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Human dietary exposure to polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in Taiwan

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ABSTRACT

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) levels in a total of 25 food items in Taiwan were surveyed. It was observed that shellfish and saltwater fish possessed the highest PCDD/Fs levels, 9.82 and 3.60 pg WHO-TEQ/g, respectively, on the lipid basis. The dietary intakes of humans at the ages of 12–18, 19–64, and over 65 were determined. The estimated intake were between 21.8 pg (female teenagers) and 37.6 pg (male seniors) WHO-TEQ/day; the levels varied with the dietary habits. The PCDD/F intakes for all human groups are far below the tolerable limit of 70 pg WHO-TEQ/kg b.w./month. In addition, the daily PCDD/F intake levels for duck-farmers consuming average and large amounts of PCDD/F contaminated duck eggs were examined. The result shows that consuming more than one duck egg with level higher than 10 pg WHO-TEQ/g lipid of PCDD/Fs per day could lead to a PCDD/F intake level higher than the tolerable limit. However, for normal population, there is a little risk to ingest intolerable amount of PCDD/Fs because of consuming contaminated duck eggs.

1. Introduction

Polychlorodibenzo-*p*-dioxin and polychlorodibenzofurans (PCDD/Fs) have received increasing attention because of their important toxicity and carcinogenic potential. In December 1990 the World Health Organization (WHO) established a tolerable daily intake (TDI) of 10 pg/kg b.w. (body weight) for TCDD on human, based on many toxicity data on experimental animals and kinetic data on human and animals. With the epidemiological and toxicological data, re-evaluation of 1–4 pg TEQ/kg b.w. as TDI was then conducted in 1998 [1,2]. Most recently in 2001, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) examined new evidence on the toxicity of these chemicals and established a Provisional Tolerable Monthly Intake (PTMI) of 70 pg of dioxins and dioxin-like PCBs [3], and the Scientific Committee on Food of the European Commission also established a 14 pg WHO-TEQ/kg b.w./day of tolerable intake on a weekly basis in 2000 [4].

Among the routes through which human exposes to PCDD/Fs, i.e., inhalation, dermal absorption, soil ingestion, and diet, the diet route is recognized to be the main course of intake. Currently, many studies focused on the contribution of PCDD/Fs in diet to the health risk and found that PCDD/Fs in meat contributed the most. About 50% of the dietary intake of PCDD/Fs by the U.S. population was from meat and dairy products [5]. For the population in Tarragona, Spain, marine foods and lipids (including oils and fats) were accounted for 33.7 and 15.3%, respectively. Of the daily dietary intake, these values obtained in recent years were lower than those before 2000 [6]. The same trend was also found in Japan that the dietary intake of 1.55 pg TEQ/kg/day PCDD/Fs in 2004 was lower than that of 2.18 pg TEQ/kg/day in 1999 [7].

In Taiwan, Chen et al. [8,9] conducted a study to correlate the consumption frequency of different food groups and the level of serum PCDD/Fs. The consumption of fish was observed to be positive correlation to the level of serum PCDD/Fs in both of the studies. For seniors in, Chen et al. [9] even found the consumption of tofu was negatively correlated with PCDD/F serum levels. Hung et al. [10] also determined the positive correlation between fishes and the pregnant women. In this study, a detail evaluation of dietary intake of PCDD/F was reported.

In 1998 in Belgian incident involved a contaminated feed and the contaminated chicken showed a level of PCDD/Fs 100 times above the recommended limit [11]. A similar incident happened in Taiwan in 2006 and the duck eggs showed a PCDD/F level higher than 30 pg TEQ/g lipid. In this study, the PCDD/F concentrations

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of a variety of foods, such as duck eggs, fruit, and bread were analyzed, and the total daily intakes were estimated to supplement the data on dietary intake of PCDD/Fs in Taiwan. Although most people in Taiwan consume fewer duck eggs than chicken eggs, the duck-farmers usually consume a large amount of duck eggs from their own farms and thus may take intolerable quantities of PCDD/Fs. Therefore, the diet intake of PCDD/Fs by duck-egg farmers was also determined to assess their daily intake levels.

2. Materials and methods

2.1. Sampling

Samples of foods were randomly chosen from supermarkets and traditional markets at two different locations in southern Taiwan for testing of PCDD/Fs. Most of the vegetables, fruits, and freshwater farm fishes are cultured in southern Taiwan. The supermarkets chosen in this study were the chain stores which supplied the same foods to all branches in Taiwan.

A total of 59 food samples including 18 major kinds of foods were selected based on the Nutrition and Health Survey in Taiwan (NAHSIT) conducted between 1993 and 1996 [12]. The selected foods included cereals (rice), vegetables (cabbage and water celery), fresh fruits (apple, banana, and pineapple), meats (pork and beef), poultry (chicken and duck), freshwater fishes (mouthbreeder and milkfish), saltwater fishes (grouper), shellfishes (shrimp, oyster, and clam), protein-rich foods (eggs, milk, cheese, soybean and soybean product), sauces and miscellaneous foods (instant noodle, bread).

2.2. PCDD/F analysis

Food samples were analyzed by using US EPA method 1613B by the Super Micro Mass Research and Technology Center of Cheng Shiu University [13]. Samples were homogenized, spiked with $^{13}C_{12}$ internal standards and Soxhlet extracted with toluene. The extracts were then cleaned up with silica gels (acid and basic), alumina, and activated carbon columns, according to method 1613. The final extracts were concentrated to about 1 ml by rotary vacuum concentrators, further concentrated to near dryness by evaporation with nitrogen blowing, and spiked with the internal standards prior to analysis by high resolution gas chromatography (HRGC) coupled with high resolution mass spectrometry (HRMS).

