A TOTAL DIET STUDY TO ESITAMATE DIOXIN-LIKE COMPOUNDS INTAKE FROM TAIWAN FOOD

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Introduction

Food is the major route of human intake of toxic dioxin-like compounds (DLCs), which include PolyChlorinated Dibenzo-*p*-Dioxins (PCDDs), PolyChlorinated Dibenzo-*p*-Furans (PCDFs), and PolyChlorinated Biphenyls (PCBs). Approximately 95% of human DLCs exposure derives from food, with nearly 80% coming from food of animal origin. The DLCs levels in foodstuffs and the food consumption rate are essential to evaluate health risk posing to humans. The lack of DLCs levels in food increases the population's risk to DLCs exposure. The Department of Health, Taiwan has entrusted us to conduct a comprehensive monitoring program on PCDD/Fs levels in Taiwan food (not including plant origin) in 2001[1] and 2002, In 2003, the program has extended the analytes to include 12 WHO-PCBs[2]. A total diet study (TDS) of DLCs intake from Taiwan food is, therefore, conducted for the first time. The DLCs concentrations in food of animal origin and the food consumption rate are collected. The average daily intake (ADI) and average weekly intake (AWI) of DLCs from food by Taiwan adults is determined.

Materials and Methods

Sample collection and preparation: The TDS was carried out from May to September in 2003. Foodstuffs were purchased from supermarkets and traditional markets located all over Taiwan. A total of 125 compound food samples were prepared by assembling from 1803 individual food samples. A total of 18 pork samples, 12 beef samples, 4 mutton samples, 12 chicken samples, 4 duck samples, 2 goose samples, 20 saltwater fish samples, 4 freshwater fish samples 4 shellfish samples, 19 milk samples, 12 diary product samples, 8 fat and oil samples, 6 egg samples were investigated in this TDS. Sample preparation and extraction were accordingly adjusted, depending on matrix type. The edible portions of all samples, except dairy products, were homogenized with a mechanical blender. About 2 to 100 grams test portions (depending on the lipid content) were used. The samples were pre-treated and analyzed using the Chinese National Standard (CNS) method of test for residual dioxins and dioxin-like PCBs in foods (CNS 14758)[3].

Instrumental analysis: Samples were analyzed for the 17 2,3,7,8-substituted PCDD/Fs and 12 WHO-PCBs. The spiked concentrates were analyzed using an Agilent 6890N high resolution gas chromatograph coupled with a Micromass Autospec NT high resolution mass spectrometer (HRGC/HRMS). The analysis of 17 PCDD/Fs congener and 12 WHO-PCBs was performed

separately with a DB-5MS capillary column (60 m × 0.25 mm i.d. × 0.25μm film thickness), dynamic mass resolution greater than 10000 was reached, and isotope dilution method was used for quantitation. The LOD for tetra~penta-chlorinated PCDD/Fs, hexa~hepta-chlorinated PCDD/Fs, octa-chlorinated PCDD/Fs, and WHO-PCBs were 01. pg, 0.2 pg, 0.5 pg, and 0.2 pg, respectively. The toxic equivalents (TEQ) were calculated using WHO-TEF system[2]. The concentrations of the non-detected congeners were calculated with zero (lower-bound concentration), half the limit of determination (LOD) and the LOD (upper-bound concentration), respectively.

Results and discussion

Food consumption rate and daily intake of DLCs: The food consumption rate[4] and daily intake of DLCs by Taiwan male adults in various food groups, all of animal origin, are listed in Table 1. Roughly 40% is from pork and 13% is from fish. The total food consumption rate is **426.4 g/day.** Saltwater fish possesses the major DLCS exposure, i.e., about 31%, followed by the freshwater fish, i.e., about 14%. The contribution from pork is about 8%. The high TEQ contribution from fish is mostly due to their high DLCs concentration, rather than their larger consumption rate.

It is noted that the daily intake of DLCs does not differ much between the ND=0, ND=½LOD, and ND=LOD values for most samples, indicating the use of larger amount of sample size and fine tune of the HRMS would lower the detection limits. This is beneficial when analyzing food samples which are generally low in DLCs. The uncertainty caused by the magnifying effect when using LOD to replace the concentrations of non-detected congeners could be reduced. The data fulfills the QA/QC requirements by the CNS method 14758. In addition, we have used the method to participate 2003 Inter-laboratory Comparison on Dioxins in Food. The Z-scores of 17 PCDD/Fs are all smaller than 1. The results for PCB-77, PCB-81, PCB-126, and PCB-169 are all being accepted (except PCB-81 in salmon).

