PCDD/Fs AND PCBs IN FISH OIL DIETARY SUPPLEMENTS

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Introduction

Fish oils are known to be a source of Vitamins A and D, and long chain fatty acids, particularly Omega-3 fatty acids. Over the last two decades the popularity of these products has increased significantly, in step with scientific research that has investigated the benefits of these oils for conditions such as cardiovascular disease, stroke, high blood pressure, rheumatoid arthritis and autoimmune disorders amongst others.

One of the main non-clinical disadvantages of using fish oils as dietary supplements that has emerged in recent years is the presence of environmental contaminants in these products. The natural habitat of the fish species (most fish oil dietary supplements are cod liver oil products) – the continental shelf especially around the North Sea and the Atlantic Ocean has suffered depositions of contaminants such as PCBs, dioxins, PBDEs, PAHs, pesticides, etc. The depositions have resulted in an enrichment of these pollutants, particularly in coastal sediments and generally in the marine food chain, especially fatty tissue. Fish oil dietary supplements are produced from such tissues and the initial crude product can contain relatively high levels of these contaminants. The actual concentrations observed are dependent not only on the species, but also on the geographical location[1,2] with products sourced from the Southern Hemisphere showing much lower levels than those from Northern Hemisphere locations such as the Baltic Sea or the North Sea.

In an effort to exclude these contaminants the industry employs a number of refining processes. General refining may include processes such as odour removal by steam stripping, exclusion of high molecular weight compounds by cold filtration, removal of other undesirable contaminants by activated charcoal filtration and other chemical methods[1]. However molecular distillation (using a temperature range of 180 – 220°C at < 100Pa) is far more effective at removing halogenated contaminants such as PCBs. A drawback of some of the refining processes is that beneficial components of the fish oils such as EPA and DHA are lost along with the contaminants, and the refining process employed must therefore strike a balance between contaminant exclusion and omega-3 fatty acid retention. Thus some manufacturers have invested in developing newer (undisclosed) purification methods that are able to achieve this balance and have claimed success. Another way of reducing the dietary exposure to consumers without further purification is of course, to reduce the recommended dosages, especially for children.

The last survey of fish oil dietary supplements for PCBs and dioxins carried out in the UK in the mid-nineties studied levels of these contaminants in retail products in 1994 and 1996. Combined (dioxin and PCB) concentrations for cod liver oils ranged from 7.3 - >44 ng WHO-TEQ/kg in 1994 and from 18 – 41 ng WHO-TEQ/kg in 1996[2]. The Tolerable Daily Intake (TDI) at the time was 10 pg I-TEQ/kg bodyweight /day and a typical consumer taking the recommended dose would not exceed this level except for toddlers (because of low body mass). In combination with

the rest of the diet however, all groups would have exceeded the 1998 WHO recommended TDI of 1-4 pg WHO-TEQ/kg bw/day[7]. This paper demonstrates the effects of improved purification procedures employed by manufacturers as well as establishing current levels of dioxins and PCBs in these products within the UK.

Sampling and Analysis

A total of 33 dietary supplement products, which in the main included cod liver oils but also salmon oils, other fish oils and formulations of cod liver oil with orange syrup, or lemon flavouring were purchased from retail outlets and by mail order during 2001-02. It was assumed that sampling would be representative as these products are distributed nationally across the UK. Products that were sampled in previous surveys in the UK[2] were included where they were still available for purchase, along with a number of additional products. Product expiry dates were checked to ensure validity of the shelf-life of the samples during the measurement period. Information on recommended dosage provided on the product labels together with other relevant sample details was recorded. Samples were stored, sealed in their original containers, at ambient temperature prior to analysis. Where samples were sold as capsules, the casings were excluded from the analytical sample. Methods used for the extraction and analysis of the samples have been reported previously[3] and were accredited to ISO 17025 standards. Sample batches included a blank and a suitable reference material. Data quality was ensured by continuous successful participation in international inter-calibration exercises.

Results and Discussion

All the samples that formed part of this study showed detectable levels of PCDD/Fs and PCBs except for one cod liver oil sample where PCDD/Fs were below detection limits. The reporting limit quoted for non-ortho-PCBs and dioxin congeners was the limit of determination that prevailed in that instance. The limit varied depending on the various different congeners, but at its lowest value was 0.02 ng/kg fat. For the ortho-PCBs, a reporting limit of 0.10 μg/kg fat was applied. A summary of the upper-bound data on a fat weight basis for the samples is presented in Table 1. One sample that was a mixture (of unspecified proportions) of cod liver oil and orange syrup is not included in the summary.

These data show that the greater contribution to the total Σ WHO-TEQ is made by the PCBs with approximately 55% from the non-ortho-PCBs and 24% from the ortho-PCBs with the remainder coming from the PCDD/F contribution. This is in agreement with observations on fish oil and fish in general[4,5]. In particular, when compared to the survey commissioned by the Food Safety Authority of Ireland[5] the contribution from PCDD/F to the Σ WHO-TEQ is the same at 21%. The data are therefore directly comparable with the UK data showing similar concentration ranges for the dioxins (0.2 -8.4 ng/kg WHO-TEQ, compared to 0.2 -11 ng/kg WHO-TEQ for the Irish results).

