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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,

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

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.

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

Table 2
PCDD/F concentrations (mean, R.S.D.[a] or R.P.D.[b]) foods (pg/g lipid)

Congeners	2,3,7,8-TeCDD	1,2,3,7,8-PeCDD	1,2,3,4,7,8-HxCDD	1,2,3,6,7,8-HxCDD	1,2,3,7,8,9-HxCDD	1,2,3,4,6,7,8-HpCDD
Pork (*n* = 2)	0.037 (2.5)	0.052 (103)	0.093 (60)	0.113 (136)	0.143 (19)	0.928 (29)
Beefs (*n* = 2)	0.113 (48)	0.187 (136)	0.259 (142)	0.361 (98)	0.115 (50)	1.53 (111)
Chickens (*n* = 2)	0.071 (3.8)	0.072 (108)	0.051 (13)	0.151 (137)	0.092 (74)	0.626 (5.1)
Ducks (*n* = 2)	0.178 (33)	0.325 (11)	0.145 (82)	0.733 (32)	0.119 (117)	1.98 (4.0)
Eggs (*n* = 2)	0.030 (70)	0.054 (73)	0.074 (90)	0.158 (51)	0.039 (13)	1.41 (86)
Duck eggs (*n* = 2)	0.051 (91)	0.106 (134)	0.065 (165)	0.122 (182)	0.059 (171)	0.593 (184)
Milk (*n* = 2)	0.087 (20)	0.323 (5.0)	0.128 (21)	0.290 (7.3)	0.134 (34)	0.499 (5.9)
Cheeses (*n* = 2)	0.049 (7.9)	0.092 (20)	0.051 (0.9)	0.088 (31)	0.088 (0.9)	0.297 (42)
Linseed oils (*n* = 2)	0.013 (0)	0.039 (0)	0.034 (0)	0.081 (110)	0.071 (35)	1.36 (142)
Soybean oils (*n* = 2)	0.006 (1.1)	0.019 (1.1)	0.017 (1.1)	0.018 (1.1)	0.040 (40)	0.278 (76)
Mouthbreeder (*n* = 3)	0.515 (38)	0.606 (28)	0.141 (33)	0.174 (35)	0.150 (65)	0.960 (66)
Milk fish (*n* = 2)	0.175 (46)	0.328 (32)	0.114 (16)	0.265 (17)	0.196 (23)	0.578 (92)
Grouper (*n* = 6)	0.617 (161)	0.975 (106)	0.330 (70)	2.17 (146)	0.783 (166)	2.83 (121)
Shrimp (*n* = 4)	0.894 (67)	1.43 (90)	1.57 (145)	4.68 (185)	2.19 (123)	2.041 (191)
Oyster (*n* = 3)	1.32 (31)	2.12 (44)	2.66 (20)	7.05 (7.6)	4.32 (9.7)	93.2 (11)
Clam (*n* = 3)	1.04 (37)	3.03 (37)	2.62 (37)	2.77 (37)	4.48 (37)	14.3 (42)
Rice (*n* = 2)[c]	0.001 (1.9)	0.003 (8.9)	0.005 (100)	0.007 (107)	0.024 (149)	0.039 (113)
Cabbage (*n* = 2)[c]	ND (NA)	ND (NA)	ND (NA)	0.0005 (33)	0.0002 (200)	0.002 (59)
Water celery (*n* = 2)[c]	0.0006 (200)	0.001 (200)	0.001 (200)	0.003 (15)	0.002 (22)	0.029 (22)
Banana (*n* = 2)[c]	0.0004 (200)	ND (NA)	ND (NA)	ND (NA)	ND (NA)	0.002 (200)
Apple (*n* = 2)[c]	ND (NA)	0.0004 (200)	0.0007 (200)	0.001 (200)	0.0009 (200)	0.020 (150)
Pineapple (*n* = 2)[c]	ND (NA)	ND (NA)	ND (NA)	ND (NA)	ND (NA)	0.001 (42)
Bread (*n* = 2)[c]	0.001 (11)	0.002 (11)	0.002 (11)	0.002 (11)	0.008 (91)	0.050 (32)
Sauce (*n* = 2)[c]	0.0005 (73)	0.001 (78)	0.001 (114)	0.001 (110)	0.003 (121)	0.006 (89)
Instant noodles (*n* = 2)[c]	0.003 (11)	0.010 (11)	0.009 (11)	0.009 (11)	0.019 (35)	0.215 (34)

