Pergamon

Chemosphere, Vol. 37, Nos 9-12, pp. 1709-1721, 1998

0045-6535/98 $19.00+0.00

PII: S0045-6535(98)00236-7

ORGANOCHLORINE RESIDUES IN FISH OIL DIETARY SUPPLEMENTS: COMPARISON WITH INDUSTRIAL GRADE OILS.

M. N. Jacobs[a], D. Santillo[a], P. A. Johnston[a], C.L.Wyatt[b] & M.C. French[b]

[a]Greenpeace Research Laboratories, Department of Biology, University of Exeter, Prince of Wales Road, Exeter EX4 4PS. [b]Institute of Terrestrial Ecology, Monks Wood, Abbots Ripton, Huntingdon PE17 2LS.

ABSTRACT

The market for fish oils as dietary supplements is of global importance. Although it is widely recognised that lipophilic organic chemicals, particularly organochlorines, can accumulate in fish oils, dietary supplements are not routinely considered when estimating average daily intakes for these contaminants. This paper reports levels of organochlorine residues in 44 fish oils, collected from 15 countries between 1994 and 1995, including 38 purchased over the counter as dietary supplements. Despite controls on the use of persistent organochlorine substances, appreciable quantities are found in oils sold as dietary supplements. Levels are discussed in relation to the significance of fish oil dietary supplements as contributors to daily intake of PCBs and pesticide residues.

Keywords: Fish-oils, PCBs, pesticides, organochlorines, diet

INTRODUCTION

Fish oils account for approximately 2% of the total world production of oils and fats[1]. They are widely used in foodstuffs, primarily in baking, but also in margerines, ice creams, animal foods, etc. A number of non-food applications exist, including manufacture of cosmetics, detergents and paints[2]. The manufacture of dietary supplements also accounts for a high demand; pharmaceutical and food grade nutritional supplements commanded a market of 50 million pounds sterling in 1992 in the UK [3]. Although cod liver oil is among the most commonly used, whole fish oils and other lipid concentrates are also processed for dietary supplements.

Procedures employed to extract and refine the oils differ among manufacturers and will depend partly on feedstock, but are essentially designed to remove pigments, odours and free fatty acids from the raw oil.

Such extraction and cleanup procedures may include direct steaming, cold extraction, flotation, solvent extraction or addition of surfaces such as clays or activated carbon [2]. Extraction of oils, particularly of pharmaceutical grade, is becoming increasingly automated in order to minimise introduction of contaminants [4].

Although the extraction and refining techniques employed may have some impact on the levels of organochlorine contaminants carried through from the raw fish oil, significant quantities may remain in refined oil. The trend towards extraction and refining at lower temperatures to maintain quality [4,5] may lead to increased carry over in the future. Residues of organochlorine pesticides and PCBs have been found in the few documented analyses of fish oils. For example, analysis of pharmaceutical grade cod liver oils sourced from the Southern Baltic between 1981 and 1989[6] showed that, while DDT and DDE levels had generally declined, PCB levels remained steady in recent times [7]. Furthermore, the appearance of such residues in oils from the South Iceland Shelf [8] indicates that these compounds are increasingly reaching remote areas, probably through atmospheric cycling. Although peak inputs for the majority of these chemicals occurred in the 1960's [9], releases to the environment continue. Levels of contaminants in unrefined fish oils could prove a useful indicator of long term global contaminant trends.

Few studies of contaminants in fish oils have included analysis of refined consumer products. Although some manufacturers do monitor contaminant levels, along with other oil quality parameters, there is no legislative requirement and the data are not in the public domain. To our knowledge, this study represents the first independent assessment of levels of organochlorine contamination in fish oils available from retail outlets. The results from analysis of 22 oils (including 17 dietary supplements), carried out in 1994, are currently in press [10]. The current paper builds on this initial study, including data from a further 23 oils obtained in 1995, again primarily through retail outlets. Levels of organochlorine contaminants are compared with those obtained from fish oils reported in other studies, and the significance of those levels discussed in the context of recommended daily intakes of fish oils as dietary supplements.

MATERIALS AND METHODS

Organochlorine pesticide and PCB residues were quantified as follows. 1ml of each fish oil sample was shaken with 5ml of HPLC grade hexane (Rathburn Chemicals) to effect complete dissolution. The lipid content was destroyed by addition of 2ml of concentrated Aristar grade sulphuric acid and the mixture allowed to separate. The hexane extract was cleaned on a column of active, neutral, Brockmann grade 1

aluminium oxide (BDH, Poole), eluting with hexane to give a final eluate volume of 5ml. 1ml eluate was transferred to a glass vial, adding 0.04 $\mu g\ ml^{-1}$ 2,6-dichlorobenzonitrile as an internal standard.

