Journal of Food and Drug Analysis

Volume 20 | Issue 4 Article 12

Dietary intake of PCDD/Fs and dioxin-like PCBs from fresh foods around Taiwan

Recommended Citation
Chang, J.-W.; Liao, P.-C.; and Lee, C.-C. (2012) "Dietary intake of PCDD/Fs and dioxin-like PCBs from fresh foods around Taiwan," *Journal of Food and Drug Analysis*: Vol. 20 : Iss. 4 , Article 12.
Available at: https://doi.org/10.6227/jfda.2012200409

Journal of Food and Drug Analysis, Vol. 20, No. 4, 2012, Pages 805-813
doi:10.6227/jfda.2012200409

Dietary Intake of PCDD/Fs and Dioxin-Like PCBs from Fresh Foods around Taiwan

JUNG-WEI CHANG[1], PO-CHI LIAO[1,2] AND CHING-CHANG LEE[1,2]*

[1.] *Department of Environmental and Occupational Health, National Cheng Kung University College of Medicine, Tainan, Taiwan, R.O.C.*
[2.] *Research Center of Environmental Trace Toxic Substance, National Cheng Kung University, Tainan, Taiwan, R.O.C.*

(Received: February 13, 2012; Accepted: July 30, 2012)

ABSTRACT

This study investigated the background levels of PCDD/Fs and dioxin-like PCBs (dl-PCBs) in fresh foods around Taiwan. In 1,029 foodstuffs, the highest PCDD/F level based on per gram fat was found in duck eggs (1.956 pg WHO-TEQ/g fat), followed by beef (1.263 pg WHO-TEQ/g fat), and egg products (1.067 pg WHO-TEQ/g fat), and the lowest was in grape seed oil (0.068 pg WHO-TEQ/g fat). The highest dl-PCB level was found in beef (0.782 pg WHO-TEQ/g fat), followed by duck eggs (0.632 pg WHO-TEQ/g fat), and mutton (0.506 pg WHO-TEQ/g fat), and the lowest was in peanut oil (0.011 pg WHO-TEQ/g fat). The average intake of boys and girls (> 6, ≤ 12 years old) were 0.70 and 0.62 pg $WHO\text{-}TEQ_{PCDD/Fs+dl\text{-}PCBs}$/kg bw/day, respectively; for adolescents (> 13, ≤ 18), 0.34 (male) and 0.30 (female) pg $WHO\text{-}TEQ_{PCDD/Fs+dl\text{-}PCBs}$/kg bw/day, respectively; for adults (19-64), 0.33 (male) and 0.31(female) pg $WHO\text{-}TEQ_{PCDD/Fs+dl\text{-}PCBs}$/kg bw/day, respectively; and for seniors (> 65), 0.42 (male) and 0.37 (female) pg $WHO\text{-}TEQ_{PCDD/Fs+dl\text{-}PCBs}$/kg bw/day, respectively. The greatest contribution (%) to the total daily intake came from meat fish, and fishery products, especially in adults (men: 51.6%; women: 47.5%) and seniors (men: 64.8%; women: 61.8%). In the Hsinchu-Miaoli area, PCDD/F concentrations were found the highest in beef and mutton, while in Keelung-Taipei-Taoyuan area the highest in mutton, duck, and goose. The distribution of dl-PCB concentrations in beef, mutton, and goose meat throughout Taiwan showed a similar trend with PCDD/Fs. The mean dioxin level in milk concurrently decreased with total dioxin emissions in Taiwan between 2004 and 2008. It is concluded that, generally, PCDD/Fs and dl-PCBs in food pose little health risk in Taiwan, except for occasionally high PCDD/F levels in beef and mutton. These data suggested that the environment near where the livestock was raised should be examined.

Key words: background levels, PCDD/Fs, dioxin-like PCBs, foods, daily intake

INTRODUCTION

Sources of human exposure to dioxins include food, drinking water, air inhalation, and skin contact. Dietary intake is by far the most important and accounts for over 90% of the exposure of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs, dioxins), and polychlorinated biphenyls (PCBs). Fatty foods such as meat, poultry, seafood, milk, eggs, and their products are the major dietary sources of dioxins(1). Many different matrices, such as milk, eggs, meat, fish, and animal feed, have been investigated. Data concerning background levels of PCDD/Fs and PCBs present in these matrices are now available and can be used to estimate typical dietary intakes for the general population(2).

A subset of dioxins and dioxin-like PCBs (dl-PCBs), comprising 17 laterally substituted PCDD/Fs and 12 non-ortho- and mono-ortho-chlorine-substituted PCBs, induce a similar spectrum of biological effects and toxic responses that are mediated through the aryl hydrocarbon (Ah) receptor, which has been the basis for establishing the toxic equivalency factors (TEF) and total toxic equivalency (TEQ) scheme(3). The World Health Organization (WHO) has set up a tolerable daily intake (TDI) range of 1-4 pg TEQ/kg of body weight (bw) for dioxins(4). Likewise, a tolerable weekly intake (TWI) of 14 pg WHO-TEQ/kg bw has been determined by the European Union (EU) through the European Commission Scientific Committee on Food (SCF) (European Commission, 2001a)(5), and a strategy to reduce human intake levels to below this threshold has been implemented. As part of this strategy, and to prevent health risks from exposure to PCDD/Fs and dl-PCBs, maximum levels for dioxins and for the sum of dioxins and dl-PCBs in foodstuffs of animal origin and vegetable oils(6), as well as target and action levels, have been

* Author for correspondence. Tel: +886-6-275-2484;
Fax: +886-6-274-3748; E-mail: cclee@mail.ncku.edu.tw

established in the EU to encourage a proactive approach to reduce the dioxins and dl-PCBs present in food[7]. Although dietary dioxin intake was determined in a number of countries in the 1990s, most surveys were focused only on those food groups in which the highest levels of PCDD/PCDFs could be expected. From 2004 to 2008, the Taiwan FDA conducted joint national surveys of dioxins and dl-PCBs in livestock, poultry, milk, eggs, oils, fish, fruit, and vegetables to obtain the background levels in these products and the primary source of their contamination. The present study is one of the limited surveys in which all food groups in generalized diets have been included. One objective of this study was to identify possible local sources of contamination. Moreover, the results were compared to the TDI for dioxins by different official organizations. We also evaluated whether there existed a trend with milk dioxin levels over time. Finally, we determined food groups which make the largest contributions to dietary exposure in different age groups.