Congeners	OCDD	2,3,7,8-TeCDF	1,2,3,7,8-PeCDF	2,3,4,7,8-PeCDF	1,2,3,4,7,8-HxCDF	1,2,3,6,7,8-HxCDF	1,2,3,7,8,9-HxCDF	2,3,4,6,7,8-HxCDF	1,2,3,4,6,7,8-HpCDF	1,2,3,4,7,8,9-HpCDF	OCDF	Total	TEQ
Pork (*n* = 2)	7.53 (46)	0.052 (93)	0.045 (37)	0.143 (38)	0.191 (87)	0.111 (18)	0.116 (80)	0.165 (47)	0.761 (36)	0.157 (81)	1.15 (7.9)	11.8	0.253
Beefs (*n* = 2)	7.70 (100)	0.093 (51)	0.055 (27)	0.180 (142)	0.224 (76)	0.213 (142)	0.136 (78)	0.288 (99)	0.692 (30)	0.089 (40)	1.86 (126)	14.1	0.551
Chickens (*n* = 2)	3.72 (18)	0.240 (54)	0.194 (55)	0.227 (51)	0.341 (116)	0.273 (76)	0.104 (109)	0.327 (93)	0.569 (16)	0.092 (50)	1.20 (54.8)	8.36	0.39
Ducks (*n* = 2)	5.27 (6.2)	0.479 (20)	0.536 (31)	0.688 (20)	0.511 (32)	0.473 (44)	0.075 (51)	0.394 (16)	0.870 (39)	0.137 (20)	1.11 (12.1)	14	1.05
Eggs (*n* = 2)	38.2 (165)	0.156 (21)	0.151 (11)	0.165 (18)	0.092 (36)	0.071 (14)	0.047 (9.2)	0.151 (12)	0.365 (11)	0.055 (7.2)	0.629 (0.6)	41.8	0.248
Duck eggs (*n* = 2)	6.27 (194)	0.323 (137)	0.232 (151)	0.385 (160)	0.371 (183)	0.201 (168)	0.066 (173)	0.181 (185)	0.600 (190)	0.048 (134)	2.21 (195)	11.9	0.434
Milk (*n* = 2)	2.65 (121)	0.307 (19)	0.179 (23)	0.835 (5.4)	0.423 (12)	0.382 (6.7)	0.081 (20)	0.340 (29)	0.281 (29)	0.126 (209)	0.228 (0.2)	7.29	0.885
Cheeses (*n* = 2)	1.33 (56)	0.245 (3.0)	0.131 (32)	0.183 (25)	0.099 (18)	0.122 (0.9)	0.081 (0.9)	0.179 (0.9)	0.208 (53)	0.126 (0.9)	0.210 (11)	3.58	0.302
Linseed oils (*n* = 2)	7.71 (48)	0.040 (0)	0.038 (0)	0.048 (0)	0.050 (0)	0.081 (0)	0.054 (0)	0.120 (0)	0.081 (0)	0.08440 (0)	0.146 (17)	10.1	0.14
Soybean oils (*n* = 2)	10.3 (147)	0.020 (1.1)	0.019 (1.1)	0.024 (1.1)	0.029 (31)	0.040 (1.1)	0.033 (40)	0.059 (1.1)	0.087 (35)	0.041 (1.1)	0.139 (108)	11.2	0.068
Mouthbreeder (*n* = 3)	7.49 (109)	3.68 (73)	0.706 (51)	0.973 (19)	0.227 (4.1)	0.198 (56)	0.080 (26)	0.231 (59)	1.04 (73)	0.223 (49)	1.37 (81)	18.8	1.95
Milk fish (*n* = 2)	2.82 (98)	1.01 (2.6)	0.588 (20)	1.01 (15)	0.283 (7.4)	0.250 (7.5)	0.142 (15)	0.283 (13)	0.362 (47)	0.135 (80)	0.574 (117)	9.12	1.09