Chromatographic analysis was carried out using a Varian 3400 gas chromatograph (GC) equipped with electron capture detection (ECD). 2µl of sample was injected in splitless mode (injection port 200°C), on to two 30m DB210 columns (J&W Scientific) connected in series, using hydrogen as a carrier (51 cm s^{-1}). An initial oven temperature of 90°C was held for 1 minute, after which the oven was ramped to 185°C (2.5 C min^{-1}) and held for 20 minutes. This was then increased to 210°C (5 C min^{-1}), and held for twelve minutes before ramping to 240°C (40 C min^{-1}). Further details of the analytical methodology are given by Jacobs *et al.* [10].

Identification and quantification of analytes was by comparison with prepared calibration standards, checking identities periodically gas chromatography/mass spectrometry. Σ-PCB values were derived from comparison with standard Aroclor-1254. Recoveries of analytes from the alumina columns exceeded 96% in every case. Recoveries from spiked samples ranged from 85.9+/-1.7% to 120.7+/-10.3%, but were judged to be acceptable in all cases.

Two cod liver oil standard reference materials (SRM) were run alongside samples (US National Institute of Standards and Technology, Standard Reference Material 1588 [for pesticide residues]; EC Community Bureau of Reference Standard CRM 349 [for 6 PCB congeners]. With the exception of p,p'-DDT, precision and accuracy were judged to be acceptable. Determinations of p,p'-DDT showed a consistent bias of 40-50% below the declared value in the NIST SRM, probably resulting from degradation on the GC column. Dieldrin is not reported since this contaminant did not survive the acid clean-up process.

Samples were collected during 1994 and 1995 and were analysed in two batches. Data for the first batch were reported in $\mu g\ l^{-1}$. In order to facilitate comparison with previously published data expressed in $\mu g\ kg^{-1}$, densities of oils were determined gravimetrically. These were found to lie between 0.911 and 0.937 g ml^{-1}. Data for the second batch were reported in ug kg^{-1}, along with densities for individual oils. These have been converted to ug l^{-1} for consistency.

RESULTS

Results for chlorinated pesticides and total PCBs are shown in Table 1, according to country of purchase or origin. Note that these are not necessarily the countries from which the oil was initially sourced. Congener

Sample	Origin	Grade	Type	HCB	a-HCH	g-HCH	p,p'-DDE	p,p'-TDE	p,p'-DDT	ΣDDT	ΣPCBs
1	Australia	F	FO	-	-	-	10	-	-	10	-
2	Australia	F	CLE	3	4	-	9	-	-	9	10
3	Australia	F	CL	19	13	-	91	122	-	213	369
4	Australia	F	CL	14	18	5	91	155	-	246	159
5	Australia	F	CL	-	-	-	4	-	-	4	-
6	Austria	P	FO	-	-	-	39	-	-	39	17
7	Austria	P	FO	-	-	-	5	-	-	5	-
8	Austria	P	FO	-	-	-	-	-	-	-	-
9	Austria	P	FO	-	-	-	-	-	-	-	6
10	Belgium	F	FO	7	23	4	13	-	-	13	22
11	Belgium	F	FO	15	10	4	97	332	492	921	805
12	Belgium	F	FO	5	-	-	28	-	-	28	40
13	Belgium	F	FO	3	-	-	26	-	-	-	228
14	Brazil	P	FO	19	7	-	141	182	-	323	346
15	Canada	F	CL	-	-	-	33	68	-	101	109
16	Canada	F	CL	-	-	-	18	-	-	18	43
17	France	F	FO	-	-	4	-	-	-	-	-
18	Germany	U	HO	35	63	13	106	117	-	223	525
19	Japan	F	FO	-	6	-	62	-	33	95	313
20	Japan	F	FO	-	-	-	2	-	-	2	-
21	NL	F	FO	4	-	-	15	-	-	15	73
22	NZ	F	CL	45	28	11	254	357	133	744	619
23	NZ	F	CL	-	-	-	14	-	-	14	-
24	Norway	P	FO	-	-	-	45	29	74	148	570
25	Norway	P	FO	-	-	-	30	19	14	63	440
26	Norway	P	CL	-	-	-	2	-	-	2	-
27	Spain	F	CL	18	93	-	59	10	-	69	261
28	Spain	F	CL	-	-	-	22	6	12	40	212
29	UK	F	CL	21	9	3	56	44	19	119	990
30	UK	P	SO	46	8	9	87	53	-	140	1132
31	UK	F	CL	-	-	-	28	27	7	62	428
32	UK	P	MLC	-	-	-	4	-	-	4	10
33	UK	F	HL	-	-	-	3	-	-	3	37
34	UK	F	CL	10	12	3	60	48	29	137	1055
35	UK	P	CL	-	7	-	6	-	-	6	14
36	UK	F	CL	10	12	-	60	47	31	138	1050
37	UK	P	MLC	-	-	-	-	-	-	-	-
38	UK	P	CFO	-	-	-	35	23	-	58	915
39	UK	F	LO	-	-	13	-	-	-	-	-
40	USA	F	CL	39	7	6	161	219	-	380	717
41	Iceland	U	FO	14	36	3	11	-	7	18	366
42	Germany	U	?	21	33	20	56	51	30	137	939
43	Germany	U	RF	24	18	-	33	37	29	99	1106
44	Germany	U	SE	8	14	11	14	23	-	37	463
45	UK	V	CL	10	26	-	9	-	-	9	183
LIMIT OF DETECTION				3	3	2	1	4	5	N/A	5

