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Cover Page Footnote

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Spatiotemporal patterns of polychlorinated dibenzo-*p*-dioxins and dibenzofurans and dioxin-like polychlorinated biphenyls in foodstuffs in air quality regions in Taiwan

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Abstract

High-fat food intake is the main source of dioxin-like compounds for humans, such as consumption of meat, dairy and eggs, and seafood products. Fruits, vegetables, and cereals have relatively low levels of dioxin-like compounds, but because of high consumption they also contribute to the food-borne intake. It is necessary to clarify dietary dioxin exposure affected by different food contamination levels and dietary habits among different geographic areas. We aimed to evaluate chronic dietary PCDD/Fs and DL-PCBs exposure in 725 individual foods in 14 categories in 6 Taiwan air quality regions (AQRs) and a total of 2441 foods from 2004 to 2018. We estimated daily PCDD/Fs + DL-PCBs intake on the basis of sex- and age-specific foodstuff ingestion rate and PCDD/Fs + DL-PCBs concentrations using a probabilistic approach. PCDD/F + DL-PCB levels among the different sampling periods exhibited a decreasing trend in fish and aquatic products (from 0.384 ± 0.764 to 0.206 ± 0.223 pg WHO_{05}-TEQ g^{-1} w.w.) (p for trend = 0.043), livestock products (from 0.133 ± 0.298 to 0.035 ± 0.043 pg WHO_{05}-TEQ g^{-1} w.w.), eggs (from 0.221 ± 0.373 to 0.056 ± 0.048 pg WHO_{05}-TEQ g^{-1} w.w.) (p for trend = 0.002), and dairy samples (from 0.066 ± 0.075 to 0.024 ± 0.026 pg WHO_{05}-TEQ g^{-1} w.w.) (p for trend = 0.001). All lifetime average daily doses (LADD) were below provisional tolerable monthly intake (PTMI) but higher than the TWI for PCDD/Fs and DL-PCBs in food. The percentages of the contribution of each food group to the total dietary intake of $TEQ_{PCDD/F+PCB}$ in different ambient air dispersion areas and age groups. The total daily intake of PCDD/Fs and DL-PCBs by Taiwanese differed between AQRs (0.188−0.397 pg WHO_{05}-TEQ kg^{-1} b.w. day^{-1}). The observed geographical variations were likely due to differences in food habits, cuisines, culture and levels of environmental contamination among various regions in Taiwan. By sensitivity analysis, we have identified the major contribution to LADD, which was the dioxin levels in marine fish, freshwater fish and fish related products, and followed by dioxin levels in duck eggs. In addition, marine and freshwater fish consumption rate accounts more than 10.2%. These major exposure variables was also consistent with the findings of total daily intake in different AQRs.

Keywords: Daily intake, Geographical variations, Taiwan air quality regions, Probabilistic approach

1. Introduction

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), and polychlorinated biphenyls (PCBs) are of global concern because of their persistence, bioaccumulation, and toxicity. More than 90% of human exposure to dioxins and dioxin-like PCBs (DL-PCBs) is estimated to occur through the diet, mainly from meat and dairy products, fish, and shellfish. Therefore, many national authorities have regular food monitoring programs. Chronic exposure to PCDD/Fs and DL-PCBs are of notable concern given their considerable toxic potential, which could have

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reproductive and developmental effects, neurological and behavioral effects, dermal toxicity, and immunomodulatory and carcinogenic effects in humans [1−6]. Exposure to dioxin is also associated with an increased risk of diabetes [7].

Food represents the primary source of nonoccupational exposure to PCDD/Fs and PCBs (more than 90% of total exposure) [8,9]. Numerous studies have confirmed that the main foods currently contributing to PCDD/Fs and PCBs exposure are fatty fish, meat, and meat products, as well as milk and dairy products [10, 11,12]. Therefore, dietary estimations are appropriate tools to estimate the exposure to such compounds and evaluate the potential risk in a population. According to geographical characteristics and air quality conditions, Taiwan Environmental Protection Agency (EPA) has divided the island into seven air quality regions (AQRs), namely North, Chu-Miao, Central, Yun-Chia-Nan, Kao-Ping, I-Lan, and Hua-Tung AQRs. Taiwan's limited land area necessitates judicious use of local resources. For instance, several fishing harbors are located in the Kao-Ping AQR, and the Central and Yun-Chia-Nan AQRs have an abundance of fields for grain and vegetables cultivation. These factors could also affect the dietary habits of residents in different AQRs. Air monitoring data for PCDD/Fs from 2013 to 2017, derived from Taiwan EPA, indicated that the highest values were recorded in the Yun-Chia-Nan AQR, followed by Kao-Ping, Central, Chu-Miao, North, and the Hua-Tung AQRs. Clarifying whether PCDD/F and DL-PCB levels in the air affect the dietary measurement of PCDD/Fs and DL-PCBs of residents in each AQR is imperative. Across various regions in Taiwan, considerable differences were observed in terms of the dietary exposure of different age groups and the pattern of contribution of food groups to total exposure because of different contamination levels and dietary habits.

