

RISK ASSESSMENT AND QUANTIFICATION OF DIETARY EXPOSURE TO PCDDs, PCDFs AND CO-PCBs IN KOREA

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Introduction

Concern of population to food safety is continuously increasing after occurrence of Belgium dioxins crisis and dioxins is now still major contaminants in food managing area. Human background exposure to PCDDs/Fs, and PCBs predominantly occurs through the diet with food from animal origin being the major source. The most usual method of assessing average dietary intake is to multiply the average consumption of each type of food by the average concentrations found in corresponding food samples, and then add together the contributions from various components of the diet.

The major effects of PCDDs/Fs include dermal toxicity, immunotoxicity, reproductive effects and teratogenicity, endocrine disruption and carcinogenicity. Recently, 2,3,7,8-tetrachlorodibenzo-p-dioxin has been newly proposed for upgrade to the "known to be a human carcinogen" category and more than 100 other dioxin-like compounds are classified as "reasonably anticipated to be human carcinogen" by U.S.EPA.

This study was conducted to quantify dietary human exposure to the PCDDs/Fs and Co-PCBs and to assess hazardous risk would be expected due to dietary exposure.

Materials and Method

Subjected Foods and Sampling

Total 75 food samples including cereal, vegetable, meat, egg, fish and dairy product were collected randomly from food markets in several sites(5cities) for representative nationwide monitoring as composite sampling. Cereals, vegetables, and dairy products were stored at 4°C and at -20°C for fishes and meats until analysis.

The subjected foods were selected through consideration of foreign contaminated data which were previously published and by regarding of highly consumed foods to general Korean people.

Edible portion of all sample was regarded for PCDDs/Fs and Co-PCBs analysis.

PCDDs/Fs and Co-PCBs Analysis

Analyses of samples for PCDDs/Fs (all seventeen 2,3,7,8-substituted congeners) have been carried out according to the method described by the U.S.EPA Method 8290 and U.S.EPA Method 1613 which is the determination of PCDDs/Fs by isotope dilution HRGC/HRMS revised in October 1994.

For the analyses of the Co-PCBs (#77,#126, #169), sample was digested with ethanolic HCl solution, extracted with hexane, and passed through florisil column. The hexane was dried

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and the residue was reconstituted with methanol and injected into GC/MSD system. The characteristic M^+ and $M^+ + 2$ ions of each congener of PCBs were monitored using selected Ion Mode (SIM) for the quantitation and confirmation of congeners. Internal reference standard for PCBs was 2,3,4,4'-TCB(IUPAC No.60). The sample amounts for cereals, vegetables, fishes and meat were 50g, 100g for whole milk and 20 gram for cheese. TEQs in this study was computed by assuming that nondetects equaled zero.

Quantification of Dietary Human Exposure

The equation and parameters in Table 1 are to quantify the dietary human exposure of PCDDs/Fs and Co-PCBs. General adult group was regarded as target population and National Nutrition Survey Report was used for food consumption of individual food.

Table 1. Human exposure parameters and equation for quantification of daily dietary intake to PCDDs/Fs and Co-PCBs

$\text{Daily Dietary Intake} = \frac{\sum_{i=1}^n C_i \times IR_i \times CF}{BW}$
C_i : Concentration (mean value) of PCDDs/Fs and Co-PCBs in food i (pg/kg) IR_i : Ingestion rate of food i (g/day, National Nutrition Survey Report, 1995) BW : Body weight (60 kg, adult mean, Korea Research Institute of Standard and Science, 1998) CF : Conversion factor

Results and Discussion

PCDDs/Fs and Co-PCBs in Foods

The analyzed results of this study are presented in Table 2. In general the levels of PCDDs/Fs and Co-PCBs found in the food samples were very low. On a whole weight basis, TEQ ranged from 0.014~0.018 pgTEQ/g for the grain samples, 0.0014~0.023 pgTEQ/g for the vegetable, 0.0125~0.313 pgTEQ/g for the fish, 0.01~0.026 pgTEQ/g for the dairy products, 0.002~0.026 pgTEQ/g for the meat and egg.