TABLE 1: Concentrations of organochlorine contaminants (ug l^{-1}) in fish-oils obtained from various sources. P: pharmaceutical grade; F: food grade; U: oil for industrial applications V: veterinary grade. (-): not detected. N/A: detection limit not applicable (ΣDDT calculated as sum of DDT, DDE & TDE); FO: Fish oil; CL: Cod liver oil; CLE: cod liver oil emulsion; SE: Sandeel; RF: Redfish Oil (Probably No: 2 cod liver oil); MLC: Marine Lipid Concentrate; HL: Halibut liver oil; CFO: Fish oil concentrate; SO: Salmon oil; LO: Linseed oil. Descriptions are those given by the manufacturer/supplier.

specific PCB analyses are reported in Table 2. Mean values, standard errors and ranges of the specific analytes are shown in Table 4, along with median values and interquartile ranges.

Sample	Origin	Grade	Type	PCB congener										
				28	52	101	118	128	138	149	153	169	170	180
1	Australia	F	FO	-	-	-	-	-	-	-	-	-	-	-
2	Australia	F	CL	-	-	-	-	-	-	-	-	-	-	-
3	Australia	F	CL	-	-	23	161	-	339	-	140	-	19	24
4	Australia	F	CL	-	-	22	118	-	124	-	70	-	-	15
5	Australia	F	MLC	-	-	-	-	-	-	-	-	-	-	-
6	Austria	P	FO	-	-	-	-	-	-	-	-	-	-	-
7	Austria	P	FO	-	-	-	-	-	-	-	-	-	-	-
8	Austria	P	FO	-	-	-	-	-	-	-	-	-	-	-
9	Austria	P	FO	-	-	-	-	-	-	-	-	-	-	-
10	Belgium	F	FO	-	-	-	17	-	-	-	11	-	-	-
11	Belgium	F	FO	-	-	89	535	17	231	93	408	-	37	48
12	Belgium	F	FO	-	-	-	43	-	-	-	25	-	-	12
13	Belgium	F	FO	-	-	-	53	-	332	-	47	-	-	15
14	Brazil	P	FO	-	-	56	344	-	210	-	203	-	-	28
15	Canada	F	CL	-	-	-	35	-	108	-	56	-	-	15
16	Canada	F	CL	-	-	-	61	-	-	-	32	-	-	-
17	France	F	FO	-	-	-	-	-	-	-	-	-	-	-
18	Japan	F	FO	-	-	-	-	-	24	10	24	14	-	21
19	Japan	F	FO	-	-	-	-	-	-	-	-	-	-	-
20	Germany	U	HO	-	26	44	158	-	758	44	103	-	24	21
21	NL	F	FO	-	-	-	-	-	86	-	42	-	-	11
22	NZ	F	CL	-	-	79	339	-	575	65	216	-	-	42
23	NZ	F	CL	-	-	-	-	-	-	-	-	-	-	-
24	Norway	P	FO	-	-	15	24	9	76	16	52	-	6	22
25	Norway	P	FO	-	-	10	14	6	49	-	37	-	10	12
26	Norway	P	CL	-	-	-	-	-	6	-	-	-	-	-
27	Spain	F	CL	-	-	-	5	-	29	-	32	-	3	7
28	Spain	F	CL	-	-	-	-	-	27	-	24	-	14	11
29	UK	F	CL	-	-	29	119	11	110	18	70	-	12	13
30	UK	P	SO	-	-	156	60	-	61	51	-	-	11	44
31	UK	F	CL	-	-	-	38	17	43	-	57	-	10	31
32	UK	P	MLC	-	-	-	9	-	-	-	-	-	-	-
33	UK	F	HL	-	-	-	-	-	12	-	-	-	-	-
34	UK	F	CL	-	-	26	117	10	122	-	79	-	13	29
35	UK	P	CL	-	-	-	-	-	-	-	-	-	-	6
36	UK	F	CL	-	-	22	116	8	60	18	75	-	14	17
37	UK	P	MLC	-	-	-	-	-	-	-	-	-	-	-
38	UK	P	CFO	-	-	19	30	-	61	52	70	-	11	13
39	UK	F	LO	-	-	-	-	-	-	-	-	-	-	-
40	USA	F	CL	-	49	69	344	22	589	26	263	-	20	29
41	Iceland	U	FO	-	-	-	36	-	16	-	-	-	13	-
42	Germany	U	?	-	-	51	76	-	65	44	80	-	21	14
43	Germany	U	RF	-	-	18	79	-	32	26	-	-	31	11
44	Germany	U	SE	-	-	21	30	-	44	41	56	-	-	12
45	UK	V	CL	-	-	-	28	-	12	-	-	-	9	-
LIMIT OF DETECTION				11	18	7	5	6	4	8	6	4	3	3