MATERIALS AND METHODS

I. *Food Sampling*

The Taiwan background food survey (TBFS) is an ongoing market-basket survey of approximately 41 core foods in the Taiwan food supply to determine levels of PCDD/Fs in foods. The planning of TBFS samples was based on the official food classification (11 groups) and data on food consumption for 1993 through 1996 by the general population were obtained from the Nutrition and Health Survey in Taiwan (NAHSIT)[8].

The sampling was conducted by a step-by-step process. First, 6 sampling areas in Taiwan were grouped according to the ambient air dispersion area defined by the Taiwan EPA: the Keelung-Taipei-Taoyuan (KTT) area, Hsinchu-Miaoli (HM) area, Taichung-Changhua-Nantou (TCN) area, Yunlin-Chiayi-Tainan (YCT) area, Kaohsiung-Pingtung (KP) area, and Yilan-Hualien-Taitung (YHT) area. Second, the quantity of production of each foodstuff was collected and evaluated in every county, village, and town in each area. Third, the foods produced in the greatest quantities in each county were selected for analysis. The foodstuff samples were purchased from traditional markets or supermarkets in selected towns around Taiwan from 2004 to 2008. Finally, we used over 1029 individual foods in the five years to prepare samples. All group samples were adequately homogenized, and then frozen at −20 °C until analysis. For example, a pork composite sample weighing 600 g was prepared by homogenizing 10 aliquots of 60 g of homogenized pork, each from separate pork samples of ca. 500-1,000 g. We investigated samples of pork (31), beef (38), mutton (45), livestock and poultry products (65), chicken (31), duck (35), goose (35), large marine fish (34), small-medium marine fish (58), freshwater fish (22), other seafood (41), fishery and seafood products (66), milk (127), dairy products (21), fat and oil (16), eggs (100), fruit (27), vegetables (197), and cereal (40).

II. *HRGC/HRMS Analysis of PCDD/Fs/dl-PCBs*

Isotope dilution high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS) was employed to measure 17 PCDD/Fs and 12 dl-PCBs in livestock, poultry, fish, seafood, eggs, milk, dairy products, and oil samples, as previously described[9]. Analytical procedures were adopted from USEPA Method 1613B[10] and USEPA Method 1668A[11] with minor modifications. The QA/QC protocols included MS Resolution, GC Resolution, calibration verification, ongoing precision and recovery, blank, and internal standard recovery. The Analytical Laboratory for Trace Environmental Pollutants, Research Center of Environmental Trace Toxic Substances, at National Cheng Kung University in Taiwan was certified by the Taiwan Accreditation Foundation (TAF). The PCDD/F and dl-PCB concentrations were given as pg WHO-TEQ/g fat and pg WHO-TEQ/g wet weight.

III. *Dioxin Intake Estimates*

In the intake calculations, the average daily consumption of each food was multiplied by the corresponding concentrations. Daily intakes (pg/day) for PCDD/Fs and dl-PCBs were calculated on a fresh-weight basis as a sum of the individual food. Intakes were calculated with upper-bound concentrations. Exposure was calculated for both PCDD/Fs and dl-PCBs. For calculations, when a congener concentration was under the limit of detection (LOD), the value was assumed to be its LOD (upper-bound approach). The TEQ data of the 17 PCDD/Fs and 12 dl-PCBs congeners were determined with respect to 2,3,7,8-TCDD toxic equivalency factors (TEFs)[3].

The dietary intake of PCDD/Fs and dl-PCBs was first calculated based on the products of multiplying the daily consumption by the mean TEQ of PCDD/Fs and dl-PCBs for each food type. To further calculate daily intake pg/kg bw, the average weight of the members of each gender and age group were used; it was also obtained from the NAHSIT[8].

IV. *Contribution Analysis*

To determine the extent which different food categories contribute to the total dioxin intake, TBFS foods were divided into the same categories as defined in NAHSIT: meat, fruits & vegetables, fish, oils, eggs, dairy, poultry, and other[8]. The contributions to the total PCDD/F and dl-PCB intake from each food category were calculated as a percentage based on the total intake (summed TEQ × food consumption) for each category.

V. *Geographical and Time-Trend Analysis*

Additional analyses were conducted to facilitate comparisons of the total dioxin intake for the purpose of identifying any time trends or geographical differences. For the time-trend analysis, only foods, PCDD/Fs, and dl-PCBs that have been analyzed for all 5 years were evaluated. The intake estimates obtained in this manner were compared for each year to determine whether any changes over time occurred. For the geographical analysis of variation, only meat, PCDD/Fs, and dl-PCBs that have been analyzed in all 6 sampling areas in Taiwan were evaluated.

RESULTS AND DISCUSSION

I. *PCDD/F and dl-PCB Concentration Levels in Food Groups*

The highest PCDD/F levels based on per gram fat were found in duck eggs (1.956 pg WHO-TEQ/g fat), followed by beef (1.263 pg WHO-TEQ/g fat), and egg products (1.067 pg WHO-TEQ/g fat), and the lowest level was found in grape seed oil (0.068 pg WHO-TEQ/g fat) (Table 1). The average PCDD/F levels based on per gram fresh weight were in seafood (0.422 pg WHO-TEQ/g fresh weight), large marine fish (0.355 pg WHO-TEQ/g fresh weight), small marine fish (0.106 pg WHO-TEQ/g fresh weight), and freshwater fish (0.099 pg WHO-TEQ/g fresh weight), and lowest in melons (0.003 pg WHO-TEQ/g fresh weight). The highest dl-PCB levels based on per gram fat were in beef (0.782 pg WHO-TEQ/g fat), followed by duck eggs (0.632 pg WHO-TEQ/g fat), mutton (0.506 pg WHO-TEQ/g fat), and the lowest level was in peanut oil (0.011 pg WHO-TEQ/g fat) (Table 1). In our study, the overall TEQ levels of PCDDs and PCDFs

