

HUMAN EXPOSURE TO PCDDs, PCDFs AND DIOXIN-LIKE PCBs ASSOCIATED WITH SEAFOOD CONSUMPTION IN KOREA FROM 2005 TO 2007

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Introduction

Seafood is a major source of proteins and healthy lipids for people. In particular, the long chain omega-3 fatty acids have been shown to have numerous beneficial roles in the human body. Despite the health benefits of seafood diet, an issue of concern related to frequent seafood consumption is the potential risk arising from exposure to toxic contaminants in fish and shellfish^{1,2}. Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (DLPCBs) accumulate in human body fat over lifetime. The levels of dioxins and dioxin-like contaminants in fish and humans have decreased considerably during the past two decades as a consequence of the ban on production and reduction in emissions^{3,4}. However, the dietary intakes of these compounds vary considerably between countries and between population groups within countries. Certain aspects of the diet within a particular subgroup of the population may differ from those of the general population. These special subgroups may have higher risks from dietary exposure of PCDD/Fs and DLPCBs than the general population and require special consideration. There has been no data published concerning exposure assessment of the populations according to the special subgroups such as infants and high seafood consumers in Korea. In this study, we estimated dietary intake of PCDD/Fs and DLPCBs by seafood consumption for the general population and the specific subpopulations according to sex, age and high seafood consumers in Korea.

Materials and methods

Twenty-six marine species ($n = 78$) were collected annually during 2005 to 2007 from Busan cooperative fish market, which is the largest fish market in Korea. These organisms are the most commonly consumed species and commercially important species in Korea. Experimental procedures and instrumental analysis of PCDD/Fs and DLPCBs in marine organisms were performed following the methods described elsewhere^{5,6}. Dietary intakes of PCDD/Fs and DLPCBs were calculated by multiplying the consumption rate of individual seafood with the corresponding chemical concentrations. The concentrations of undetected congeners were calculated as one half of the respective LOD. Average daily consumption rates of foodstuffs for general populations of Korea were obtained from 'National Health and Nutrition Survey' by the Ministry of Health and Welfare⁷. Average daily consumption of the 26 seafoods analyzed in this study amounted to 50.6 g, which accounted for 3.8% of the total food ingested. The 95th percentile data of individual seafoods were used for the risk assessment of high seafood consumers in Korea. The dietary intakes of PCDD/Fs and DLPCBs were estimated according to sex and age, to assess the risks in specific subpopulations. Age groups were classified into six categories; < 2 years, 3–6 years, 7–12 years, 13–19 years, 20–29 years, 30–49 years, 50–64 years, and > 65 years.

Results and discussion

Concentrations of PCDD/Fs and DLPCBs

Concentrations of lipids, PCDD/Fs and DLPCBs in individual seafoods in Korea collected from 2005 to 2007 are summarized in Table 1. The lipid contents in seafood samples ranged from 0.56% to 54% on a dry weight basis. The concentrations of PCDD/Fs and DLPCBs ranged from 0.008 to 2.85 pg TEQ/g wet wt and from 0.002 to 4.76 pg TEQ/g wet wt, respectively. Overall concentrations of DLPCBs in seafood samples were twice those of PCDD/Fs. The total TEQ concentrations ranged from 0.01 to 6.76 pg TEQ/g wet wt with a mean of 1.18 pg TEQ/g wet wt. The highest TEQ concentration in marine species was found for mackerel (4.20 ± 2.23 pg TEQ/g wet wt), followed by herring (4.05 ± 0.73 pg TEQ/g wet wt), eel (4.04 ± 0.59 pg TEQ/g wet wt) and tuna (4.03 ± 0.46 pg TEQ/g wet wt). Hairtail and Spanish mackerel showed relatively high residues of PCDD/Fs and DLPCBs. These species are long-lived and are larger predatory fish with high lipid contents ($28 \pm 15\%$), compared with other species ($7.6 \pm 8.4\%$).

Congener profiles of PCDD/Fs and DLPCBs

The contribution of 2, 3, 7, 8-substituted PCDFs (average 82%) to the total PCDD/Fs concentration was higher than that of PCDDs (average 18%). The predominant congener of PCDD/Fs was 2, 3, 4, 7, 8-PeCDF, which accounted for $31 \pm 27\%$ in all of the seafood samples, followed by 2, 3, 7, 8-TCDF ($25 \pm 25\%$) and 1, 2, 3, 7, 8-PeCDF ($10 \pm 12\%$). A moderate proportion with high standard deviation ($7 \pm 20\%$) of OCDD is due to the contributions by shellfish and cephalopod samples, which had high concentrations of OCDD relative to the total PCDD/F concentrations. The congener profiles of DLPCBs were similar for all of the seafood samples. Pentachlorobiphenyls (PCB 123, 118, 114, 105 and 126) were the predominant homologue group, which contributed more than 75% to the total concentrations of DLPCBs. PCB 118 was present in the highest proportion ($53 \pm 10\%$) among the DLPCB congeners. The next dominant congener was PCB 105 with a percentage of $16 \pm 4\%$. These congener patterns of PCDD/Fs and DLPCBs in seafood samples were similar to those previously reported⁵.