TABLE 2: Concentrations of individual chlorobiphenyl congeners (ug l^{-1}) in fish oils from various sources. ICES congeners are 28, 52, 101, 118, 138, 153, 180 [11]. All abreviations as Table 1.

With the exception of sample 37, a marine lipid concentrate from the UK, and sample 8, a fish oil purchased in Austria, all oils contained detectable levels of organochlorine residues. Levels varied greatly from oil to oil, showing a skewed distribution resulting from a minority of relatively high values. The highest total PCB concentration was found in a salmon oil preparation (Sample 30) from a retail outlet in the UK (1132 ug l^{-1}).

Sample	Origin	Grade	Type	PCB congener										
				28	52	101	118	128	138	149	153	169	170	180
R1	N/A	N/K	CL	15	39	54	71	nv	94	nv	124	nv	nv	38
R2	N/A	N/K	CL	28	80	129	91	179	267	105	276	49	nv	108
R3	N/A	P	CL	10	23	45	80	nv	160	nv	120	nv	nv	50
R4	Norway	P	CL	72*	96	240	380	48	520	140+	590	nv	57	230
R5	Iceland	P	CL	-	45	88	120	38	220	64+	250	nv	19	87

TABLE 3: Concentrations of individual chlorobiphenyl congeners (ug l^{-1}) in fish oils from other studies. R1: values (ug kg^{-1}) for reference fish oil from North Sea, used in the QUASIMEME scheme[12]. R2: concentrations of CBs (ug kg^{-1}) in Canadian reference material issued in 1989 [13]. R3: mean values (ug kg^{-1}) for five samples of pharmaceutical grade oils [14]. nv: :no value given. R4: sample supplied in 1983 from the same source as sample 1; R5: cod liver oil from Iceland in 1984 [7]. * : aggregate value for CBs 28 & 31, +: aggregate value for CBs 144 & 149.

Prominent among other samples with high PCB concentrations were the cod liver oils and concentrated fish oils, whereas the limited number of marine lipid concentrates analysed showed only low levels of PCB contamination. Fish oils showed a more even spread of contaminant levels, probably as a result of their more heterogenous origin. Indeed, a fish oil, sample 11 (purchased in Belgium), yielded the highest combined DDT concentration of all samples.

In addition to the salmon oil, a sample of herring oil from Germany (sample 18, probably unrefined) showed relatively high levels of PCBs. An oil sold in the UK as halibut liver oil (sample 33) and showing only intermediate concentrations of PCBs, was actually a mixture of halibut liver oil and soya oil in unknown proportions; the levels of contaminants found cannot, therefore, be regarded as representative of halibut liver oil alone. A single sample of linseed oil included as a reference (sample 39) contained only very low levels of residues.