In this study, we evaluated chronic dietary exposure to PCDD/Fs and DL-PCBs of people across Taiwan's AQRs to assess the health risks derived from them. We compared total dioxin intake to identify any time trends or geographical differences. Our results are useful for temporal trend analysis to evaluate the effectiveness of the dioxin strategy implemented by Taiwan EPA.

2. Materials and methods

2.1. Sampling strategy

We integrated results from two-stage food sampling to monitor PCDD/Fs and DL-PCBs (Fig. 1). Between 2004 and 2012, we have conducted a monitoring program on PCDD/Fs and DL-PCBs in foods from traditional markets or supermarkets in selected towns around Taiwan using the European Commission's standard for dioxins (stage I) [13]. During that period, we collected high-lipid food, cereals, fruits and vegetables, and various types of processed foods in each town that produced them in greatest quantity. However, a representative dataset on food consumption is more appropriate to derive dietary exposure. Therefore, in the following stage during 2013−2018, we started a new study (stage II), to monitor the background levels of PCDD/Fs and DL-PCBs in selected foods based on Taiwanese dietary habits derived from the Nutrition and Health Survey in Taiwan (NAHSIT) were monitored in Taiwan's AQRs in sequence [14]. Levels were monitored first in the North (including Taipei, New Taipei, and Taoyuan City) in 2013, Kao-Ping (Kaohsiung and Pingtung City) in 2014, Yun-Chia-Nan (Yunlin, Chiayi, and Tainan City) in 2015, Central (Taichung, Changhua, and Nantou City) in 2016, Chu-Miao (Hsinchu and Miaoli City) in 2017, and Hua-Tung (Hualien and Taitung City) in 2018. In the NAHSIT, a multistage, stratified, probability sampling design was employed to select participants representative of the Taiwanese population for all ages, and then face-to-face interviews were conducted. Consequently, a new study was conducted in six Taiwan AQRs, in which a representative dataset on food consumption was combined with data on the concentration of the compounds of interest in foods to derive the exposure.

Abbreviations

PCDD/Fs	polychlorinated dibenzo-p-dioxins and dibenzofurans
DL-PCBs	dioxin-like polychlorinated biphenyls
AQRs	air quality regions
EFSA	European Food Safety Authority
EPA	Environmental Protection Agency
NAHSIT	Nutrition and Health Survey in Taiwan
TWI	tolerable weekly intake
LADDs	lifetime average daily doses
CONTAM	Contaminants in the Food Chain

2.2. Foodstuff sampling criteria

The first step consisted of establishing the list of foods to be analyzed. Food items were selected on the basis of the following criteria: the foods most consumed in terms of quantity, (with a consumption rate of at least >2 g person^{-1} day^{-1}) and the main known or assumed contributors to PCDD/Fs and DL-PCBs exposure, such as meat, poultry, seafood,