Table 1. Distribution of PCDD/Fs and dioxin-like PCBs in Taiwan foods

		pg WHO-TEQ/g, fat		pg WHO-TEQ/g, wet weight	
Food	N	PCDD/Fs	dl-PCBs	PCDD/Fs	dl-PCBs
Livestock					
Pork	31	0.100 ± 0.060 (0.043-0.281)	0.092 ± 0.086 (0.014-0.384)	0.027 ± 0.020 (0.008-0.092)	0.027 ± 0.031 (0.002-0.134)
Beef	38	1.263 ± 1.191 (0.061-6.006)	0.782 ± 0.788 (0.012-3.382)	0.091 ± 0.128 (0.010-0.522)	0.057 ± 0.090 (0.002-0.435)
Mutton	45	0.939 ± 1.045 (0.084-4.006)	0.506 ± 0.696 (0.016-2.879)	0.154 ± 0.307 (0.004-1.475)	0.084 ± 0.152 (0.001-0.789)
Livestock products	54	0.162 ± 0.131 (0.057-0.718)	0.076 ± 0.062 (0.018-0.362)	0.018 ± 0.008 (0.007-0.046)	0.010 ± 0.007 (0.001-0.032)
Poultry					
Chicken	31	0.405 ± 0.432 (0.099-2.405)	0.187 ± 0151 (0.047-0.828)	0.031 ± 0.052 (0.007-0.303)	0.012 ± 0.008 (0.005-0.036)
Duck	35	0.639 ± 0.347 (0.207-1.729)	0.334 ± 0.443 (0.059-2.798)	0.029 ± 0.021 (0.009-0.085)	0.015 ± 0.017 (0.003-0.084)
Goose	35	0.445 ± 0.222 (0.091-1.042)	0.257 ± 0.285 (0.084-1.810)	0.044 ± 0.027 (0.011-0.132)	0.024 ± 0.015 (0.005-0.067)
Poultry products	11	0.784 ± 0.911 (0.083-2.430)	0.424 ± 0.524 (0.018-1.360)	0.141 ± 0.174 (0.014-0.561)	0.087 ± 0.116 (0.003-0.299)
Milk					
Whole fat milk	103	0.889 ± 0.474 (0.198-2.891)	0.490 ± 0.237 (0.068-1.672)	0.032 ± 0.017 (0.007-0.105)	0.018 ± 0.008 (0.002-0.057)
Whole fat milk powder	5	0.163 ± 0.036 (0.124-0.216)	0.065 ± 0.037 (0.043-0.131)	0.047 ± 0.010 (0.035-0.062)	0.019 ± 0.011 (0.012-0.039)
Whole fat sheep milk	19	0.658 ± 0.184 (0.392-1.196)	0.412 ± 0.085 (0.251-0.576)	0.024 ± 0.007 (0.014-0.044)	0.015 ± 0.003 (0.009-0.022)
Dairy products					
Cream	3	0.310 ± 0.108 (0.186-0.379)	0.255 ± 0.133 (0.133-0.397)	0.115 ± 0.040 (0.069-0.141)	0.095 ± 0.049 (0.049-0.147)
Butter	3	0.408 ± 0.066 (0.332-0.446)	0.209 ± 0.027 (0.193-0.241)	0.338 ± 0.052 (0.279-0.375)	0.174 ± 0.019 (0.163-0.195)
Cheese	7	0.321 ± 0.223 (0.127-0.795)	0.229 ± 0.215 (0.045-0.640)	0.081 ± 0.059 (0.029-0.208)	0.059 ± 0.058 (0.010-0.171)
Fermented milk	5	1.018 ± 0.444 (0.353-1.442)	0.422 ± 0.148 (0.228-0.589)	0.030 ± 0.017 (0.008-0.051)	0.013 ± 0.006 (0.005-0.021)
Condensed milk	3	0.690 ± 0.786 (0.212-1.598)	0.204 ± 0.196 (0.045-0.423)	0.062 ± 0.068 (0.021-0.140)	0.019 ± 0.017 (0.004-0.037)
Eggs					
Chicken eggs	32	0.459 ± 0.230 (0.200-1.131)	0.179 ± 0.199 (0.064-1.209)	0.043 ± 0.024 (0.018-0.130)	0.016 ± 0.018 (0.007-0.114)
Duck eggs	39	1.956 ± 4.118 (0.270-23.685)	0.632 ± 0.624 (0.122-3.440)	0.243 ± 0.510 (0.038-3.000)	0.079 ± 0.073 (0.016-0.399)
Egg products	29	1.067 ± 1.190 (0.238-5.118)	0.454 ± 0.305 (0.137-1.368)	0.118 ± 0.124 (0.029-0.616)	0.052 ± 0.033 (0.018-0.159)
Oils					
Soybean oil	2	0.079 ± 0.025 (0.061-0.097)	0.009 ± 0.000 (0.009-0.009)	0.079 ± 0.025 (0.061-0.097)	0.009 ± 0.000 (0.009-0.009)
Peanut oil	6	0.121 ± 0.098 (0.061-0.314)	0.011 ± 0.005 (0.007-0.017)	0.121 ± 0.098 (0.061-0.314)	0.011 ± 0.005 (0.007-0.017)
Grape seed oil	2	0.068 ± 0.044 (0.037-0.100)	0.026 ± 0.030 (0.004-0.047)	0.068 ± 0.044 (0.037-0.100)	0.026 ± 0.030 (0.004-0.047)
Sunflower seed oil	2	0.101 ± 0.011 (0.093-0.109)	0.014 ± 0.002 (0.012-0.015)	0.101 ± 0.011 (0.093-0.109)	0.014 ± 0.002 (0.012-0.015)
Olive oil	2	0.082 ± 0.017 (0.070-0.095)	0.027 ± 0.013 (0.018-0.036)	0.082 ± 0.017 (0.070-0.095)	0.027 ± 0.013 (0.018-0.036)
Pork fat	2	0.165 ± 0.023 (0.149-0.181)	0.044 ± 0.004 (0.041-0.046)	0.165 ± 0.023 (0.149-0.181)	0.044 ± 0.004 (0.041-0.046)