ANALYTE	HCB	a-HCH	g-HCH	p,p'-DDE	p,p'-TDE	p,p'-DDT	ΣDDT	ΣPCBs
RANGE	ND-46	ND-93	ND-20	ND-254	ND-357	ND-492	ND-921	ND-1132
MEAN	9.5	10.8	2.7	41.8	45.3	21.8	121.0	332.0
SE	1.9	2.7	0.6	7.7	12.6	11.5	30.7	56.5
MEDIAN	ND	ND	ND	27	ND	ND	58	198
IQ RANGE	13.0	11.5	2.5	52.0	47.0	8.5	129.5	537.5
SI	73-100	42-71	5-9	340-440	-	15-120	650-950	1.9*
Mean	87	53	6	400		76	860	Nv
SB	170-370	280-400	100-160	1500-5100	-	440-2100	3100-1200	8100-16000
Mean	280	320	140	2600		1300	6300	10

TABLE 4: Range, mean, standard error (SE), median and interquartile range (IQ RANGE) for organochlorine contaminants in fish oil (μg l^{-1}). ND denotes analyte undetected. Mean values calculated using one half of the detection limit shown in TABLE 3 where contaminant levels were below this value. n=44 except for ΣDDT where n=39 due to omission of six values below detection limit. Lower section shows comparative values in ug kg-1 for fish oils sampled between 1984 and 1989 from the Shelf of Iceland (SI) and Southern Baltic (SB) [8]. * single value. ΣDDT values not directly comparable due to summation of different isomers in each case.

Quantitatively, the dominant contaminants were the PCBs, followed by DDT and its metabolites. Notably high levels of DDT were recorded in samples 40 (USA, cod liver oil), 11 (Belgium, fish oil), 22 (New Zealand, cod liver oil) and 14 (Brazil, fish oil). Note that sample 10, submitted as a duplicate of 11, nevertheless showed much lower levels of organochlorine contamination.

Hexachlorobenzene (HCB) and hexachlorcyclohexane (HCH) isomers contributed relatively little to the overall contaminant burden, particularly in dietary supplement oils, and were absent from many samples. These residues were more abundant in some unrefined and industrial grade oils (eg. samples 18, 41, 42 and 43). Nevertheless, levels of other contaminants in the industrial and veterinary grade oils did not differ markedly from concentrations present in pharmaceutical or food grade supplements. Oils with higher PCB contents generally showed higher levels of other residues. Although the numerical correlation is poor, there is a good non-parametric association between total DDT and PCB levels (Pearsons rank coefficient 0.84, $p<0.0001$).

The PCB congener specific values obtained are of the same order as values obtained for an oil derived from a batch of North Sea fish for intercalibration purposes in 1993 (denoted R1, Table 3). Concentrations are generally an order of magnitude or more lower than those recorded for cod liver oil in 1983 from the North Sea (R4), supplied from the same manufacturer as Samples 1 & 2. Similarly, an order of magnitude difference generally exists between values reported for a 1989 Canadian Reference Material and the present samples. Organochlorine concentrations in the more contaminated cod liver oils, nonetheless, are of a comparable order of magnitude to those determined for a 1984 sample obtained from Iceland and for which a PCB value of 1.9ppm is given [7].

CBs 28 and 52 were not detected in any of the samples analysed in this study although this may be a function of relatively high detection limits for these congeners. While CBs 138 and 153 were generally dominant in the cod liver oil samples, the relative proportions do not appear to be as high as in samples R1-R5 (Table 3). In the current study, CB 118 appears to contribute a higher proportion of the congener content. Although 118 can be over estimated as a result of co-elution with 149, the two peaks were clearly resolved in the current investigation.

Recommended daily consumptions of fish oils, where specified on the packaging, are included in Table 5. These quantities have been used to estimate the contribution made by recommended doses of the dietary supplements to daily intake of PCBs and pesticide residues. Consumption of the recommended daily dose of one of the more contaminated fish oils (sample 11) could account for intakes up to 16.1 ug day^{-1} ΣPCBs and 18.4 ug day^{-1} ΣDDT.

SAMPLE	RECOMMENDED DAILY INTAKE	ΣPCB DAILY INTAKE (μg)	ΣDDT DAILY INTAKE (μg)
1	0.275-0.55g	nd	nd
2	30ml	0.30	0.27
3	4.5ml	1.66	0.96
4	2-4ml	0.32-0.64	0.49-0.98
5	3g	nd	0.013
7	1g	nd	0.006
8	0.81g	nd	nd
9	1.5-3g	0.01-0.02	nd
10	0.5-1g	0.30-0.60	0.28-0.56
13	5-20ml	0.11-0.44	0.07-0.13
14	5-20ml	4.03-16.10	4.61-18.42
16	0.5g	0.026	0.018
17	1.0g	0.25	0.028
19	0.25ml	0.08	0.02
20	0.3ml	nd	0.0005
21	10ml	0.73	0.15
22	0.275-0.55g	0.26-0.54	0.32-0.63
23	5ml	nd	nd
24	5-7ml	2.85-3.99	0.74-1.036
25	5ml	2.2	0.32
25	5ml	nd	0.01
27	0.2-1.2ml	0.05-0.31	0.01-0.08
29	1ml	0.99	0.12
30	4ml	4.5	0.56
31	0.25ml	0.12	0.02
32	1-6ml	0.01-0.06	0.004-0.024
33	0.25ml	0.009	0.0007
34	0.54ml	0.56	0.074
35	1ml	0.014	0.006
36	0.54ml	0.57	0.074
37	2-8ml	nd	nd
38	1-4ml	0.92-3.66	0.06-0.23
39	5-15ml	nd	nd
40	5ml	3.59	1.90