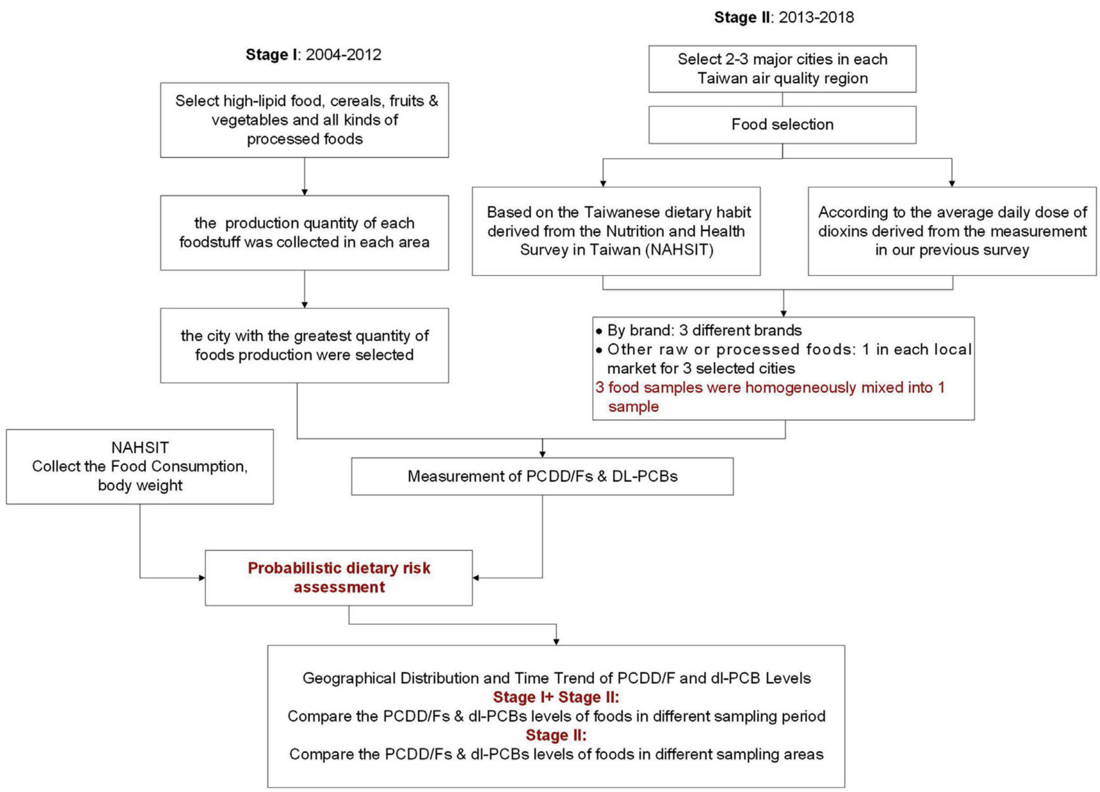

Fig. 1. Study flow.

milk, eggs, and their products. We also considered the daily intake of PCDD/Fs and DL-PCBs in foodstuffs from our previous measurements [13].

Furthermore, we collected and integrated the quantity of production of each foodstuff in every county, village, and town in each AQR. The foodstuff samples including the raw sample and brand sample were purchased from traditional markets or supermarkets in selected towns around Taiwan from 2004 to 2018. The raw foods produced in the greatest quantities in each county were selected for analysis. We have also confirmed that the raw sample were mainly produced from the local farms, pastures or fisheries from the inquiry from the vendors. The individual raw food sample for each AQR was purchased in two or three cities. Finally, the same matrix of three raw food samples was homogeneously mixed into one food sample and then frozen at −20 °C until analysis. For example, 600 g of the pork composite sample was prepared by separating pork samples of 200–300 g purchased from three cities. In addition, the individual brand sample, such as dairy products, seasonings, composite foods and soups, and beverages were purchased from famous brands based on the market share. And the brand sample was collected and analyzed individually. Finally, 2441 individual foods in 14 categories were collected and analyzed (stage I, n = 1716; stage II, n = 725). The investigated samples were divided into 14 categories as follows. Cereals, grains, tubers and roots: rice and its products, wheat and its products, and carbohydrate-rich tubers, roots, and their products (sample size = 138); beans and nuts: beans, processed bean products, and nuts and its products (sample size = 39); fish and aquatic products: freshwater fish, marine fish, fish and its products, and other aquatic animals and their products (sample size = 546); meats: beef, pork, mutton, chicken, duck, and goose (sample size = 585); dairy: whole milk, low-fat/fat-free milk, whole sheep milk, fermented milk, other milk, powdered milk, and cheese (sample size = 281); eggs (sample size = 196), cereals (sample size = 138), fruits (sample size = 70); vegetables: leafy vegetables, fruit crops, bean sprouts, gourd, stem vegetables, mushrooms, and others (sample size = 371); and fats and oils (sample size = 41). All these foodstuffs were prepared as described above. The details of the geographical origin of the samples are reported in the Supplementary Materials (Table S1).

