Estimation of dietary intake for PCDDs, PCDFs and DLPCBs in various marine organisms from the coastal areas of Korea

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Introduction

A large variety of organic chemicals, primarily anthropogenic pollutants, are transported into coastal marine ecosystems through various routes. Marine organisms may be exposed to toxic organic contaminants by contact with contaminated seawater and sediments, either on the seabed or through suspended sediments, or by ingestion of contaminated prey[1]. Due to low water soulubility and high octanol/water partition coefficients (K_{ow}) in seawater, these chemicals can retain and concentrate in fatty tissues of these marine organisms such as migrate into fish, shellfish and invertebrates. The different chemical characterisation of toxic contaminants has been observed in organisms of different trophic levels[2,3]. These differencies were attributed to a partial biotransformation of the contaminants in the organisms of higher trophic levels[4]. In aquatic systems, the highly lipophilic and hydrophobic organic pollutants tend to bioconcentrate from water to aquatic animal and then biomagnify up through the multistep food chain[5,6]. Hence, these organisms reflect the pollution extent of persistent toxic organic pollutants and some species are used as bio-indicators at different environmental conditions and foodweb[7].

Exposure of humans to dioxins and dioxin-like contaminants occurs mainly (>95%) through contamination of food and the inhalatory route contributes only a neglible extent[8]. The present risk assessment will briefly outline the major exposures of PCDDs/DFs and dioxin-like PCBs (DLPCBs) and their toxicity, and finally characterize the health risk for humans associated with dietary uptake of contaminated seafood.

Materials and Methods

Sample collection: Seventy marine organism samples (40 species) were purchased at local fisheries markets from various locations distributed over Korean coastal areas from July 2001 to October 2003. These organisms are common edible species and are commercially important food items in Korea. In addition, these samples can be classified with five groups of fish, crustacea, bivalvia, gastropoda and cephalopoda. Information on selected marine organisms in this study was summarized in Table 1.

Table 1: Information on selected marine organisms in this study

Classification	Species no.	Sample no.	Moisture (%)	Lipid (%)[a]
Fish	24	38	62 – 84	1.1 – 48
Crustacea	5	9	58 – 79	2.4 – 17
Bivalvia	6	13	46 – 84	2.0 – 10
Gastropoda	3	6	65 – 78	2.1 – 4.0
Cephalopoda	2	4	63 – 79	3.1 – 4.7

[a]in the dry weight.

Sample preparation and analysis: Marine organism samples were pooled and then homogenized with an ultra-disperser. After the internal standards for PCDDs/DFs and DLPCBs spiked into homogenized samples (100 g), they were decomposed in 200 mL of 1 N KOH ethanolic solution (Dioxin analysis, Wako) for 2 hours by mechanical shaking. The digest was liquid-liquid extracted with twice using 150 mL of *n*-hexane after the addition of water and 50 g of anhydrous Na_2SO_4. The extracts were reduced to small volume in a rotary evaporator and then adjusted to a volume of 10 mL. After pre-cleaned up with a multi-layer slilica gel column chromatography containing $AgNO_3$-silica gel, H_2SO_4-silica gel and KOH-silica gel, the extract was cleaned up on an activated alumina column chromatography with successive elutants of 3% methylene dichloride in *n*-hexane and 50% methylene dichloride in *n*-hexane. The second fraction was concentrated to less than 1 mL, and left at a room temperature for one or two days to evaporate to dryness. The residue was dissolved with 30 µL of *n*-nonane and determined for PCDDs/DFs and DLPCBs using HRGC/HRMS (JMS 700D, JEOL). Further details of the experimental procedure and instrumental analysis of PCDDs/DFs and DLPCBs are presented elsewhere [9,10].

Results and Discussion

Residues of PCDDs/DFs and DLPCBs: Residues of PCDDs/DFs and DLPCBs in various marine organisms from Korean coastal areas were presented in Figure 1.

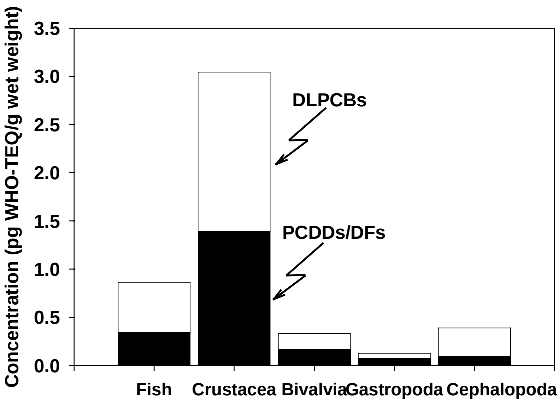

Figure 1. Comparison of the residues of PCDDs/DFs and DLPCBs in selected marine organisms in Korea.

In general, the crustacea species had the highest residues in PCDDs/DFs and DLPCBs, followed by fish, cephalopoda, bivalvia and gastropoda species. The concentrations of PCDDs/DFs in fish varied from 0.028 to 1.63 pg WHO-TEQ/g wet weight and DLPCBs concentrations varied from 0.023 to 4.16 pg WHO-TEQ/g wet weight. The highest contents of PCDDs/DFs and DLPCBs were detected in Roundnose flounder (*Eopsetta grigorjewi*) from Jumunjin coast. Roundnose flounder (*Eopsetta grigorjewi*) and Herring (*Clupea pallasii*) from Pohang coast and Mi-iuy croaker (*Michthys miiuy*) from Mokpo coast also showed relatively high contents of these chemicals. The residues of PCDDs/DFs in curstacea species were in the range of 0.102–4.39 pg WHO-TEQ/g wet weight and the DLPCBs residues were in the range of 0.079–6.0 pg WHO-TEQ/g wet weight. For bivalvia species, the levels of PCDDs/DFs and DLPCBs revealed from 0.02 to 0.42 pg WHO-TEQ/g wet weight and from 0.02 to 0.52 pg WHO-TEQ/g wet weight, respectively. Gastropoda species showed the concentration ranges of 0.031–0.14 pg WHO-TEQ/g wet weight for PCDDs/DFs and of 0.008–0.087 pg WHO-TEQ/g wet weight, respectively. The concentrations of PCDDs/DFs and DLPCBs in cephalopoda species were in the ranges of 0.058–0.17 pg WHO-TEQ/g wet weight and 0.021–0.61 pg WHO-TEQ/g wet weight, respectively.