TABLE 5: Recommended daily intakes of fish oil according to product manufacturers, where information is available. Total PCB and DDT intake are shown as calculated from the analytical determinations presented in Table 1. nd indicates value below limit of detection for that analyte.

DISCUSSION

The data presented above extend those already published by Jacobs *et al.* [10]. Despite the inclusion of an additional 22 samples to the data set, drawn from a further 8 countries, the levels and patterns of contaminant distribution are broadly similar to those for the preliminary data set. It seems reasonable to suggest, therefore, that the range of levels reported here may be typical for fish oils available as dietary supplements around the world.

Again, none of the samples exceeds the 2.0 ppm regulatory limits specified for total PCBs in foodstuffs by the US Food and Drug Administration [15], although in 4 of the 45 samples, the 1.0 ppm limit specified in

Switzerland is exceeded. No specific legal requirements relating to PCB or pesticide residue contamination of foods apply in the UK, beyond the general requirement under the Food Safety Act (1990) that commodities should be safe for human consumption. Similarly, the Codex Alimentarius published by the United Nations World Health Organisation contains no recommendations for limits on organochlorines in fish oils, although oils derived from fish and marine mammals are a recognised commodity group [16].

Overall, the levels of organochlorine contaminants reported in this study are somewhat lower than those reported in previous studies[8,17]. This may reflect, in part, a decline in concentrations of pesticide residues in the marine environment. For example, concentrations of DDT are substantially lower than previously reported figures for the Shelf of Iceland samples taken between 1984 and 1987 (Table 4). A similar decline has been reported for organochlorine pesticides through comparison of samples analysed in 1985 and 1991 [18]. Although controls on PCBs and the organochlorine pesticides may be partially responsible, it is probable that changes in the sourcing and processing of oils have also contributed.

More detailed comparison with existing data sets is confounded by the fact that many previous studies quote only means and ranges. That data distributions in the current study were skewed is clear from the large differences between mean and median values reported in Table 4. Comparison of median values, if possible, would perhaps be more robust. Nevertheless, data obtained in the current study were of a similar order of magnitude as values reported by Himberg *et al.* [14] for pharmaceutical grade fish oils, and by Falandysz [7] for a sample obtained in Iceland in 1984.

A high degree of variation in levels of contaminants between products of a similar nature is a characteristic of the data. In addition to changes in feedstock, variations in oil processing procedure parameters could result in batch-to-batch differences in contaminant levels even in the final product from a single manufacturer. It is interesting to note, for example, that samples 10 and 11 showed large differences in levels of all major contaminants despite being sold as the same product. The most likely explanation is that the two samples were derived from different batches of fish oil stock, one being more highly contaminated than the other. The lower density of sample 11 compared to 10 (0.811 g ml^{-1} and 0.980 g ml^{-1} respectively) suggests that the former may have been blended with another oil or other contaminant prior to bottling. We are currently investigating these differences in more detail. Nevertheless, should this anomalous result be shown to reflect inter-batch variation, perhaps as a consequence of contamination during refining, it would bring into question the quality control of products sold for human consumption. Such large differences would complicate further intercomparison of data and analysis of geographical and temporal trends. However, there are currently insufficient data available in the open literature to allow further assessment of the importance of such inter-batch variation.

The high value reported here for salmon oil (sample 30) may not be representative of the natural content of salmon oil since this oil may be extracted using pilchard or Greyfish oil [2] followed by simple centrifugation to separate the oil mixture. More probably, however, it reflects the use of farmed salmon reared in coastal waters and fed on synthetic foods which contain between 15 and 30% added fish oil (Sargent pers. comm.). The derivation of oils from wastes from farmed fish is likely to increase in the future [19].

Note that this study has focussed on a limited range of organochlorine residues; other organochlorine chemicals, which may well have been present in the samples, would not have been detected using the analytical techniques employed in this investigation. Chlorinated dioxins [20] and toxaphene [21], for example, have also been detected at significant concentrations in fish livers and in the oils derived from them.