Table 1: Food consumption rate by Taiwan male adults and intake of DLCs

Groups	Consumption rate (g/day)	DLCs Mean±SD (pg WHO-TEQ/day)		
		ND=0	ND=½ LOD	ND=LOD
Pork	126.2 ± 182.8	7.25 ± 10.8	7.29 ± 10.9	7.29 ± 10.9
Beef	7.9 ± 36.3	1.04 ± 4.90	1.05 ± 4.89	1.05 ± 4.90
Other livestock	3.3 ± 27.5	0.333 ± 2.78	0.333 ± 2.78	0.333 ± 2.78
Chicken	52.2 ± 137.0	3.80 ± 10.4	3.86 ± 10.3	3.86 ± 10.4
Duck	9.2 ± 52.0	2.67 ± 15.1	2.67 ± 15.1	2.67 ± 15.1
Other fowl	3.8 ± 30.8	4.02 ± 32.8	4.02 ± 32.8	4.02 ± 32.8
Freshwater fish	29.0 ± 141.6	13.4 ± 66.3	13.4 ± 66.3	13.4 ± 66.3
Saltwater fish	34.4 ± 111.1	29.6 ± 105.7	29.6 ± 105.6	29.6 ± 105.7
Shellfish	28.1 ± 89.2	4.05 ± 15.2	4.26 ± 14.3	4.26 ± 15.2
Milk	55.9 ± 156.3	1.85 ± 5.28	1.85 ± 5.27	1.85 ± 5.28
Diary products	28.0 ± 78.2	2.50 ± 7.62	2.55 ± 7.49	2.55 ± 7.62
Fat	3.0 ± 18.0	5.09 ± 31.4	5.15 ± 31.1	5.15 ± 31.4
Oil	11.1 ± 21.9	13.3 ± 28.0	13.6 ± 27.4	13.6 ± 28.0
Egg	34.3 ± 50.0	6.87 ± 10.8	6.90 ± 10.7	6.90 ± 10.8
Total	426.4	95.8 ± 138.3	96.5 ± 138.7	96.5 ± 139.1

Similar phenomenon is observed in results from Taiwan female adults. Table 2 lists the food consumption rate[4] and daily intake of DLCs by Taiwan female adults in various food groups, all of animal origin. The total food consumption rate is 297.4 g/day, which is about 70% to that of Taiwan male adults. The daily intake of DLCs by Taiwan female adults is 73.9 pg WHO-TEQ/day, which is

about 77% to that of Taiwan male adults.

Table 2: Food consumption rate by Taiwan female adults and intake of DLCs

Groups	Consumption rate (g/day)	DLCs Mean±SD (pg WHO-TEQ/day)		
		ND=0	ND=½ LOD	ND=LOD
Pork	88.2 ± 138.0	5.06 ± 8.13	5.09 ± 8.16	5.12 ± 8.20
Beef	3.1 ± 19.5	0.41 ± 2.60	0.41 ± 2.61	0.41 ± 2.61
Other livestock	1.3 ± 14.4	0.13 ± 1.45	0.13 ± 1.45	0.13 ± 1.46
Chicken	31.1 ± 129.2	2.27 ± 9.47	2.30 ± 9.61	2.34 ± 9.76
Duck	3.5 ± 18.6	1.02 ± 5.40	1.02 ± 5.40	1.02 ± 5.40
Other fowl	2.7 ± 28.5	2.86 ± 30.3	2.86 ± 30.3	2.86 ± 30.3
Freshwater fish	19.3 ± 82.1	8.90 ± 38.6	8.91 ± 38.6	8.91 ± 38.6
Saltwater fish	21.1 ± 56.6	18.2 ± 56.0	18.2 ± 56.0	18.2 ± 56.0
Shellfish	28.5 ± 121.8	4.11 ± 18.1	4.32 ± 19.1	4.55 ± 20.1
Milk	28.3 ± 78.7	1.87 ± 5.31	1.87 ± 5.31	1.87 ± 5.31
Diary products	28.3 ± 78.7	2.53 ± 7.41	2.58 ± 7.54	2.63 ± 7.67
Fat	2.8 ± 14.1	4.75 ± 24.1	4.81 ± 24.4	4.86 ± 24.7
Oil	12.5 ± 21.6	15.0 ± 26.5	15.3 ± 27.1	15.7 ± 27.8
Egg	26.7 ± 49.6	5.35 ± 10.3	5.37 ± 10.4	5.40 ± 10.4
Total	297.4	72.4 ± 86.8	73.2 ± 87.3	73.9 ± 87.9

Estimation of average daily and weekly intake of DLCs: Table 3 lists the average daily and weekly dietary intake of DLCs from foods of animal origin by Taiwan adults. The upper-bound dietary intake of DLCs is **1.62 ± 2.32** and **1.23 ± 1.46** pg WHO-TEQ/kg b.w./day, for male and female adults, respectively. The ADI of DLCs from foods of animal origin by Taiwan adults is within the tolerable daily intake (TDI), i.e., 1-4 pg WHO-TEQ/kg b.w./day, recommended by WHO in 1998. Recently, the Scientific Committee on Food (SCF) of European Communities proposed a tolerable weekly intake (TWI) for dioxins and dioxin-like PCBs of 14 pg WHO-TEQ/kg b.w./week. The upper-bound dietary intake of DLCs of **11.4 ± 16.2** and **8.62 ± 10.3** pg WHO-TEQ/kg b.w./week, for Taiwan male and female adults, respectively, is also within the recommended TWI. However, the large standard deviation, which is mainly from the uncertainty associated with food consumption rate, indicates that a small portion of the population might exceed the TDI or WDI because of dietary habits. It is also noted that current study does not include foods of plant origin. The intake values estimated in this study are therefore under estimated.

Table 3: Intake of DLCs from foods of animal origin by Taiwan adults

Sex	Average daily intake of DLCs, assuming 70kg male adult weight, 60kg female adult weight (pg WHO-TEQ/kg b.w./day)		
	ND=0	ND=1/2 LOD	ND=LOD
Male	1.60 ± 2.31	1.61 ± 2.31	1.62 ± 2.32
Female	1.21 ± 1.45	1.22 ± 1.46	1.23 ± 1.46
	Average weekly intake of DLCs, assuming 70kg male adult weight, 60kg female adult weight (pg WHO-TEQ/kg b.w./week)		
	ND=0	ND=1/2 LOD	ND=LOD
Male	11.2 ± 16.1	11.3 ± 16.2	11.4 ± 16.2
Female	8.45 ± 10.1	8.53 ± 10.2	8.62 ± 10.3

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