Grouper (*n* = 6)	18.3 (158)	5.37 (153)	2.47 (127)	2.35 (78)	1.04 (158)	1.02 (93)	0.147 (96)	0.888 (98)	1.91 (138)	0.3166 (136)	6.622 (153)	48.2	3.6
Shrimp (*n* = 4)	5.390 194)	3.31 (79)	2.09 (91)	3.47 (101)	2.45 (99)	2.5 (105)	0.673 (71)	2.28 (78)	12.3 (65)	1.02 (64)	24.2 (84)	5,660	9.19
Oyster (*n* = 3)	1.080 (14)	10.3 (17)	2.57 (57)	4.3 (64)	1.13 (10)	3.85 (19)	9.12 (7.3)	1.15 (21)	55.9 (17)	3.23 (17)	3151 (18)	1,600	10.7
Clam (*n* = 3)	1.112 (28)	5.58 (24)	3.12 (37)	4.6 (35)	3.82 (37)	6.2 (37)	9.12 (37)	4.15 (37)	11.1 (45)	6.4 (37)	20.5 (46)	2143	9.77
Rice (*n* = 2)[c]	0.107 (37)	0.008 (1.3)	0.005 (40)	0.003 (8.6)	0.008 (89)	0.009 (103)	0.019 (139)	0.015 (110)	0.037 (113)	0.020 (128)	0.095 (113)	0.412	0.0165
Cabbage (*n* = 2)[c]	0.013 (0.3)	0.001 (61)	0.0009 (32)	0.001 (52)	0.0009 (45)	0.0008 (41)	0.0008 (53)	ND (NA)	0.002 (50)	0.0002 (200)	0.003 (31)	0.029	0.025
Water celery (*n* = 2)[c]	0.119 (23)	0.005 (15)	0.006 (17)	0.006 (12)	0.007 (12)	0.007 (11)	0.008 (14)	ND (NA)	0.029 (29)	0.005 (60)	0.037 (6.7)	0.001	0.009
Banana (*n* = 2)[c]	0.019 (33)	0.0004 (200)	0.0005 (200)	0.0007 (200)	0.001 (57)	0.001 (74)	ND (NA)	0.0006 (200)	0.006 (41)	NDD (NA)	0.012 (35)	0.046	0.001
Apple (*n* = 2)[c]	0.059 (139)	0.001 (4.9)	0.001 (36)	0.001 (53)	0.001 (32)	0.001 (33)	0.001 (77)	ND (NA)	0.003 (39)	0.0004 (200)	0.004 (114)	0.101	0.002
Pineapple (*n* = 2)[c]	0.007 (39)	0.0007 (58)	ND (NA)	0.0003 (200)	ND (NA)	ND (NA)	ND (NA)	ND (NA)	0.0003 (200)	NDD (NA)	0.001 (200)	0.011	0.0002
Bread (*n* = 2)[c]	1.69 (17)	0.007 (117)	0.004 (82)	0.008 (111)	0.007 (98)	0.008 (55)	0.010 (27)	0.003 (11)	0.037 (22)	0.005 (11)	0.062 (82)	1.91	0.013
Sauce (*n* = 2)[c]	0.022 (133)	0.001 (73)	0.002 (125)	0.002 (101)	0.003 (129)	0.003 (96)	0.004 (73)	0.002 (97)	0.006 (122)	0.004 (107)	0.011 (122)	0.08	0.005
Instant noodles (*n* = 2)[c]	15.72 (18)	0.010 (11)	0.011 (19)	0.014 (14)	0.033 (63)	0.023 (3)	0.033 (0.2)	0.014 (11)	0.123 (1.5)	0.022 (11)	0.141 (109)	16.42	0.043