It is clear that fish oils taken as dietary supplements may contribute significantly to daily intakes of organochlorine contaminants. Estimates of dietary intake vary greatly from country to country [22]. The most recent estimate for dietary intake of PCBs in the UK is 0.34 μg $person^{-1}$ day^{-1} (congener specific) [23]. For 15 of the supplements included in the current investigation, consumption at manufacturers recommended doses would lead to exceedence of this estimated average intake, in some cases by a wide margin. This is before other dietary sources of PCBs are taken into consideration. Even in countries such as Finland and the Netherlands, where high fish consumption results in higher estimated daily intakes of PCBs[22], the contribution from single daily doses of fish oil supplements could be significant. It follows that fish oil contamination requires careful monitoring if intended for use as a dietary supplement or in the preparation of other foodstuffs.

The therapeutic benefits conferred by the high vitamin and omega-3 fatty acid contents of fish oils have long been recognised [24]. More recently, their application in the treatment of a wide range of clinical conditions has been investigated, including cardiovascular disease[25,26,27,28], rheumatoid arthritis [29] and colon cancer [30]. Given that, in these applications, prescribed doses may be substantially above normal dietary supplementation, the elevated intake of organochlorine contaminants which may result warrants serious consideration.

REFERENCES

[1] A.P. Bimbo and J.B. Crowther, Fish oils: processing beyond crude oil, *INFOFISH International* **6/91**: 20-25 (1991).

[2] H.W. Ockerman, Fishery By-products. In:, (Ed) *Fish Processing Technology* (Edited by G..M. Hall), Chap. 6. Blackie Academic, London (1992).

[3] Euromonitor. Dietary Supplements. In *Market Research GB*, Volume XXXV: 121-138 (1994).

[4] K. Hansen, Developments in fish-processing equipment. Presented at *Developments in Fish Processing Conference*, North East Lincolnshire Council, UK, 24th-26th June (1997).

[5] K. Angsupanich and D.A. Ledward, High pressure processing of fish. Presented at *Developments in Fish Processing Conference*, North East Lincolnshire Council, UK, 24th-26th June (1997).

[6] K. Kannan, J. Falandysz, N. Yamashita, S. Tanabe and R. Tatsukawa, Temporal trends of organochlorine concentrations in cod-liver oil from the southern Baltic proper, 1971-1989. *Mar. Pollut. Bull.*, **24**: 358-63 (1992).

[7] J. Falandysz, Polychlorinated biphenyl concentrations in cod-liver oil: Evidence of a steady state condition of these compounds in the Baltic area oils and levels noted in Atlantic oils, *Arch. Environ. Contam. Toxicol.*, **27**: 266-271 (1994).

[8] J. Falandysz, K. Kannan, S. Tanabe and R. Tatsukawa, Organochlorine pesticides and polychlorinated biphenyls in cod-liver oils: North Atlantic, Norwegian Sea, North Sea and Baltic Sea, *Ambio*, **23**: 288-293 (1994).

[9] G. Sanders, S.J. Eisenreich and K.C. Jones, The rise and fall of PCBs: Time trend data from temperate industrialised countries, *Chemosphere*, **29**: 2201-2208 (1994).

[10] M.N. Jacobs, P.A. Johnston, C.L. Wyatt, D. Santillo and M.C. French (1997) Organochlorine pesticide and PCB residues in pharmaceutical, industrial and food grade fish oils. *Int. J. Environment and Pollution* **8(1/2)**: IN PRESS

[11] R.J. Law, C.R. Allchin and J. Harwood, Concentrations of organochlorine compounds in the blubber of seals from Eastern and North Eastern England, 1988, *Mar. Pollut. Bull.*, **20**: 110-115 (1989).

[12] D.E. Wells and J. De Boer, The 1993 QUASIMEME laboratory performance study: Chlorobiphenyls in fish oil and standard solutions, *Mar. Pollut. Bull.*, **29**: 174-184 (1994).

[13] M.M. Schantz, R.M. Parris, S.A. Wise, H.T. Won and R. Turie, Polychlorinated biphenyl (PCB) congeners and chlorinated pesticides in a cod liver oil reference material, *Chemosphere*, **24**: 1687-1698 (1992).

[14] K. Himberg, A. Hallikainen and K. Louekari, Intake of polychlorinated biphenyls (PCB) from the Finnish diet, *Z. Lebensm. Unters. Forsch.*, **196**: 126-130 (1993).

[15] M.P. Simmonds, P.A. Johnston, M.C. French, R. Reeve and J.D. Hutchinson, Organochlorines and mercury in pilot whale blubber consumed by Faroe islanders, *Sci. Tot. Environ.*, **149**: 97-111 (1994).