Recoveries of internal standards, errors of duplicate analyses and detections of blank samples were all within the analytical standard method [13]. The ranges of limits of detection (LOD) of the 17 congeners of PCDD/Fs are shown in Table 1.

2.3. Estimation of daily intake of PCDD/Fs

The food consumption data for persons of 19–64 years old, the body with average weight of 64.8 kg for male and 56.3 kg for female adult were obtained from the NAHSIT [12]. The toxic equivalency (TEQ) data of 17 PCDD/Fs congeners were determined with respect to the 2,3,7,8-TCDD toxicity (TEFs) [14]. The PCDD/F dietary intake was calculated based on the products of multiplying the daily consumption by the mean TEQ of PCDD/Fs for each food type. The PCDD/F concentrations of non-detected PCDD/F congeners are assumed to be half of the respective limits of detection.

3. Results and discussion

The concentrations of 17 PCDD/F congeners in 25 food items were given in Table 2. The observed PCDD/F levels in meats, eggs, diary products, seafood and oils are on the lipid weight basis; vegetables, fruits, soybean foods, breads, and instant noodles were given on the fresh-weight basis. All 17 congeners detected in all food groups are lager than the LOD except for those in vegetables and fruits.

Table 1
Ranges of limits of detection (LOD) of the 17 congeners of PCDD/Fs

Congeners	LOD (pg/g fresh weight)
2,3,7,8-TeCDD	0.042–0.294
1,2,3,7,8-PeCDD	0.085–0.297
1,2,3,4,7,8-HxCDD	0.054–0.628
1,2,3,6,7,8-HxCDD	0.051–0.336
1,2,3,7,8,9-HxCDD	0.042–0.495
1,2,3,4,6,7,8-HpCDD	0.058–0.585
OCDD	0.185–1.263
2,3,7,8-TeCDF	0.091–0.4
1,2,3,7,8-PeCDF	0.095–0.396
2,3,4,7,8-PeCDF	0.086–0.449
1,2,3,4,7,8-HxCDF	0.06–0.301
1,2,3,6,7,8-HxCDF	0.059–0.495
2,3,4,6,7,8-HxCDF	0.069–0.327
1,2,3,7,8,9-HxCDF	0.065–0.461
1,2,3,4,6,7,8-HpCDF	0.072–0.544
1,2,3,4,7,8,9-HpCDF	0.071–0.322
OCDF	0.115–0.922

The observed PCDD/F levels of meats on the lipid basis in an increasing order are: pork (0.253 pg WHO-TEQ/g), chicken (0.39 pg WHO-TEQ/g), beef (0.551 pg WHO-TEQ/g) and duck (434 pg WHO-TEQ/g), and those on the fresh-weight basis show the same trend. Among fish and shellfish, the levels of three kinds of shellfishes (oysters, clams, and shrimps) are comparatively higher than those of fishes (mouthbreeder, milkfish, and grouper) on the lipid basis as shown in Table 2. However, all fish and shellfish show relatively comparable PCDD/F levels (mouthbreeders 0.087 pg WHO-TEQ/g, milkfish 0.178 pg WHO-TEQ/g, groupers 0.143 pg WHO-TEQ/g, oysters 0.173 pg WHO-TEQ/g, clams 0.108 pg WHO-TEQ/g, and shrimps 0.079 pg WHO-TEQ/g) when expressed on the fresh-weight basis. For all food items, concentrations of OCDD were the highest among the 17 PCDD/F congeners.

A number of other studies focused on determining PCDD/F levels in foods. Llobet et al. [15] measured the concentration of 59 kinds of foods from retailed stores and brands/trademarks. The oils and fats showed the highest WHO-TEQ values on the fresh-weight basis (0.223 pg/g), and the seafood and fish (0.131 pg/g) were followed, while fruits (0.003 pg/g) and vegetables (0.09 pg/g) showed the lowest concentrations.

In Taiwan, Chen et al. [16] examined 37 different cooked foodstuffs (including meat, fish, milk and diary product, oil, and eggs) from markets located in eight cities of counties. The maximum total PCDD/Fs level corresponded to duck meat (0.182 pg WHO-TEQ/g lipid weight) and needle fishes (0.185 pg WHO-TEQ/g lipid weight), followed by goldfishes and orbfishes, while the lowest levels were detected in the nonfat milk (0.006 WHO-TEQ/g wet weight). Furthermore, PCDD/Fs levels in animal fat were notably higher than those in vegetable oil. Another study in Taiwan [17] in which the PCDD/F levels of 14 food groups (excluding vegetables, cereals, and fruits) were analyzed also found the highest level in fishes.

Other studies in recent years in France [18], in Belgium [19], and in China [20], similar to this study, all indicated the highest PCDD/Fs levels in fishes and lowest levels in vegetables, fruits, and cereals. Additionally, duck level was higher than other meat in this study and in the previous one by Chen et al. [16], which should be noteworthy, especially in Asia.

Average PCDD/F concentrations, consumption rates of the adult population, and estimated of PCDD/F dietary intakes via all food items are shown in Table 3. The dietary intakes of