Table 2 PCDDs/Fs and Co-PCBs levels in foods (mean)

Congener	I-TEQ*	Rice (n=5)	Barely (n=5)	Bean (n=5)	Radish (n=5)	Korean Cabbage (n=5)	Mackerel (n=5)	Pollack (n=5)
2,3,7,8,-TCDD	1.0	0	0	2.6	0	0	3.32	0
1,2,3,7,8-PeCDD	1.0	2.15	2.76	4.59	0.06	22.42	26.37	0
1,2,3,4,7,8-HxCDD	0.1	0.876	0.52	0.338	0	0	0	0
1,2,3,6,7,8-HxCDD	0.1	0.736	1.226	0.888	0	0	1.552	0
1,2,3,7,8,9-HxCDD	0.1	0.524	0.074	0.082	0	0	0	0
1,2,3,4,6,7,8-HpCDD	0.01	0.577	0.084	0.135	0	0	0.1894	0
OCDD	0.0001	0.045	0.098	0.058	0.0373	0.166	0.745	1.793
Total PCDDs (fg TEQ/g)		4.908	4.762	8.690	0.0973	22.586	32.176	1.793

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Table 2 .Continued

2,3,7,8-TCDF	0.1	0	0.56	0.678	0.316	0.508	8.78	9.452
1,2,3,7,8-PeCDF	0.05	0.261	0.676	0.429	0.006	0	0.658	1.308
2,3,4,7,8-PeCDF	0.5	2.91	6.02	2.01	0	0	0	0
1,2,3,4,7,8-HxCDF	0.1	2.63	1.792	0.796	0.008	0	0	0
1,2,3,6,7,8-HxCDF	0.1	1.302	1.884	0.964	0.012	0	1.898	0
1,2,3,7,8,9-HxCDF	0.1	1.502	0.942	0.638	0	0	0	0
2,3,4,6,7,8-HxCDF	0.1	2.044	1.426	0.65	0	0	0	0
1,2,3,4,6,7,8-HpCDF	0.01	0.449	0.206	0.198	1.03	0.194	0.561	0
1,2,3,4,7,8,9-HpCDF	0.01	0.545	0.13	0.184	0	0	0	0
OCDF	0.0001	0.012	0.019	0.003	0	0	0.022	0.0372
Total PCDFs (fg TEQ/g)		11.66	13.66	6.550	1.372	0.702	11.919	10.797
3,3',4,4'-TCB(#77)	0.0001	0	0	2.48	0	0	0.14	0
3,3',4,4',5-PeCB (#126)	0.1	0	0	0	0	0	0	0
3,3',4,4',5,5'-HxCB (#169)	0.01	0	0	0	0	0	0	0
Total Co-PCBs(fg TEQ/g)		0	0	2.48	0	0	0.14	0
Total pgTEQ/g		0.0166	0.0184	0.0177	0.0015	0.0233	0.0442	0.0126
Congener	Croaker (n=5)	Squid (n=5)	Milk (n=2)	Cheese (n=2)	Beef (n=7)	Pork (n=7)	Chicken (n=5)	Egg (n=5)
2,3,7,8-TCDD	0	0	3.05	0	0	0	0	0.4
1,2,3,7,8-PeCDD	9.64	7.39	3.95	0	3.143	0	0	0.9
1,2,3,4,7,8-HxCDD	0	0	1.51	0	0.1	0.357	0	0.1
1,2,3,6,7,8-HxCDD	0	0	1.41	0	4.514	0.614	0.02	0.58
1,2,3,7,8,9-HxCDD	0	0	0.855	0	0.371	0	0	0.29
1,2,3,4,6,7,8-HpCDD	0.506	0	0.110	0	1.460	0.650	0.006	0.166
OCDD	1.150	1.882	0.001	0.183	0.017	0.031	0.018	2.436
Total PCDDs (fg TEQ/g)	11.296	9.272	10.886	0.183	9.605	1.652	0.044	0.003
2,3,7,8-TCDF	5.216	14.612	0.39	0	0.271	0.8	0	1.58
1,2,3,7,8-PeCDF	1.838	2.926	0.59	0	0.021	0.021	0.03	0.32
2,3,4,7,8-PeCDF	0	2.36	3.35	16.625	5.000	1.643	0.1	5.1
1,2,3,4,7,8-HxCDF	0	0	0.905	0	1.314	1.943	0.02	0.86
1,2,3,6,7,8-HxCDF	0	3.736	0.82	0	0.886	1.100	0	0.4
1,2,3,7,8,9-HxCDF	0	0	0.765	0	0.729	0.286	0.04	0.74
2,3,4,6,7,8-HxCDF	2	0	1.12	0	0	0.243	0	0
1,2,3,4,6,7,8-HpCDF	0.9528	0	0.0885	1.2075	0.311	0.684	0.028	0.17
1,2,3,4,7,8,9-HpCDF	1.7752	0	0.1215	0	0.007	0.107	0.006	0.004
OCDF	0.015	0.04	0.0014	0.0203	0.101	0.002	0.0042	0.010
Total PCDFs (fg TEQ/g)	9.797	23.675	8.1539	17.853	8.541	6.830	0.2282	9.184
3,3',4,4'-TCB(#77)	0.08	0.1	0.6	0.85	0.027	0.713	0	0.061
3,3',4,4',5-PeCB (#126)	0	280	0	150	7.786	2.057	1.32	10.460
3,3',4,4',5,5'-HxCB (#169)	0	0	0	0	0.200	0.439	0.016	0.212
Total Co-PCBs(fg TEQ/g)	0.08	280.1	0.6	150.85	8.013	3.209	1.336	10.733
Total pgTEQ/g	0.0212	0.3131	0.0196	0.1689	0.026	0.012	0.002	0.022

