxins and related compounds. Chemosphere 20, 751–757.

Liem, A.K.D., Theelen, R.M.C., 1997. Dioxins: chemical analysis, exposure and risk assessment. Ph.D. Thesis, National Institute of Public Health and the Environment, Bilthoven, The Netherlands.

Ministry of Agriculture, Fisheries and Food, 1995. Dioxins in food – UK dietary intakes. Food Safety Information Sheet 71, MAFF, London, UK.

Ministry of Agriculture, Fisheries and Food, 1996. Polychlorinated biphenyls in food – UK dietary intakes. Food Surveillance Information Sheet 89, MAFF, London. UK.

Ministry of Agriculture, Fisheries and Food, 1997. Dioxins and polychlorinated biphenyls in fish oil dietary supplements and licensed medicines. Food Surveillance Information Sheet 106, MAFF, London, UK.

Ministry of Agriculture, Fisheries and Food/Department of Health, 1998. Dioxins and polychlorinated biphenyls in farmed trout. Food Safety Information Bulletin 94, MAFF, London, UK.

Ministry of Agriculture, Fisheries and Food, 1999. Dioxins and polychlorinated biphenyls in marine fish, fish and fish products. Food Safety Information Sheet 184, MAFF, London, UK.

North Sea Task Force, 1993. North Sea Quality Status Report, 1993. Oslo and Paris Commissions/International Council for Exploration of the Sea, London.

Parsley, K., Wright, C., Thorpe, S.A., 1999. PCDDs, PCDFs and PCBs in marine fish, salmon and fish fingers. Report Number FD 97/66, CSL Food Science Laboratory, MAFF, London, UK.

Rappe, C., Bergqvist, P.-A., Kjeller, L.O., 1989. Levels, trends and patterns of PCDDs and PCDFs in Scandinavian environmental samples. Chemosphere 18 (1–6), 651–658.

Royce, W.F., 1996. Introduction to the Practice of Fishery Science. Academic Press, London.

Scientific Committee on Food, 2000. Opinion of the SCF on the risk assessment of dioxins and dioxin like PCBs in food. Adopted November 22, 2000. European Commission Health and Consumer Protection Directorate-General. Directorate C – Scientific Health Opinions Unit C3. Available from http://europa.eu.int/comm/food/fs/sc/scf/index_en.html.

Scientific Committee on Food, 2001. Opinion of the SCF on the risk assessment of dioxins and dioxin like PCBs in food. Update based on new scientific information available since the adoption of the SCF opinion of November 22, 2000. Adopted May 30, 2001. European Commission Health and Consumer Protection Directorate-General. Directorate C – Scientific Health Opinions Unit C2. CS/CNTM/DIOXIN20 final. Available from http://europa.eu.int/comm/food/fs/sc/scf/index_en.html.

Svensson, B.-G., Nilsson, A., Hansson, M., Rappe, C., Åkesson, B., Skerfving, S., 1991. Exposure to dioxins and dibenzofurans through the consumption of fish. New England Journal of Medicine 324, 8–12.

Trouw Aquaculture, (no date). Fish Feed. Feeding for Profit. Trouw Aquaculture, Wincham, Northwich, Cheshire, UK.

UK Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment, 2000. Statement on the dietary exposure to dioxins and dioxin like PCBs. August 2000 COT Statement 2000/03.

Van den Berg, M., Birnbaum, L., Bosveld, A.T.C., Brünstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., van Leeuwen, F.X.R., Liem, A.K.D., Nolte, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillit, D., Tysklind, M., Younes, M., Wærn, F., Zacharewski, T., 1998. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. Environmental Health Perspectives 106 (12), 775–792.

van Leeuwen, F.X.R., Younes, M., 1998. WHO revises the tolerable daily intake (TDI) for dioxins. Organohalogen Compounds 38, 295–298.

Veterinary Medicines Directorate, 1998. The Veterinary Medicines Directorate Annual Report on Surveillance for Veterinary Residues in 1997. MAFF, London, UK.

Vuorinen, P.J., Vartiainen, T., Keinänen, M., 1997. Polychlorinated dibenzo-*p*-dioxins, dibenzofurans and biphenyls in salmon (*Salmo salar*) from the southern Baltic sea and the arctic Tenjoki river. In: Vartiainen, T., Komulainen, H. (Eds.), 7th Nordic Symposium on Organic Pollutants. Natural and Environmental Sciences, 68. Kuopio University Publications, Finland.

World Health Organisation, 1999. Food safety issues associated with products from aquaculture. Report of a Joint FAO/NACA/WHO Study Group. WHO Technical Report Series 883. World Health Organisation, Geneva.

Wright, C., Startin, J.R., 1995. Concentrations of PCDD's and PCDF's in total diet study samples from 1982 to 1992. Report No. FD94/200. UK Ministry of Agriculture, Fisheries and Food, London.