Table 1. Continued

Food	N	pg WHO-TEQ/g, fat		pg WHO-TEQ/g, wet weight	
		PCDD/Fs	dl-PCBs	PCDD/Fs	dl-PCBs
Fish					
Large marine fish	34	3.912 ± 5.813 (0.507-30.318)	10.754 ± 15.230 (0.798-84.028)	0.355 ± 0.773 (0.00-4.003)	0.953 ± 2.061 (0.003-11.095)
Small marine fish	58	4.070 ± 5.708 (0.385-35.181)	11.642 ± 17.885 (0.209-118.456)	0.106 ± 0.153 (0.005-0.933)	0.328 ± 0.524 (0.002-2.712)
Freshwater fish	22	1.784 ± 2.277 (0.558-10.308)	2.049 ± 1.378 (0.476-5.061)	0.099 ± 0.080 (0.014-0.369)	0.164 ± 0.194 (0.024-0.840)
Fishery products	48	1.126 ± 1.805 (0.076-10.282)	2.146 ± 3.597 (0.037-17.665)	0.068 ± 0.075 (0.005-0.317)	0.183 ± 0.310 (0.003-1.315)
Seafood					
Shellfish	19	3.300 ± 2.765 (0.583-9.773)	5.823 ± 6.722 (0.273-23.705)	0.051 ± 0.043 (0.009-0.167)	0.099 ± 0.128 (0.005-0.485)
Shrimp	9	4.107 ± 2.927 (2.566-11.746)	3.237 ± 2.894 (0.135-9.588)	0.048 ± 0.035 (0.019-0.126)	0.037 ± 0.031 (0.002-0.103)
Crab	7	15.847 ± 11.678 (5.309-39.889)	13.156 ± 11.888 (3.111-34.229)	0.422 ± 0.408 (0.058-1.025)	0.329 ± 0.304 (0.021-0.780)
Molluscsa	6	1.800 ± 0.775 (1.035-3.024)	3.210 ± 2.134 (0.360-5.649)	0.025 ± 0.007 (0.016-0.032)	0.042 ± 0.022 (0.007-0.064)
Seafood products	18	0.512 ± 0.481 (0.099-1.602)	0.482 ± 0.464 (0.052-1.431)	0.027 ± 0.028 (0.007-0.126)	0.024 ± 0.024 (0.004-0.087)
Fruits					
Fruits with peels	12	0.054 ± 0.038 (0.014-0.140)	0.005 ± 0.002 (0.002-0.009)	0.009 ± 0.005 (0.002-0.019)	0.001 ± 0.000 (0.000-0.002)
Fruits without peels	15	0.081 ± 0.076 (0.015-0.258)	0.012 ± 0.012 (0.003-0.048)	0.008 ± 0.006 (0.001-0.019)	0.001 ± 0.001 (0.000-0.003)
Vegetables					
Leafy vegetables	92	0.278 ± 0.426 (0.015-3.194)	0.035 ± 0.033 (0.001-0.157)	0.015 ± 0.031 (0.001-0.271)	0.002 ± 0.002 (0.000-0.006)
Root vegetables	39	0.127 ± 0.286 (0.006-1.700)	0.027 ± 0.065 (0.002-0.358)	0.015 ± 0.023 (0.001-0.112)	0.003 ± 0.007 (0.000-0.035)
Beans	15	0.063 ± 0.053 (0.017-0.208)	0.008 ± 0.006 (0.003-0.023)	0.016 ± 0.008 (0.004-0.030)	0.002 ± 0.002 (0.000-0.008)
Bamboo shoots	9	0.087 ± 0.036 (0.054-0.171)	0.011 ± 0.005 (0.006-0.020)	0.006 ± 0.002 (0.004-0.012)	0.001 ± 0.000 (0.000-0.001)
Melons	18	0.051 ± 0.043 (0.013-0.165)	0.007 ± 0.004 (0.002-0.018)	0.003 ± 0.002 (0.001-0.009)	0.000 ± 0.000 (0.000-0.001)
Mushrooms	24	0.094 ± 0.106 (0.018-0.441)	0.007 ± 0.004 (0.002-0.020)	0.009 ± 0.009 (0.002-0.037)	0.001 ± 0.000 (0.000-0.002)
Cereals[b]					
Cereals	22	0.014 ± 0.008 (0.005-0.032)	0.003 ± 0.002 (0.001-0.008)	0.009 ± 0.006 (0.002-0.026)	0.002 ± 0.001 (0.000-0.005)
Cereal products	18	0.017 ± 0.006 (0.008-0.027)	0.002 ± 0.001 (0.001-0.005)	0.014 ± 0.005 (0.007-0.023)	0.002 ± 0.001 (0.001-0.004)

Note: mean ± standard deviation and minimum to maximum included in parentheses.
[a]Cuttle fish, octopus, squid, and neritic squid; [b]rice, glutinous rice, and corn.

were much higher than that of dl-PCBs. In another words, the contribution ratio of dl-PCBs to total intake is lesser than those of PCDDs and PCDFs.

II. *Estimated Daily, Weekly, and Monthly Intake of PCDD/Fs and dl-PCBs*

Estimations of the food consumption by children, adolescents, adults, and the elderly (≥ 65 years old) were conducted based on a 1993- 1996 investigation by the Nutrition and Health Survey in Taiwan (NAHSIT)[8]. For older children, the daily, weekly, and monthly intakes of PCDD/Fs and dl-PCBs are 0.70 (male) and 0.62 (female) pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/day; 4.89 and 4.36 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/week; 20.95 and 18.7 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/month, respectively (Table 2). For adolescents, they are 0.34 and 0.30 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/day; 2.37 and 2.12 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/week; 10.16 and 9.07 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/month, respectively. For adults, they are 0.33 (male) and 0.31 (female) pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/day; 2.29 and 2.14 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/week; 9.82 and 9.16 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/month, respectively (Table 2). For the elderly, they are 0.42 and 0.37 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/day; 2.97 and 2.57 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/week; 12.74 and 11.03 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/kg bw/month, respectively. It is difficult to compare results of intake estimations reported from different countries due to varying methodologies used for calculation. In these studies, there are significant differences in sampling strategy (including food type and region), values for undetected congeners (0, 1/2LOD or LOD) and methods of studying food consumption. A comparison of the results on total dietary PCDD/F intake of recent reports from a number of countries, and those of the present study, is shown in Table 3. The daily intake of dioxins ranged between 75 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/day in Sweden[12] and 161 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/day in the Japan[13]. The recent Taiwanese TEQ estimates of daily intakes (96.6 for male and 74.1 pg WHO-$TEQ_{PCDD/Fs+dl-PCBs}$/day for female) were within these ranges reported from other countries[12-21]. However, these PCDD/F dietary intake values were observed

Table 2. The average daily dose of PCDD/Fs/dl-PCBs of each food in different age groups