Both sets of data are positively skewed although the UK data shows a much higher proportion of lower concentrations (median 0.9 ng/kg WHO-TEQ) compared to the Irish data which show similar mean and median values (3.6 and 3.1 ng/kg WHO-TEQ respectively). However data for the PCB TEQ shows slightly different ranges with a lower range of 0.2 – 29.8 for the Irish data compared to 1.1 – 41.5 ng/kg PCB TEQ.

However it is interesting to note that the mean and median values for the UK data are much lower despite the higher range. This suggests that the mean (9.4 ng/kg) for the UK PCB WHO-TEQ data is influenced by a small number of high concentration samples which tend to exaggerate this value. This view is supported by the lower median value of 4.9 ng/kg and a skewness value of 1.7. The Irish data on the other hand shows a much more even distribution with good agreement between the mean and median values and a very low skewness value (0.2). The distribution for both sets of PCB WHO-TEQ data are shown in Figure 1.

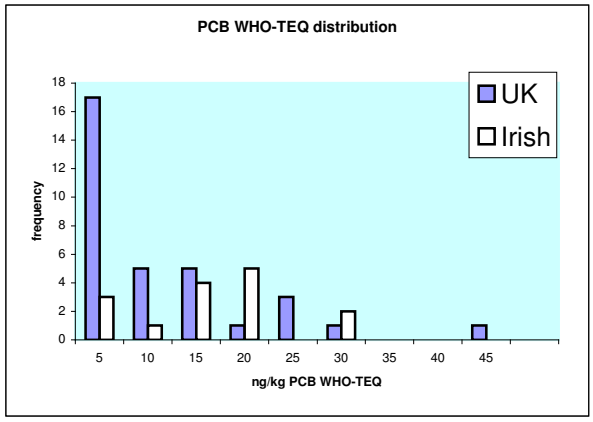

Fig 1: Comparison of UK and Irish survey results

A more detailed assessment within individual analyte groups shows that in order of contribution, PCB 126 is the largest contributor to total Σ WHO-TEQ followed by PCB 118 and PCB 156. 12378 PCDD, 23478 PCDF, 2378 TCDD and 2378 TCDF also make significant, though smaller contributions. It must be emphasised that these observations refers to the fish oil data in the group of samples discussed in this paper and probably reflect the effects of the purification procedures used by manufacturers, rather than the original contaminant levels in the oil. An important implication that follows is that the reduction in levels that is observed in this data set compared to previous data could well be the result of better purification procedures or more selective sourcing of products based on geography, rather than actual reductions in the crude oil products. This view needs to be balanced with the observations on gradually declining dioxin levels in fish samples in general.

The study also revealed that 12 of the 33 products showed concentrations in excess of the EU maximum limit for fish oil (2 ng/kg; maximum limit and 1.5 ng/kg; target limit). Of these 11 were cod liver oils and one was a salmon oil. An additional cod liver oil product showed a concentration above the action limit. Approximately 64% of the products surveyed showed concentrations of dioxins well below the EU maximum limits.

Thus where to some extent contaminant removal methodologies have had some success in reducing the dioxin component of the WHO-TEQ, the emphasis should now move to excluding PCBs as well, from these products.

Conclusions

This survey of fish oils dietary supplements for dioxin and dioxin-like PCBs demonstrates continued surveillance in the UK of a product that can make a significant contribution to the total human exposure to these contaminants. The data obtained for dioxins are in good agreement with the survey of fish oils carried out by the Food Safety Authority of Ireland. The results suggest that some products show lower PCDD/F concentrations relative to earlier surveys. Estimates of human exposure of different population groups resulting from consumption of the fish oil products of

their own as well as in combination with the diet show that some products would give intakes in excess of the current UK TDI of 2 pg (dioxin + dioxin-like PCB) WHO-TEQ/kg bodyweight/day[6].

Table 1: Survey results: PCDD/Fs and PCBs in fish oil

Parameter	Σ PCDD/F WHO-TEQ	Σ non-*o* PCBs WHO-TEQ	Σ *o*-PCBs WHO-TEQ	Σ Total WHO-TEQ	Σ *o* -PCBs & non-*o* PCBs WHO-TEQ
n=32*		ng/kg fat weight			
Mean	2.5	6.6	2.8	11.9	9.4
Median	0.9	2.4	2.4	5.7	4.9
Skewness	1.0	1.5	1.6	1.3	1.7
Minimum	0.2	0.6	0.3	1.9	1.1
25th Percentile	0.6	1.1	1.8	3.2	3.0
50th Percentile	0.9	2.4	2.4	5.7	4.9
75th percentile	4.6	9.9	3.1	17.1	13.0
Maximum	8.4	31.3	10.1	46.0	41.5
SD	2.6	7.7	2.2	11.8	9.6
Mean+2SD	7.6	21.9	7.2	35.6	28.7
% contribution to Σ WHO-TEQ	21	55	24		

***** One syrup formulation sample not included due to unspecified fish oil content.

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