ND: not detected; NA: not available.
[a] Relative standard deviation.
[b] Relative percentage difference.
[c] Results are given in pg/g fresh weight.

Table 3
Total PCDD/F levels, the consumption rates, and the daily intakes of PCDD/Fs of each food group by adults in Taiwan

Food groups	Food concentration		Consumption rate of adults (g)		Daily intake (pg WHO-TEQ/day)	
	pg WHO-TEQ/g fat	pg WHO-TEQ/g fresh weight	Male	Female	Male	Female
Cereals	–	0.0166	210	129	2.24	1.39
Dark green vegetables	–	0.0086	84.4	79.0	1.30	1.31
Light green Vegetables	–	0.0256	153	154	0.084	0.0843
Vegetable oils	0.104	–	11.1	12.5	0.750	0.830
Chickens	0.390	0.056	52.2	31.1	2.93	1.74
Ducks	1.05	0.220	9.21	3.46	2.02	0.76
Pork	0.253	0.0339	126	88.2	4.28	2.99
Beef	0.074	0.0744	0.622	3.12	0.585	0.232
Freshwater fish	1.52	0.123	29.0	19.3	4.48	2.98
Saltwater fish	3.60	0.143	34.4	21.1	4.92	3.03
Shellfish	9.82	0.116	28.1	28.5	3.39	3.44
Eggs	0.248	0.0692	34.3	26.7	2.56	1.99
Milk and dairy products	0.594	0.0475	55.9	58.5	2.66	2.78
Soybean and soybean food	–	0.0156	78.9	46.5	1.23	0.72
Fresh fruits	–	0.0011	172	208	0.193	0.234
Breads	0.303	0.0127	10.6	10.1	0.135	0.129
Sauces	–	0.00494	51.4	29.6	0.254	0.146
Instant noodles	0.177	0.0425	8.50	6.12	0.361	0.260
Total	–	–	1124	955	34.4	25.1

PCDD/Fs were estimated to be 34.4 and 25.1 pg WHO-TEQ for male and female, respectively. The daily, weekly, and monthly intakes (pg WHO-TEQ/day/b.w., pg WHO-TEQ/week/b.w., and pg WHO-TEQ/month/b.w.) and the fractions (%) contributed by 10 major food groups to the daily intakes of PCDD/Fs by teenagers, adults and seniors in Taiwan were shown in Table 4. The estimations of PCDD/F intake of the teenagers and the senior persons were according to the intake amount ratio of the teenagers to the adults and the senior persons to the adults on each group of food investigated by Department of Health, Taiwan (1999–2000) [21]. The results range from 21.8 pg WHO-TEQ/day for female teenagers to 38.6 pg WHO-TEQ/day for male seniors. The daily, weekly and monthly intakes for all groups of population were lower than the corresponding tolerable limits. Rice, the staple of Taiwanese (0.0166 pg WHO-TEQ/g), contributed 2.24 and 1.39 pg WHO-TEQ for male and female, respectively. Fish and shellfish are the main contributor to dietary intakes of adults (37.2 and 37.7% for male and for female, respectively) and seniors (42.6 and 39.8% for male and for female, respectively).

Even the PCDD/F concentrations of meats were lower than those of fish and shellfish, the teenagers' estimated great higher consumption rates of meat (204 and 138 g/day for male and female) than fish and shellfish (62.9 and 45.9 g/day for male and female) caused the higher PCDD/F intake rate of meats (31.9 and 28.8% for male and female) than fish and shellfish (26.2 and 28.8% for male and female). As for the intake trend of seniors, fish and shellfish account for 42.6 and 39.8% daily intake of PCDD/Fs for male and female seniors, respectively, and meats contributed only 20.0 and 16.7%. Besides fish, shellfish, and meat, eggs and dairy products are other important sources for intake of PCDD/Fs, especially for teenagers who intakes more than 10% of PCDD/Fs from eggs. Furthermore, rice and vegetables, often neglected as sources for intake of PCDD/Fs, are estimated to contribute 5.40–7.21%, respectively, to the dietary intake of PCDD/Fs. Because of different dietary habits, the main food items contributing to the dietary intake of PCDD/Fs for teenagers, adults and seniors are clearly different.

Male teenagers and seniors are the highest PCDD/F intake population in this study because of their lower body weights. Llobet et al. [22] have found that children could be exposed to the highest PCDD/F level per unit body burden through diet. The same situation might hold for Taiwanese children though the estimation of daily intake for children is not included because of the

Table 4
Daily, weekly, and monthly intakes (pg WHO-TEQ/day/b.w., pg WHO-TEQ/week/b.w., and pg WHO-TEQ/month/b.w.) and the fractions (%) contributed by 10 major food groups to the daily intakes of PCDD/Fs by teenagers, adults and seniors in Taiwan