Food groups	Food levels[a]	Average daily dose (pg WHO-TEQ/kg bw/day)							
		6-12 years old		13-18 years old		19-64 years old		> 65 years old	
		male	female	male	female	male	female	male	female
Cereals, grains, tubers and roots		0.025	0.022	0.020	0.015	0.015	0.014	0.017	0.015
Rice and its products	0.007	0.014	0.011	0.011	0.008	0.010	0.007	0.009	0.009
Wheat and its products	0.015	0.007	0.008	0.008	0.006	0.004	0.005	0.007	0.004
Carbohydrate's tubers, roots, and their products	0.006	0.003	0.002	0.001	0.001	0.001	0.001	0.001	0.001
Beans, lotus-seed, chestnut and their Products	0.025	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000
Fats and oils		0.027	0.031	0.012	0.014	0.015	0.019	0.015	0.015
Vegetable oils	0.114	0.024	0.023	0.007	0.008	0.009	0.012	0.011	0.010
Animal fats	0.210	0.003	0.006	0.004	0.005	0.004	0.005	0.002	0.003
Nuts and their products	0.075	0.001	0.001	0.001	0.001	0.001	0.001	0.002	0.002
Poultry and their products		0.034	0.027	0.011	0.012	0.012	0.008	0.007	0.006
Chicken and its products	0.041	0.026	0.023	0.010	0.010	0.008	0.006	0.004	0.004
Duck and its products	0.097	0.007	0.003	0.001	0.002	0.003	0.001	0.002	0.001
Goose and its products	0.068	0.001	0.001	0.000	0.000	0.001	0.001	0.001	0.001
Livestock and their products		0.074	0.066	0.049	0.030	0.040	0.028	0.031	0.021
Pork and its products	0.039	0.048	0.042	0.032	0.021	0.028	0.023	0.024	0.019
Beef and its products	0.133	0.011	0.010	0.011	0.007	0.007	0.004	0.006	0.001
Mutton and its products	0.238	0.015	0.014	0.006	0.002	0.005	0.002	0.002	0.001
Fish and Aquatic Products		0.301	0.254	0.124	0.109	0.169	0.145	0.275	0.227
Freshwater fish	0.263	0.018	0.018	0.016	0.014	0.026	0.020	0.043	0.031
Marine fish	0.757	0.194	0.165	0.067	0.062	0.112	0.085	0.201	0.169
Fish and its products	0.251	0.048	0.032	0.020	0.015	0.009	0.011	0.016	0.016
Other aquatic animals and their products	0.187	0.042	0.038	0.021	0.017	0.022	0.030	0.014	0.010
Other proteinaceous products		0.192	0.177	0.096	0.092	0.043	0.049	0.040	0.037
Chicken eggs and its products	0.064	0.030	0.027	0.021	0.017	0.013	0.012	0.009	0.006
Duck eggs and its products	0.264	0.002	0.002	0.003	0.002	0.002	0.002	0.004	0.002
Dairy products	0.064	0.155	0.144	0.068	0.070	0.025	0.032	0.026	0.027
Soybean and its products (Tofu)	0.005	0.004	0.004	0.003	0.002	0.003	0.002	0.002	0.002
Vegetables		0.029	0.031	0.019	0.020	0.026	0.032	0.028	0.034
Dark green vegetables	0.022	0.023	0.024	0.016	0.017	0.022	0.027	0.022	0.029
Light color vegetables	0.004	0.003	0.003	0.002	0.002	0.002	0.002	0.003	0.002
Bamboo shoots	0.007	0.001	0.001	0.000	0.000	0.001	0.001	0.000	0.001
Melons	0.004	0.002	0.001	0.000	0.001	0.001	0.001	0.002	0.002
Beans	0.013	0.001	0.001	0.000	0.000	0.000	0.000	0.001	0.001
Mushrooms	0.010	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000
Fruits and their products		0.017	0.016	0.007	0.010	0.008	0.012	0.012	0.012
Fresh fruits	0.009	0.017	0.016	0.007	0.010	0.008	0.012	0.012	0.012
Total[b]		24.79	21.19	20.99	15.52	23.12	17.83	27.35	20.78
Total[c]		0.70	0.62	0.34	0.30	0.33	0.31	0.42	0.37
Total[d]		4.89	4.36	2.37	2.12	2.29	2.14	2.97	2.57
Total[e]		20.95	18.70	10.16	9.07	9.82	9.16	12.74	11.03

[a]pg WHO-TEQ/g fresh weight; [b]daily intake (pg WHO-TEQ/day); [c]total daily intake (pg WHO-TEQ/kg bw/day); [d]total weekly intake (pg WHO-TEQ/kg bw/week); [e]total monthly intake (pg WHO-TEQ/kg bw/month).

Table 3. Average daily intakes of PCDD/Fs/dl-PCBs TEQs as pg and (pg/kg bw)

Country, study period	Daily intakes, pg, (pg/kg bw)		Method[a]	Ref.
	PCDD/Fs+PCBs	only PCBs		
Finland, 1999	114 (1.50)	−	0	Kiviranta *et al*., 2004
Finland, 1999	116 (1.53)	−	LOQ	Kiviranta *et al*., 2004
Japan, 2000	161 (3.23)	−	0.5 × LOQ	Tsutsumi *et al*., 2001
Sweden, 1999	75 (1.05)	−	0.5 × LOQ	Lind *et al*., 2002
The Netherlands, 1999	91 (1.23)	−	0	Freijer *et al*., 2001
The United Kingdom, 2001	(0.9)	−	LOQ	FSA report 38/03
USA, 1995	146 (2.33)	−	0.5 × LOQ	Schecter *et al*., 2001
Taiwan, 2007	Male:96.6 (1.49) Female:74.1 (1.32)	−	LOQ	Hsu *et al*., 2007
Japan, 2000, 2001, 2002	139.28 (2.79), 111.99 (2.24), 133.99 (2.68)	−	0.5 × LOQ	Nakatani *et al*., 2011
Finland, 1999	−	53 (0.70)	0	Kiviranta *et al*., 2001
Sweden, 1999	−	63 (0.85)	LOQ	SCOOP, 2000
The Netherlands, 1991	−	81	LOQ	SCOOP, 2000
The United Kingdom, 1992	−	57 (0.81)	LOQ	SCOOP, 2000

[a]Method of denoting concentrations of unquantified congeners in intake calculations: 0 = lower bound, 0.5 × LOQ = medium bound, LOQ = upper bound.

to be all comparatively higher than that for people in Taiwan in this study[(14)]. For example, in the recent Finish survey 40 samples were analyzed[(15)]. In another German study, 3000 dioxin data from food samples were collected and analyzed through 5 years (1995-1999)[(22)]. Between 1995 and 1999, probably there were significant decreases in the concentrations of PCDD/F in food, which entails another distortion factor for the estimation of the dietary PCDD/PCDF intake. In addition, cereals and pulses were not included in that study. In USA, Schecter *et al*. (2001) analyzed 110 food samples divided into pooled lots by category. Only 12 separate analyses were conducted[(16)].