[16] FAO. *Codex Alimentarius, Volume 2, Pesticide Residues in Food.* Food and Agriculture Organisation of the United Nations/ World Health Organisation, Rome (1993).

[17] K. Ballschmiter, C. Rappe and H.-R. Buser, Chemicals properties, analytical methods and environmental levels of PCBs, PCTs, PCNs and PBBs, In *Halogenated Biphenyls, Terphenyls, Naphthalenes, Dibenzodioxins and Related Products* (Edited by R.D. Kimbrough and A.A. Jensen) Elsevier, Amsterdam: 47-102 (1989)

[18] MAFF. *Annual Report of the Working Party on Pesticide Residues*: 1991. Ministry of Agriculture Fisheries and Food/ Health and Safety Executive, HMSO: 76-77 (1992)

[19] L.J. Farmer, Effects of farming on the eating quality if fish. Presented at *Developments in Fish Processing Conference*, North East Lincolnshire Council, UK, 24th-26th June (1997).

[20] R.M.C. Theelen, Modelling of human exposure to TCDD and I-TEQ in the Netherlands: Background and occupation, In *Biological Basis for Risk Assessment of Dioxins and Related Compounds* (Edited by M.A. Gallo, R.J. Scheuplein and K.A. Van der Heijden, Banbury Report No: 35, Cold Spring Harbor Laboratory Press, New York: 277-290 (1991).

[21] J. De Boer, P. and Wester, Determination of toxaphene in human milk from Nicaragua and in fish and marine mammals from the North Eastern Atlantic and the North Sea, *Chemosphere*, **27**: 1879-1890 (1993).

[22] G. Moy, F. Kaferstein, Y.M. Kim, Y. Motarjemi and F. Quevedo, Dietary exposure to lead, cadmium, mercury and polychlorinated biphenyls, *Archiv fur Lebensmittelhygiene*, **44**: 45-51 (1993).

[23] Ministry of Agriculture, Fisheries and Food: Food Safety Directorate. *Polychlorinated biphenyls in food - UK dietary intakes,* Food Surveillance Information Sheet No. **89**: 14 pp. (1996).

[24] J. Falandysz, S. Tanabe R. and Tatsukawa Most toxic and highly bioaccumulative PCB congeners in cod-liver oil of Baltic origin processed in Poland during the 1970s and 1980s, their TEQ-values and possible intake, *Sci. Tot. Environ.*, **145**: 207-212 (1994)

[25] T.H. Lee, R.L. Hoover, J.D. Williams, R.I. Sperling, J. Ravalese, B.W. Spur, D.R. Robinson, D.R., E.J. Corey, R.A. Lewis and K.F. Austen, Effect of dietary enrichment with eicosapentaenoic and docosahexaenoic acids on in vitro neutrophil and monocyte leukotriene generation and neutrophil function, *New England J. Med.*, **312**: 1217-1224 (1985).

[26] M. Kestin, P. Clifton, G.B. Belling P.J. Nestel, n-3 Fatty acids of marine origin lower systolic blood pressure and triglycerides but raise LDL cholesterol compared with n-3 and n-6 fatty acids from plants, *Am. J. Clin. Nutr.*, **51**: 1028-1034 (1990).

[27] M.E. Turini, W.S. Powell, S.R. Behr and B.J. Holub, Effects of a fish-oil and vegetable-oil formula on aggregation and ethanolamine-containing lysophospholipid generation in activated human platelets and leukotriene production in stimulated neutrophils, *Am. J. Clin. Nutr.*, **60**: 717-724 (1994).

[28] T.A. Mori, R. Vandongen, L.J. Beilin, V. Burke, J. Morris and J. Ritchie, Effects of varying dietary fat, fish, and fish oils on blood lipids in a randomized controlled trial in men at risk of heart disease, *Am. J. Clin. Nutr.*, **59**: 1060-1068 (1994).

[29] J.M. Kremer, W. Jubiz, A. Michalek, R.I. Rynes, L.E. Bartholomew, J. Bigaouette, M. Timchalk, D. Beeler, and L. Lininger, Fish oil fatty acid supplementation in active rheumatoid arthritis, *Ann. Internal Med.* **106**: 497-503 (1987)

[30] H.-P. Bartram, A. Gostner, W. Scheppach, B.S. Reddy, C.V. Rao, G. Dusel, F. Richter, A. Richter and H. Kasper, Effects of fish oil on rectal cell proliferation, mucosal fatty acids and prostaglandin E2 release in healthy subjects, *Gastroenterology*, **105**: 1317-1322 (1993).