Comparison of PCDDs/DFs and DLPCBs residues in fish with other countries: The PCDDs/DFs and DLPCBs residues in various fish species measured in this study were compared with those in other countries. PCDDs/DFs levels in fishes purchased from local market of Seoul and Busan of Korea were in the range of 0.01–0.58 pg I-TEQ/g wet weight[11]. These experimental results were lower than or comparable to those in this investigation. Also, PCDDs/DFs residues in fishes from Tokyo Bay of Japan[12] and from Adriatic Sea of Itlay[13] were similar to those in this study. However, other reports from several countries were about an order of 2-10 times greater than in this study. In particular, PCDDs/DFs concentrations in fishes from Lough Neagh of UK[14], the southern Germany[13] and Baltic Sea of Sweden[15] were much higher values than the different locations surveyed including this study. The DLPCBs concentrations in various fish species in this study were comparble to those from Seoul of Korea[11]. However, other results from other countries were much higher than DLPCBs residues in fishes in this study. In particular, California of USA revealed the higher levels of DLPCBs with a residue range of 1.19–32 pg I-TEQ/g wet weight. Therefore, the PCDDs/DFs and DLPCBs levels in fishes from the Korean coastal areas were likely to be moderate in comparison to those of other countries.

Estimation of daily intake on PCDDs/DFs and DLPCBs in marine organisms: The daily intake of individual organism published from the Korea Ministry of Health & Welfare was used[16]. Human dietary intake of PCDDs/DFs and DLPCBs through some kinds of marine organisms has been determined and daily doses of uptake have been calculated as shown in Table 2.

Table 2: Estimated daily intake of different organism species for PCDDs/DFs and DLPCBs

	Intake amount (g/day)	Daily dietary intake (pg WHO-TEQ/kg body weight/day)		
		PCDDs/DFs	DLPCBs	Total
Fish	30.5	0.175	0.262	0.437
Crustacea	3.3	0.077	0.091	0.167
Bivalvia	4.4	0.012	0.012	0.024
Gastropoda	0.5	0.001	0.0003	0.001
Cephalopoda	7.8	0.013	0.038	0.051
Total	46.5	0.28	0.40	0.68

Dietary daily intake of PCDDs/DFs for various marine organism species was estimated to be 0.28 pg WHO-TEQ/kg body weight/day and that for DLPCBs was estimated to be 0.40 pg WHO-TEQ/kg body weight/day. Also, total dietary daily intake of dioxins and dioxin-like PCBs was estimated to be 0.68 pg WHO-TEQ/kg body weight/day.

Average daily dietary intake of fishes from the Korean coastal environments by PCDDs/DFs was estimated at 0.175 pg WHO-TEQ/kg body weight/day and 0.262 pg WHO-TEQ/kg body weight/day by DLPCBs. Daily dietary intakes of PCDDs/DFs and DLPCBs for crustacea species were 0.077 and 0.091 pg WHO-TEQ/kg body weight/day, respectively. Bivalvia daily intakes for PCDDs/DFs and DLPCBs were estimated at 0.012 and 0.012 pg WHO-TEQ/kg body weight/day, respectively. In the case of cephalopoda species, the daily intake of PCDDs/DFs was 0.013 pg WHO-TEQ/kg body weight/day and DLPCBs intake was 0.038 pg WHO-TEQ/kg body weight/day. Gastropoda species was the lower values for PCDDs/DFs and DLPCBs.

In Korea Food and Drug Administration (KFDA), a TDI for PCDDs/DFs and DLPCBs in Korea was proposed as 1–4 pg WHO-TEQ/kg body weight/day. PCDDs/DFs and DLPCBs dietary intakes in this investigation did not exceed a TDI proposed by KFDA as well as tolerable weekly intake (TWI) and tolerable monthly intake (TMI) suggested by EU and UK[17]. Therefore, the dietary intake for dioxins and dioxin-like contaminants in Korean marine organisms was relatively safe as an aspect of risk assessment for human body.

Contribution of various marine organisms for the daily intake: The contributions to the estimated dietary intakes of different organisms for PCDDs/DFs and DLPCBs were presented in Figure 2. Contribution of individual marine organism for dietary intake of PCDDs/DFs and DLPCBs was the highest proportion in fish species (64.2%), followed by crustacea (24.6%), cephalopoda (7.4%), and bivalvia species (3.6%).

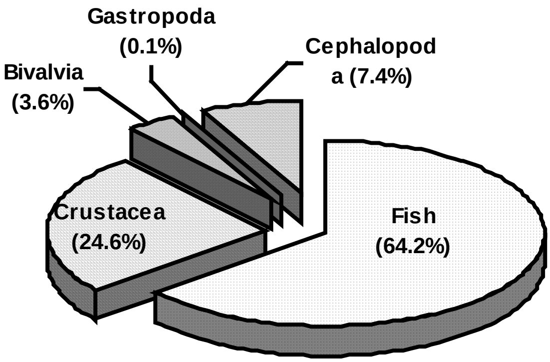

Figure 2: Contribution to the estimated dietary intakes of various marine organisms for PCDDs/DFs and DLPCBs.

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