	Teenagers (12–18 years old)		Adults (19–64 years old)		Intake fractions of seniors (more then 65 years old)	
	Male	Female	Male	Female	Male	Female
Intake fractions (%)						
Cereals	7.21	6.29	6.53	5.54	5.30	5.40
Fats and oils	0.94	2.50	2.18	3.31	1.20	2.34
Soybeans and soybean products	4.39	2.49	3.57	2.89	1.63	1.96
Meats	31.9	28.8	28.6	22.9	20.0	16.7
Eggs	10.7	11.4	7.45	7.96	4.09	3.60
Milk and dairy products	13.2	12.8	7.7	11.1	18.9	22.6
Seafood	26.2	28.9	37.2	37.7	42.6	39.8
Vegetables	2.62	3.62	4.03	5.56	3.56	4.87
Fruits	0.58	0.80	0.56	0.93	0.80	0.77
Others	2.24	2.45	2.18	2.14	2.00	1.93
Dietary intake of PCDD/Fs						
Daily intake (pg WHO-TEQ/day)	33.5	21.8	34.4	25.1	37.6	27.7
Total daily intake (pg WHO-TEQ/day/b.w.)	0.609	0.428	0.531	0.445	0.614	0.492
Total monthly intake (pg WHO-TEQ/week/b.w.)	4.27	3.00	3.71	3.11	4.30	3.44
Total monthly intake (pg WHO-TEQ/month/b.w.)	18.3	12.9	15.9	13.3	18.4	14.8

lack of detailed food intake data, another subject requiring further investigations.

In a study on dietary intake with CALUX bioassay in Belgium, dietary intake of PCDD/Fs (including six kinds of meat and meat products, nine kinds of fishes and shellfishes, three dairy products, cereals, eggs, and oil) were estimated to be 2.24, 2.09, and 1.74 pg CALUX TEQ/kg b.w./day for adolescents, mothers, and adults, respectively [19]. A previous study in the same country (including meat, poultry, eggs, milk and dairy products, fish and seafood) reported the intake level of 65.29 pg/WHO-TEQ/day and 1.00 pg/WHO-TEQ/b.w./day. Fish and seafood, and milk and dairy products contributed about 40 and 30% to the total intake value. [23].

In Spain, Bocio and Domingo [6] examined the intake of PCDD/Fs from vegetables, pulses, cereals, fruits, seafoods, meats, eggs, dietary products, and oil in 1998 and 2002, and a significant decrease were observed from 210 to 59.6 pg WHO-TEQ/day due to the use of new technologies in the flue gas cleaning systems of incinerators and vehicles. In a more recent study [24], the intake in Tarragona Spain further decreased from 63.8 pg WHO-TEQ/day in 2002 to 27.81 pg WHO-TEQ/day in 2006.

In Swedish, the estimated PCDD/F dietary intake based on a market basket study including fish, meat, dairy product, egg, fat, pastry in 1999 was reported to be 54.4 pg TEQ/day [25]. In France, the intake of meat products, fish and fish products, fats, dairy products, eggs, cereals, fruits and vegetables in 2005 were estimated to be 0.53 pg WHO-TEQ/kg b.w./day. Besides seafoods, dairy products were another important source of PCDD/Fs [18]. The PCDD/F dietary intake from vegetables, fruits, meats, fish and shellfish was estimated to be 44.7 pg WHO-TEQ/day in Japan [26].

These PCDD/F dietary intake values were observed to be all comparatively higher than that for people in Taiwan in this study, except for that in France [18] and Spain [24] which was very close to the value in this study. However, similar to this study, seafood all contributed the most to the dietary intake for most population despite that seafood was a more important source of the PCDD/F intake in Japan (67%) [26] and France (45.2%) [18] than that in Taiwan (37.2 and 37.7% and for male and female adults). Since fish and seafood were main contributors to the dietary intakes, Domingo and Bocio [27] reported detail PCDD/F concentration and the intake from these species and indicated that some population who frequently consuming fish and seafood could be significantly increasing health risk due to PCDD/F exposure.