Dietary intakes of PCDD/Fs and DLPCBs by seafood consumption

Estimated dietary intakes of PCDD/Fs and DLPCBs through seafood consumption for the general population and some subpopulations in Korea are summarized in Table 1. The average dietary intake of PCDD/Fs and DLPCBs through seafood consumption in Korea was estimated to be 1.23 pg TEQ/kg body weight/day. The contribution of dietary intake of DLPCBs, an average 70%, was higher than PCDD/Fs. Among marine edible species investigated in the present study, mackerel contributed to the highest intake of 0.43 pg TEQ/kg bw/day, accounting for 35.3% of the total dietary TEQ intake. The dietary intakes through the consumption of tuna and hairtail were 0.22 and 0.16 pg TEQ/kg bw/day, respectively. The yellow croaker, eel, anchovy and Spanish mackerel showed moderate contributions, which accounted collectively for 20% of the total dietary TEQ intake. Other species contributed less than 15% of the total TEQ intake. Interestingly, species such as mackerel, tuna and anchovy, which contributed to high dietary intakes of PCDD/Fs and DLPCBs, also have high levels of omega-3 fatty acids⁸. Recently, the debate on health benefits versus risks of seafood consumption has attracted a great scientific and social interest^{1,8}. Several studies reported that the increasing evidence of health benefits of seafood consumption, which may be promoted by efforts to reduce dioxin contamination, rather than restricting seafood intake^{1,8}.

The dietary intakes of PCDD/Fs and DLPCBs through seafood consumption for males and females of the Korean population were estimated to be 1.40 and 1.06 pg TEQ/kg bw/day, respectively. The males had a higher intake rate than the females, because males (58.2 g/day) consume higher levels of seafood than females (43.5 g/day). The current dietary intake of PCDD/Fs and DLPCBs by seafood consumption for the general population (1.23 pg TEQ/kg bw/day), females (1.40 pg TEQ/kg bw/day) and males (1.06 pg TEQ/kg bw/day) in Korea was within the range of TDI (1–4 pg TEQ/kg bw/day) recommended by the WHO. However, it is below the TDI or TWI proposed by the Korea Food and Drug Administration, UK Committee on Toxicity and the EU Scientific Committee on Food. Therefore, Korean seafood contributes to low dietary intake compared with the guidelines established by international organizations. The dietary intakes of PCDD/Fs and DLPCBs in the present study did not include the exposures from other foodstuffs in Korea.

The dietary intakes (66.8 pg TEQ/day) of PCDD/Fs and DLPCBs from seafood consumption in Korea are compared with the data recently reported in the literature. The dietary intake by seafood consumption in Finland (95 pg TEQ/day)⁹ was higher than that in the present study. The dietary intakes in Belgium (71 pg TEQ/day)¹⁰, Japan (63.8 pg TEQ/day)¹¹, Guangzhou (66 pg TEQ/day) and Zhoushan (58.2 pg TEQ/day) in China¹² were similar to that obtained in our study. The dietary intakes of total TEQs by seafood consumption in some countries such as Spain (34.9 pg TEQ/day)¹³, Taiwan (34.5 pg TEQ/day)¹⁴ and Egypt (32.4 pg TEQ/day)¹⁵ were one half of the values found in the present study. Sweden (15.6 pg TEQ/day)¹⁶ and Uzbekistan (6.8 pg TEQ/day)¹⁷ had lower values than those reported from the present study and other countries. In particular, Uzbekistan had a low consumption rate (1.32 g/day) of seafood compared with other countries.

Dietary intakes of PCDD/Fs and DLPCBs by high seafood consumer

The dietary intakes of PCDD/Fs and DLPCBs by high seafood consumers (95th percentiles) were estimated to be 4.16 pg TEQ/kg bw/day, which exceeded the TDIs proposed by the WHO, KFDA and the UK committee. This

result indicates that high seafood consumers in Korea have the potential risk from seafood consumption alone. Therefore, risk management activities such as limiting the consumption frequency or the consumption of contaminated fish are essential to protect their health. High contributors among marine species are mackerel, tuna and anchovy, similar to those for the general population.

Table 1. Concentrations of PCDD/Fs and DLPCBs and estimated dietary intakes (pg TEQ/kg bw/day) of PCDD/Fs and DLPCBs from seafood consumption by sex groups, high seafood consumers and the general population