2.3. High-resolution gas chromatography/high-resolution mass spectrometry for PCDD/Fs + DL-PCBs

Isotope dilution high-resolution gas chromatography/high-resolution mass spectrometry was employed to determine the levels of 17 PCDD/Fs and 12 DL-PCBs in fish, seafood, meats, eggs, milk, dairy products, and oil samples, as described previously [13]. Analytical procedures were adopted from the US Environmental Protection Agency (USEPA) Methods 1613B [14] and 1668A [15], with minor modifications. Three extraction procedures (I, II, and III) were applied for various sample matrices. Quality assurance and quality control protocols were established in the laboratory according to those defined in USEPA Method 1668A [15] to ensure positive identification and measurement quality. The quality assurance and quality control protocols included mass spectrometry resolution, gas chromatography resolution, calibration verification, ongoing precision and recovery, blank, and internal standard recovery. The analytical laboratory, Trace Environmental Pollutant, Research Center of Environmental Trace Toxic Substances (RCETTS), at National Cheng Kung University in Tainan, Taiwan, is certified by the Taiwan Accreditation Foundation and responsible for all the analyses. We have participated the interlaboratory Comparison on Dioxins in Food which were to assess the in-between laboratory reproducibility, to offer a quality assurance instrument regularly and have a good performance (Table S2). In addition, we have also ascertained that the recovery of internal standard in all samples for PCDD/Fs and DL-PCBs were meet the criteria which was shown in Table S3.

LODs for all measured analytes were estimated dynamically during the specific period of analysis and were dependent on parameters such as sample weight, type of matrix and instrument performance at the time of measurement. Typical LODs were 0.01−0.021 pg g^{-1} lipid for PCDD/Fs and 0.102 to 0.564 for DL-PCBs (Table S4). We have also randomly analyzed Certified Reference Material (CRM) samples in routine sample analysis every six months. The analysis results were also meet with the criteria of reference value (Table S5).

The PCDD/F + DL-PCB concentrations were stated as fat weight and wet weight (pg World Health Organization-Toxic Equivalent (WHO-TEQ) g^{-1} fat, and pg WHO-TEQ g^{-1} wet weight [w.w.]).

2.4. Exposure assessment

In the intake calculations, the dietary intake of PCDD/Fs and DL-PCBs was first calculated by multiplying the daily consumption by the mean TEQ of PCDD/Fs and DL-PCBs for each food type. To further calculate daily intake (in pg kg^{-1} body weight [b.w.]), the average weights of the members of each sex and age group were used; values were also obtained from the NAHSIT [14]. The TEQ data of the 17 PCDD/Fs and 12 DL-PCBs congeners were determined with respect to WHO_{2005} Toxic Equivalency Factors (TEFs). Intake was calculated using 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) according to EFSA recommendations [16]. According to EU analytical regulations for foodstuffs, it requires the difference between UB and LB values to be less than 20% for confirmations of regulatory maximum exceedances (Commission Regulation 589/2014). We presented summary analyte concentrations in UB values, and are thus precautionary, ‘worst case’ estimates.

We estimated the average daily dose of PCDD/Fs + DL-PCBs based on the ingestion rate of foodstuffs from a sex- and age-specific population database derived from the NAHSIT conducted in 2001−2002 and 2005−2008 and from the measured concentration of PCDD/Fs + DL-PCBs in the corresponding food items. The estimated daily intake (EDI) was evaluated using a probabilistic approach. Intake calculations were performed using @RISK, a Monte Carlo computational system for stochastic modeling of dietary exposure [17]. The exposure of a randomly selected person from the consumption database was the result of multiplying the consumption of each relevant foodstuff the person consumed in one day by a randomly selected concentration per commodity from the concentrations database. To model the intake as accurately as possible, this calculation was repeated 10,000 times and a sensitivity analysis was performed during each model run. The sensitivity analysis helped identify which of the selected model parameters had the greatest effect on the output parameter—initial dioxin concentration in food—by determining the input parameter's contribution to the variance of the output parameter. The different possible outcomes generated iteratively were assembled to create a probabilistic statement of the range of results obtained. A distribution of daily intake was thus generated, including variability and uncertainties.

Table 1. Distribution of PCDD/Fs and DL-PCBs in Taiwan food from 2004 to 2018.