Although the PCDD/F daily intake for most populations were below 70 WHO-TEQ/month/kg b.w., some specific population may expose to higher risk of PCDD/F intake. To understand the PCDD/F intake of the duck-farmer population who usually consumes a large amount of duck eggs and duck meat from their own farms, this study examines several scenarios on the contaminated duck-eggs issue. The PCDD/F level of 0.434 pg WHO-TEQ/g lipid was taken as normal level in duck eggs and the chicken consumption were replaced with duck meat. It is assumed that the farmers consume from 34.3 and 26.7 g of duck eggs for male and female (the average consumption) to 165 g (the upper limit) per day. Further, the assumed PCDD/F levels of 1, 3, 10, 20, and 30 pg WHO-TEQ/g lipid via duck eggs were also used in estimation (Fig. 1). The estimated intake of PCDD/Fs was still less than 70 pg WHO-TEQ/kg b.w./month even when three duck eggs with 1 pg WHO-TEQ/g lipid were consumed each day. The intake level became higher than 70 pg WHO-TEQ/kg b.w./month when one to two duck eggs with 3 pg WHO-TEQ/g lipid weight were consumed. While the PCDD/F level in duck eggs were as high as 10 pg WHO-TEQ/g lipid, even ingesting less than one egg everyday would cause intolerable intake of PCDD/Fs both for adults and teenagers. In addition, teenagers in farmer's family could expose slightly higher risk than adults due to the low body weight,

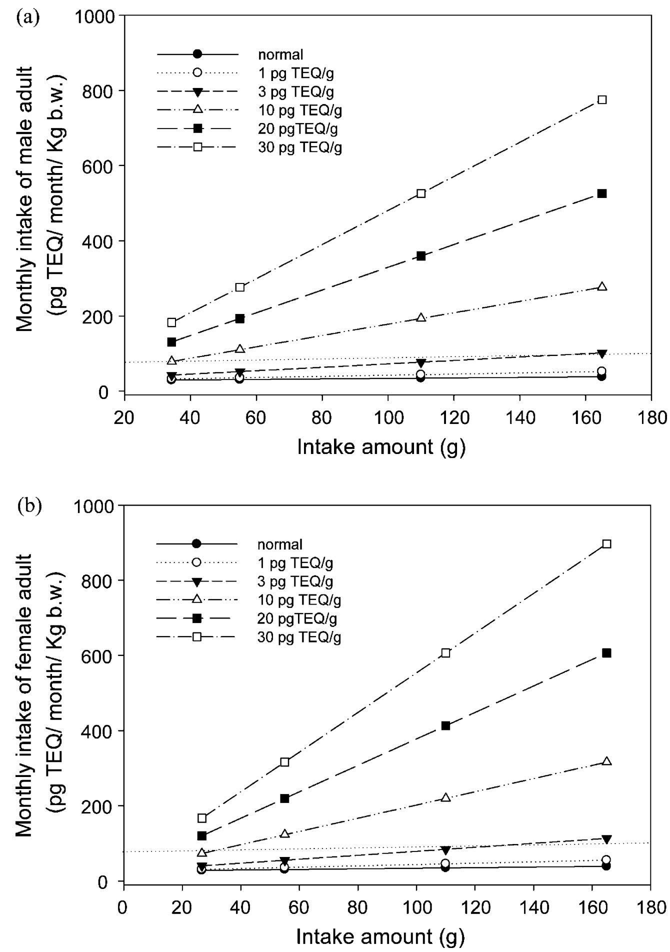

Fig. 1. Daily intake of male (a) and female (b) duck-farmers consuming different PCDD/F levels of duck eggs.

while the seniors expose the last due to low consuming amount of eggs.

As for normal population who only consume duck eggs amount of 1/14 eggs, the intake of PCDD/Fs could be raised to more than three times (from 16.3 to 49.9 pg WHO-TEQ/month/b.w.) because of contaminated duck eggs (30 pg WHO-TEQ/g lipid) though its still below the tolerable level (70 pg WHO-TEQ/month/kg b.w.). For teenagers, the ingestion of contaminated duck eggs of the same level PCDD/Fs would cause 74.2 pg WHO-TEQ/month/kg b.w. (male) and 54.8 pg WHO-TEQ/month/kg b.w. (female) of PCDD/Fs exposure, while the temporary daily exposure was lower than 4 pg WHO-TEQ/kg b.w./day. From the above simulation results, there is a little risk to cause intolerable intake except for the continually consuming contaminated eggs with extreme PCDD/Fs levels for normal population.

Although most important food has been included in this study, the PCDD/F intake by Taiwanese from various fish and shellfish such as crabs, yellow croaker, the other foods such as animal fat, squash, nuts and nut products still need further investigation.

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