* I-TEF : International Toxic Equivalency factor

* Detection limit : PCDDs/Fs 0.001ppt, Co-PCBs 1ppt (whole weight basis)

Quantification of Dietary Exposure to PCDDs/Fs and Co-PCBs

The estimated dietary daily intake of PCDDs/Fs and Co-PCBs was 0.1757 pg TEQ/kg/day as mean value(Table 3). This value was approximately 10 fold lower than background exposure estimate for PCDDs/Fs of 119 pgTEQ/day (1.7 pg/kg/day, 70kg base) which is exposure level for the United States (North America, 1994) on the basis of diet.

Table 3. Dietary daily intake of PCDDs/Fs and Co-PCBs for Korean population.

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Food	Ingestion Rate (g/day)	Dietary Daily Intake (pgTEQ/kg/day)	Exposure Contribution (%)
Rice	261.7	7.223×10^{-2}	41.11
Barely	5.04	1.547×10^{-3}	0.88
Bean	3.6	1.063×10^{-3}	0.61
Radish	27.3	6.685×10^{-4}	0.38
Korean Cabbage	13.1	5.084×10^{-3}	2.89
Mackerel	10.9	8.037×10^{-3}	4.57
Croaker	10.2	2.140×10^{-3}	1.22
Pollak	6.3	2.223×10^{-3}	1.27
Squid	7.1	3.705×10^{-2}	21.09
Milk	56.2	1.840×10^{-2}	10.47
Cheese	0.2	5.629×10^{-4}	0.32
Beef	31.4	1.368×10^{-2}	7.79
Pork	21.6	3.909×10^{-3}	2.22
Chicken	8.2	2.460×10^{-4}	0.14
Egg	21.6	8.856×10^{-3}	5.04
Total		0.1757	100

The order of high exposure contribution to the total exposure was rice, squid, milk, beef, egg and mackerel. The feature of Korean food consumption pattern is that the ingestion rate of cereal and vegetable is higher than meat and dairy product. It was recognized that the exposure pattern of PCDDs/Fs and Co-PCBs is very different with that of Europe and America.

Risk Assessment

The Korea Food and Drug Administration (KFDA) organized a domestic consultation on the domestic evaluation of the national Tolerable Daily Intake (TDI) and assessment of the health risk of dioxins to dietary exposure in last year. KFDA recommended tentatively TDI value as 4 pg/kg/day through review of revised WHO TDI. Although the estimated dietary intake based on this results is except air inhalation and soil ingestion level, current exposure level (0.17pg/kg/day) is much lower than revised WHO TDI, 1-4 pg/kg/day. Then, we can suggest that the occurrence of the hormonal, reproductive and developmental effect, which is toxicological endpoint of revised WHO TDI due to current dietary exposure of PCDDs/Fs and Co-PCBs, would not be expected.

Acknowledgement

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