In all age groups, the daily intake of PCDD/Fs and dl-PCBs were within the TDI for dioxins established by the WHO (1-4 pg TEQ/kg/day), with a prevailing tendency towards the lower value of the range, 1 pg TEQ/kg/day. In addition, it is below the temporary weekly intake (t-TWI) of 7 pg WHO-$TEQ_{PCDD/Fs+dl\text{-}PCBs}$/kg bw recommended by the European Community SCF[(23)]; the monthly intake is also below the TDI of 14 pg TEQ/kg/week. Moreover, it is below the provisional tolerable monthly intake (PTMI) of 70 pg WHO-$TEQ_{PCDD/Fs+dl\text{-}PCBs}$/kg bw recommended by the Joint FAO/WHO Expert Committee on Food Additives[(24)].

III. *Contribution of PCDD/Fs and dl-PCBs to TEQ in Different Food Samples*

The greatest percentages of total daily dietary dioxin intake corresponded with fish and fishery products, especially in adults (51.6 [male] and 47.5% [female]) and the elderly (64.8 [male] and 61.8% [female]) (Figure 1). Moreover, the contribution of TEQ in this food is primarily from marine fish. The results suggested that changing fish dietary habits (that is, eating freshwater fish with lower TEQ levels) should reduce human exposure to PCDD/Fs and dl-PCBs. Other proteinaceous products made the second largest contributions, which account for 9.5-30.3% of the total intake. Even the PCDD/F concentrations of other proteinaceous products were lower than those of fish and shellfish. The estimated higher adolescents' consumption rates of other proteinaceous products (253.4 [male] and 185.2 [female] g/day) than fish and shellfish (42.4 [male] and 29.6 [female] g/day) resulted in a similar PCDD/F intake rate of other proteinaceous products (28.2 [male] and 30.3% [female]).

Moreover, the contribution of TEQ in proteinaceous products is primarily from dairy products. Children and adolescents eat many eggs and drink a lot of cow's milk instead of soybean milk in daily life. We previously[(25)] found that when elderly people ingested considerable amounts of soybean protein, such as tofu, they typically had low serum PCDD/F levels because of reduced body fat content and the induction of metabolic enzymes. Therefore, we hypothesize that soybean milk is a beneficial alternative drink for people with a higher dioxin burden. Fish and shellfish account for 64.8 and 61.8% of the daily intake of PCDD/Fs for elderly men and women, respectively, and poultry and livestock meat contributed only 9.0 and 7.4%. Furthermore, vegetables, often neglected as sources of PCDD/Fs, are estimated to contribute 4.1-10.4% to the dietary intake of PCDD/Fs, especially in adult and elderly women. Because of different dietary habits, the main food items contributing to the dietary intake of PCDD/Fs for adolescents, adults, and the elderly are clearly different. Male and female children had the highest PCDD/F intake in this study because of their lower body weights. It is important to remark the significant contribution to PCDD/PCDF intake of fish and shellfish, which has been

Journal of Food and Drug Analysis, Vol. 20, No. 4, 2012

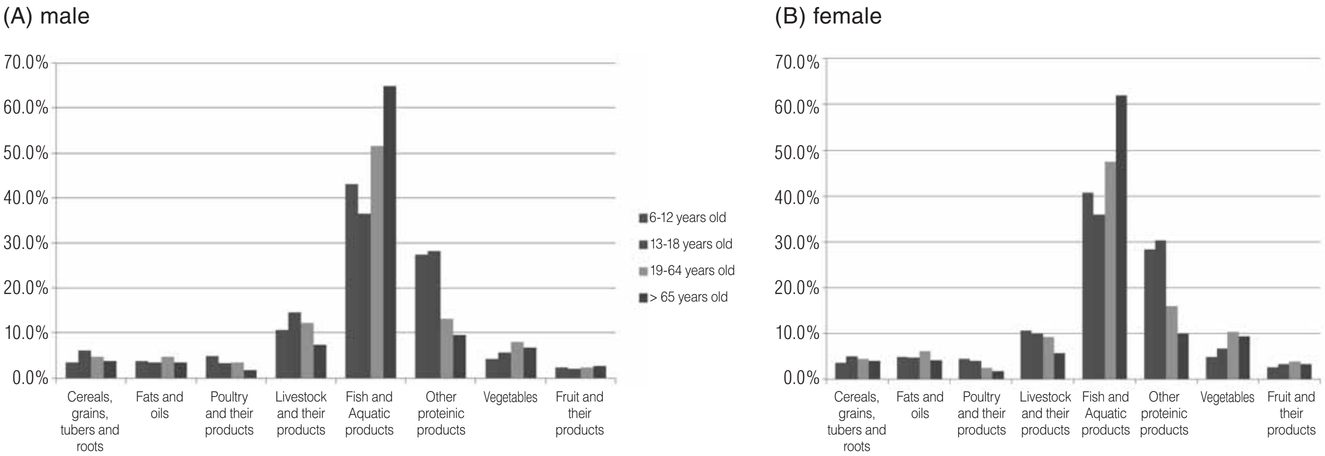

Figure 1. Percentage of contribution from each food group to the total daily intake of PCDD/Fs and dl-PCBs by Taiwanese in different age groups: (A) male; (B) female.