	Concentrations (pg TEQ/g wet wt)		Dietary intakes (pg TEQ/kg bw/day)			
	PCDD/Fs	DLPCBs	Male	Female	95 th	Average
Mackerel	0.06 ± 0.08	0.10 ± 0.02	0.51	0.36	2.87	0.43
Tuna	0.04 ± 0.02	0.12 ± 0.12	0.25	0.19	0.60	0.22
Hairtail	1.37 ± 0.64	2.83 ± 1.68	0.18	0.14	0	0.16
Yellow croaker	0.22 ± 0.22	0.65 ± 0.97	0.09	0.09	0.18	0.09
Eel	0.31 ± 0.48	0.55 ± 0.32	0.07	0.04	0	0.06
Anchovy	1.01 ± 0.39	3.02 ± 0.16	0.06	0.05	0.21	0.05
Spanish mackerel	0.87 ± 0.45	2.57 ± 0.58	0.05	0.04	0	0.05
Squid	0.01 ± 0.003	0.09 ± 0.09	0.02	0.02	0.12	0.02
Crab	0.38 ± 0.08	0.23 ± 0.18	0.02	0.02	0	0.02
Saury	0.14 ± 0.22	0.14 ± 0.20	0.03	0.01	0	0.02
Alaska pollack	0.08 ± 0.03	0.40 ± 0.32	0.02	0.02	0.12	0.02
Rockfish	0.04 ± 0.04	0.07 ± 0.05	0.02	0.01	0	0.02
Olive flounder	0.88 ± 0.21	1.42 ± 0.33	0.02	0.01	0	0.01
Oyster	0.22 ± 0.10	0.28 ± 0.29	0.01	0.01	0	0.01
Flounder	0.46 ± 0.16	0.71 ± 0.54	0.01	0.01	0	0.01
Shrimp	0.10 ± 0.09	0.29 ± 0.15	0.01	0.01	0.03	0.01
Herring	0.41 ± 0.44	0.61 ± 0.10	0.01	0.01	0	0.01
Cod	0.84 ± 0.68	3.20 ± 1.09	0.01	0.01	0	0.01
Clam	0.31 ± 0.30	0.41 ± 0.41	0.004	0.004	0.024	0.004
Octopus	0.06 ± 0.08	0.04 ± 0.01	0.003	0.002	0	0.003
Ray	0.06 ± 0.08	0.05 ± 0.04	0.003	0.001	0	0.002
Mussel	0.13 ± 0.21	0.05 ± 0.04	0.002	0.002	0	0.002
Angler fish	0.04 ± 0.05	0.07 ± 0.07	0.001	0.001	0	0.001
Catfish	0.27 ± 0.15	0.11 ± 0.03	0.002	0.001	0	0.001
Ark shell	0.09 ± 0.11	0.24 ± 0.32	0.001	0.001	0	0.001
Filefish	1.98 ± 0.82	2.07 ± 0.09	0.001	0.0004	0	0.001
Sum			1.40	1.06	4.16	1.23

Dietary intakes of PCDD/Fs and DLPCBs according to age

The total body burden from dietary intake of PCDD/Fs and DLPCBs increases with increasing age, with the exception of > 65 years-old age group ((a) in Figure 1). The dietary intake of PCDD/Fs and DLPCBs according to age ranged from 15.4 pg TEQ/day for < 2 years old to 86.5 pg TEQ/day for 50–64 years. However, considering both factors of seafood consumption rate and body weight, the dietary intakes of PCDD/Fs and DLPCBs ranged from 0.9 to 1.6 pg TEQ/kg bw/day ((b) in Figure 2). The age group with the highest exposure rate was 3–6 years old, followed by 50–64 years, 30–39 years and < 2 years. In particular, children of 3–6 years and < 2 years have high risks compared with adults. These age groups also have potential risks from exposure through human milk¹⁸. Taking into account both exposure routes to infants (seafood consumption and human milk), the adequate risk management programs should be instituted for infant health, although both exposure

routes contribute to good nutritional benefits.

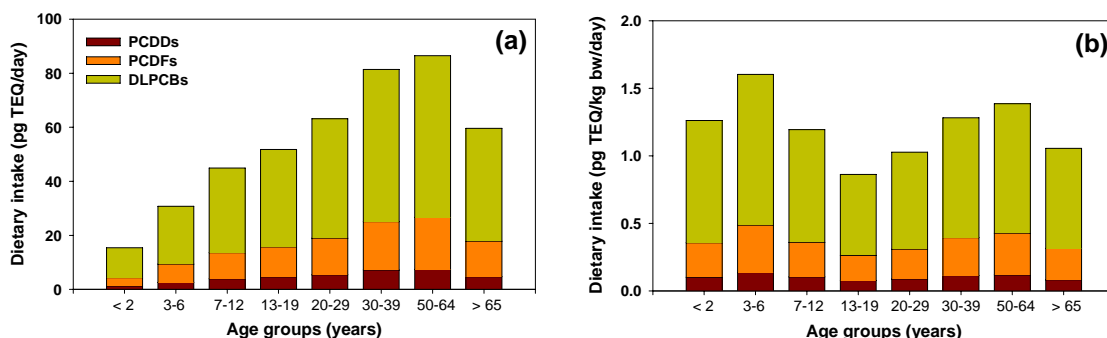


Figure 1. Estimated dietary intakes of PCDD/Fs and DLPCBs through seafood consumption for six age groups. Dietary intake (a, pg TEQ/day) is total burden of PCDD/Fs and DLPCBs and dietary intake (b, pg TEQ/kg bw/day) is re-calculated by body weights for respective age groups.

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