Food group	N	pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} wet weight	pg WHO_{05}-TEQ_{PCB} g^{-1} wet weight	pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} wet weight
Cereals, grains, tubers and roots				
Rice and its products	65	0.016 (0.001−0.042)	0.002 (<0.001−0.01)	0.018 (0.002−0.044)
Wheat and its products	52	0.015 (0.003−0.048)	0.002 (<0.001−0.011)	0.017 (0.003−0.050)
Carbohydrate's tubers, roots, and their products	21	0.009 (0.002−0.044)	0.001 (<0.001−0.005)	0.010 (0.002−0.047)
Beans and nuts				
Beans	15	0.029 (0.006−0.066)	0.005 (0.001−0.014)	0.034 (0.008−0.070)
Bean processed products	20	0.005 (0.002−0.024)	0.001 (<0.001−0.004)	0.005 (0.002−0.024)
Nuts and its products	4	0.017 (0.010−0.022)	0.003 (0.002−0.003)	0.020 (0.014−0.024)
Fats and oils				
Vegetable oils	28	0.073 (0.022−0.299)	0.010 (0.003−0.036)	0.084 (0.025−0.305)
Animal fats	8	0.109 (0.063−0.169)	0.055 (0.028−0.129)	0.164 (0.092−0.251)
Others	5	0.018 (0.007−0.035)	0.002 (0.001−0.004)	0.020 (0.010−0.039)
Poultry and their products				
Chicken and its products	96	0.023 (0.006−0.260)	0.009 (0.002−0.039)	0.031 (0.009−0.265)
Duck and its products	88	0.055 (0.004−0.503)	0.033 (0.002−0.663)	0.088 (0.007−0.782)
Goose and its products	63	0.055 (0.010−0.256)	0.029 (0.004−0.116)	0.084 (0.014−0.306)
Livestock and their products				
Pork and its products	163	0.020 (0.003−0.185)	0.012 (<0.001−0.110)	0.032 (0.003−0.265)
Beef and its products	94	0.064 (0.004−0.442)	0.004 (0.001−0.410)	0.104 (0.005−0.809)
Mutton and its products	81	0.109 (0.003−1.281)	0.070 (0.001−0.833)	0.179 (0.004−2.067)
Fish and Aquatic Products				
Freshwater fish	70	0.100 (0.012−0.568)	0.147 (0.010−1.011)	0.246 (0.033−1.327)
Marine fish	266	0.118 (0.003−3.330)	0.359 (0.001−9.036)	0.477 (0.005−12.365)
Fish and its products	89	0.066 (0.005−0.527)	0.155 (0.003−1.378)	0.220 (0.008−1.582)
Other aquatic animals and their products	121	0.076 (0.004−0.899)	0.116 (0.002−3.839)	0.192 (0.005−4.668)
Food group	N	pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} wet weight	pg WHO_{05}-TEQ_{PCB} g^{-1} wet weight	pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} wet weight
Eggs				
Chicken eggs	89	0.040 (0.009−0.194)	0.0124 (0.018−0.104)	0.052 (0.011−0.202)
Duck eggs	63	0.149 (0.025−2.473)	0.033 (0.002−0.663)	0.211 (0.038−2.622)
Other eggs	44	0.095 (0.014−0.516)	0.053 (0.004−0.482)	0.148 (0.020−0.997)
Dairy				
Whole fat milk	204	0.023 (0.003−0.089)	0.014 (0.001−0.058)	0.037 (0.004−0.142)
Low fat/fat free milk	6	0.010 (0.003−0.017)	0.004 (0.002−0.009)	0.014 (0.005−0.026)
Whole fat sheep milk	24	0.020 (0.009−0.037)	0.013 (0.005−0.020)	0.034 (0.014−0.054)
Fermented milk	14	0.013 (0.001−0.043)	0.006 (0.001−0.021)	0.019 (0.002−0.064)
Other milk	10	0.025 (0.003−0.111)	0.010 (<0.001−0.038)	0.035 (0.003−0.149)
Powdered milk	13	0.033 (0.005−0.082)	0.014 (0.002−0.037)	0.047 (0.007−0.117)
Cheese	13	0.119 (0.026−0.346)	0.075 (0.009−0.174)	0.194 (0.036−0.505)
Fruits				
Berries	32	0.005 (0.001−0.024)	0.001 (<0.001−0.004)	0.006 (0.002−0.027)
Pomaceous fruits	9	0.004 (0.001−0.007)	<0.001 (<0.001−0.001)	0.004 (0.001−0.009)
Stone fruits	9	0.006 (0.001−0.014)	0.001 (<0.001−0.001)	0.006 (0.001−0.015)
Melon and fruit	6	0.002 (0.001−0.004)	<0.001 (<0.001−0.001)	0.002 (0.001−0.004)
Citrus Fruit	9	0.004 (0.002−0.010)	0.001 (<0.001−0.001)	0.005 (0.002−0.011)
Sugar-cane	5	0.005 (0.003−0.006)	0.001 (<0.001−0.001)	0.006 (0.004−0.007)
Vegetables				
Leafy vegetables	203	0.011 (<0.001−0.294)	0.004 (<0.001−0.229)	0.014 (<0.001−0.295)
Fruit crops	12	0.005 (0.001−0.016)	0.001 (<0.001−0.003)	0.006 (0.001−0.019)
Bean sprouts	16	0.006 (0.001−0.025)	0.001 (<0.001−0.002)	0.007 (0.001−0.027)
Gourd	25	0.002 (0.001−0.008)	<0.001 (<0.001−0.001)	0.002 (0.001−0.009)
Stem vegetables	76	0.005 (0.001−0.077)	0.001 (<0.001−0.018)	0.006 (0.001−0.095)
Mushrooms	32	0.008 (0.001−0.033)	0.001 (<0.001−0.003)	0.009 (0.001−0.035)
Others	7	0.007 (0.001−0.020)	0.001 (<0.001−0.005)	0.008 (0.001−0.025)
Food group	N	pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} wet weight	pg WHO_{05}-TEQ_{PCB} g^{-1} wet weight	pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} wet weight
Seasonings				