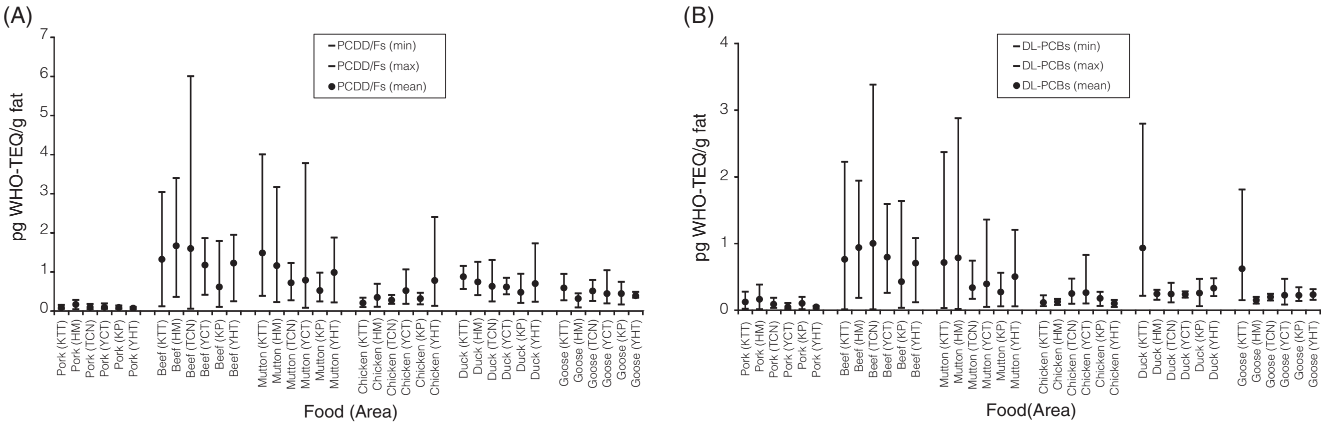

Figure 2. PCDD/F and dl-PCB levels in meat samples in 6 ambient air dispersion areas.

observed in most studies. However, the impact of other food groups, such as vegetables, fruits, and cereals, should not be ignored, especially in those countries or regions in which the consumption of these items is notable.

Llobet *et al*.[26] found that children could be exposed to the highest PCDD/F level per unit of body burden through their diet. Although the present analysis shows that children are exposed to more dietary dioxins on a body-weight basis than adolescents and adults, a greater risk to children cannot be presumed. Because TDIs are established based on chronic lifetime exposure, comparing childhood dioxin exposure to a TDI assumes that diet, and thus dioxin exposure, remains constant over a lifetime. That assumption overestimates potential risk, especially in view of decreasing environmental levels and body burdens.

IV. *Geographical Distribution of PCDD/F and dl-PCB Levels in Meat Samples*

In Taiwan during 2004-2008, the distribution of PCDD/Fs in meat samples in the 6 sampling areas were as follows: beef and mutton were the top 1 and 2 in all livestock samples in the HM area, and mutton, duck, and goose were higher in the KTT area than in all the other sampling areas (Figure 2A). The distribution of dl-PCBs showed a similar trend (Figure 2B). After the government forensic scientists identified the source of the contaminant: the mutton and beef were a local pollution episode caused primarily by the open burning of industrial and commercial wastes and by using the polluted animal feed.

V. *Time Trend of PCDD/F and dl-PCB Levels in Milk Samples*

Given that the primary mechanism for dioxins entering the food chain is through atmospheric deposition, cow's milk is considered a particularly suitable matrix for assessing their presence in the environment, because cows tend to graze over relatively large areas, and these compounds will, if present, concentrate in the fat content of milk. The mean value for the distribution of PCDD/Fs and dl-PCBs on milk fat in a

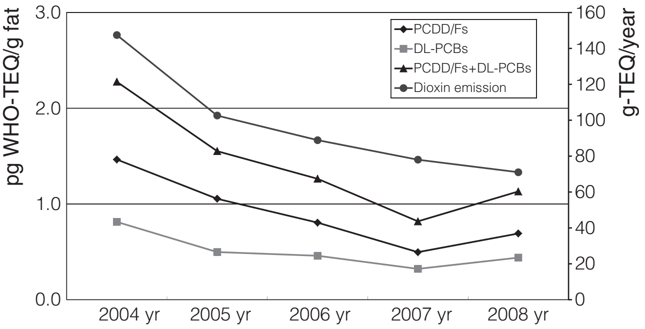

Figure 3. Annual distribution of PCDD/Fs and dl-PCBs in milk samples from 2004 to 2008.

2004- 2008 survey was 1.31 pg WHO-TEQ/g fat compared with corresponding mean values of 0.82 for 2007, 1.26 for 2006, 1.55 for 2005, and 2.28 pg WHO-TEQ/g fat for 2004 (Figure 3). The downward trend in milk samples mirrored the concomitant downward trend in total dioxin emissions in Taiwan[27].

VI. *Suggestions for Reducing the Intake of Dietary PCDD/Fs and dl-PCBs*

One food safety tip is that consumers adopt risk management by obtaining food from different sources. It is better not to buy food products of the same category, from the same area, or of the same brand. And food had better be changed as often as possible. Animal meat products like pork and fish, parts like skin, fat, and viscera, where dioxins easily accumulate, should be avoided. Of course, autonomous management and inspections by food makers themselves over their raw materials and products is also crucial to reducing dioxin contamination in food.

CONCLUSIONS

Data from this study suggested that, generally, there is no health risk from PCDD/Fs and dl-PCBs in food in Taiwan, except for occasionally high PCDD/F levels in polluted beef and mutton. These data suggest that the environment near where the livestock was farmed should be examined. The highest levels of PCDD/Fs and dl-PCBs were found in fish; however, freshwater fish had lower levels than did marine fish. A number of studies have shown that, since the 1980s, efforts to control emissions and reduce human exposure have been successful in some industrialized countries[28,29]. In these countries, emission, food levels, and body burden of dioxins have been reduced several times, whereas, in Taiwan, measures for controlling contamination have only recently been implemented. Although estimated daily intake for our study population is low, there is potential for higher levels of contamination in Taiwan in the future. Continual monitoring of PCDD/F and dl-PCB contamination, especially in food, is absolutely essential.

REFERENCES

1. Liem, A. K. D, Fürst, P. and Rappe, C. 2000. Exposure of populations to dioxins and related compounds. Food Addit. Contam. 17: 241-259.
2. Focant, J. F., Eppe, G. and Pirard, C. *et al.* 2002. Levels and congener distributions of PCDDs, PCDFs and non-ortho PCBs in Belgian foodstuffs. Assessment of dietary intake. Chemosphere 48: 167-179.
3. Van den Berg, M., Birnbaum, L. S. and Denison, M. *et al.* 2006. The 2005 World Health Organization reevaluation of human and mammalian toxic equivalency factors for dioxins and dioxin-like compounds. Toxicol. Sci. 93: 223-241.
4. van Leeuwen, F. X., Feeley, M. and Schrenk, D. *et al.* 2000. Dioxins: WHO's tolerable daily intake (TDI) revisited. Chemosphere 40: 1095-1101.
5. European Commission. 2001a. Communication from the Commission to the Council, the European Parliament and the Economic and Social Committee of 24 October 2001. Community Strategy for Dioxins, Furans and Polychlorinated Biphenyls. Off. J. Eur. Comm. C322.
6. European Commission. 2006a. Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. Off J. Eur. Union L364, 5-24.
7. European Commission. 2006b. Commission Recommendation of 6 February 2006 on the reduction of the presence of dioxins, furans and PCBs in feeding stuffs and foodstuffs (notified under document number C(2006) 235). Off J. Eur. Union L42, 26-28.
8. Wu, S. J., Chang, Y. H., Fang, C. W. and Pan, W. H. 1999. Food sources of weight, calories and three macronutrients-NAHSIT 1993-1996. Nutr. Sci. J. 24: 41-58.
9. Chen, H. L., Su, H. J., Hsu, J. F., Liao, P. C. and Lee, C. C. 2008. High variation of PCDDs, PCDFs, and dioxin-like PCBs ratio in cooked food from the first total diet survey in Taiwan. Chemosphere 70: 673-681.
10. USEPA. 1994. Method 1613B: Tetra-through Octa-chlorinated Dioxin and Furans by Isotope Dilution HRGC/HRMS (Revision B). USEPA Office of Water.
11. USEPA. 1999. Method 1668, Revision A: Chlorinated Biphenyl Congeners in Water, Soil, Sediment, and Tissue by HRGC/HRMS. USEPA Office of Water.
12. Lind, Y., Darnerud, P. O., Aune, M. and Becker, W. Exponering för organiska miljökontaminanter via livsmedel. Report from the Swedish NFA (in Swedish), report 26-2002.
13. Tsutsumi, T., Yanagi, T. and Nakamura, M. *et al.* 2001. Update of daily intake of PCDDs, PCDFs, and dioxin-like PCBs from food in Japan. Chemosphere 45:1129-1137.
14. Hsu, M. S., Hsu, K.Y. and Wang, S. M. *et al.* 2007. A total diet study to estimate PCDD/Fs and dioxin-like PCBs intake from food in Taiwan. Chemosphere 67: S65-S70.
15. Kiviranta, H., Hallikainen, A., Ovaskaiens, M. L., Kumpulainen, J. and Vartiainen, T. 2001. Dietary intakes of polychlorinated dibenzo-p-dioxins, dibenzofurans

and polychlorinated biphenyls in Finland. Food Add. Contam. 18: 945-953.
16. Schecter, A., Cramer, P., Boggess, K., Stanley, J., Päpke, O., Olson, J., Silver, A. and Schmitz, M. 2001. Intake of dioxins and related compounds from food in the U.S. population. J. Toxicol. Environ. Health A 63: 1-18.
17. Kiviranta, H., Ovaskainen, M. L. and Vartiainen, T. 2004. Market basket study on dietary intake of PCDD/Fs, PCBs, and PBDEs in Finland. Environ. Int. 30: 923-932.
18. Freijer, J. I., Hoogerbrugge, R. and van Klaveren, J. D. *et al.* 2001. Dioxins and dioxin-like PCBs in foodstuffs: Occurrence and dietary intake in The Netherlands at the end of the 20th century. RIVM report 639102022, Bilthoven: The Netherlands.
19. Food Standards Agency (FSA). Dioxins and dioxin-like PCBs in the UK diet: 2001 total diet study samples. Food Survey Information Sheets on the WWW: http://www.food.gov.uk/science/surveillance/. Report 38/03; 2003.
20. Nakatani, T., Yamamoto, A. and Ogaki, S. 2011. A survey of dietary intake of polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans, and dioxin-like coplanar polychlorinated biphenyls from food during 2000-2002 in Osaka City, Japan. Arch. Environ. Contam. Toxicol. 60: 543-555.
21. SCOOP: Scientific co-operation on questions relating to food ''assessment of dietary intake of dioxins and related PCBs by the population of EU member states'' Task 3.2.5—Final report—7 June, 2000.
22. Vieth, B., Heinrich-Hirsch, B. and Mathar, W. 2000. Trends in dioxin intake and human milk levels in Germany. Organohalogen Compd. 47: 300-303.
23. European Commission. 2001b. Opinion of the Scientific Committee on Food on the risk assessment of dioxins and dioxin-like PCBs in food. Update based on new scientific information available since the adoption of the SCF opinion of 22nd November 2000. CS/CNTM/DIOXIN/20 final. Adopted on 30 May 2001.
24. Joint FAO/WHO Expert Committee on Food Additives (JECFA). 2001. Summary of the Fifty-seventh Meeting of the Joint FAO/WHO Expert Committee on Food Additives.
25. Chen, H. L., Su, H. J. and Lee, C. C. 2007. Association between tofu intake and serum polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) in the elderly Taiwanese. Environ. Int. 33: 265-271.
26. Llobet, J. M., Martí-Cid, R., Castell, V. and Domingo, J. L. 2008. Significant decreasing trend in human dietary exposure to PCDD/PCDFs and PCBs in Catalonia, Spain. Toxicol. Lett. 178: 117-126.
27. Taiwan EPA. 2008. The National Implementation Plan of Republic of China (R.O.C., Taiwan) under the Stockholm Convention on Persistent Organic Pollutants. http://ivy1.epa.gov.tw/Dioxin_Toxic/Index/InstructionFrameset.aspx?type=Mg==&main=YXAyL25pcC5hc3A=
28. Harrison, N., Wearne, S. and De Gem, M. G. *et al.* 1998. Time trends in human dietary exposure to PCDDs, PCDFs and PCBs in the UK. Chemosphere 37: 1657-1670.
29. Charnley, G. and Doull, J. 2005. Human exposure to dioxins from food 1999-2002. Food Chem. Toxicol. 43: 671-679.