(*continued on next page*)

Table 1. (continued)

Food group	N	pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} wet weight	pg WHO_{05}-TEQ_{PCB} g^{-1} wet weight	pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} wet weight
Salt	5	0.013 (0.007−0.024)	0.002 (0.001−0.002)	0.015 (0.009−0.025)
MSG	1	0.005	0.001	0.006
Soy sauce	18	0.014 (0.001−0.036)	0.004 (<0.001−0.019)	0.028 (0.002−0.197)
Curry sauce	17	0.021 (0.005−0.183)	0.007 (0.001−0.062)	0.028 (0.006−0.245)
Composite foods and Soups				
Rice	22	0.008 (0.003−0.041)	0.001 (<0.001−0.004)	0.009 (0.004−0.044)
Wheat	90	0.018 (0.003−0.052)	0.007 (<0.001−0.032)	0.025 (0.004−0.069)
Others	2	0.031, 0.099	0.003, 0.089	0.013, 0.120
Candies and Snacks	11	0.013 (0.007−0.021)	0.005 (<0.001−0.015)	0.018 (0.007−0.025)
Beverages	5	0.004 (0.003−0.005)	0.0004 (<0.001−0.001)	0.005 (0.003−0.006)

Note: PCDDs, polychlorinated dibenzo-*p*-dioxins; PCDFs, polychlorinated dibenzofurans; DL-PCBs, dioxin-like polychlorinated biphenyls.

The toxicity of PCDD/Fs and DL-PCBs is related to the amount accumulated in the body during a lifetime, the so-called body burden. A tolerable weekly intake (TWI) of 14 pg WHO-TEQ kg^{-1} b.w. has been established by the Scientific Committee on Food [18]. Likewise, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) set up a provisional tolerable monthly intake (PTMI) of 70 pg WHO-TEQ kg^{-1} b.w. $month^{-1}$ [19].

The statistical significance between food PCDD/Fs and DL-PCBs among different food category and AQRs was evaluated by one-way ANOVA. In addition, PCDD/F + DL-PCB levels among the different sampling periods was evaluated by linear trend test. SPSS 22 was used for all analyses. Significance was set at $P < 0.05$.

3. Results

3.1. PCDD/F and PCB concentration in foodstuffs

Table 1 presents the sample size for each location and the PCDD/F and DL-PCB levels in alternative food categories. For PCDD/F levels in different foodstuff, the highest levels were observed in duck eggs (average, 0.149 pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} w.w.), followed by cheese (0.119 pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} w.w.) > marine fish (0.118 pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} w.w.) > mutton (0.109 pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} w.w.) > animal fats (0.109 pg WHO_{05}-$TEQ_{PCDD/F}$ g^{-1} w.w.) ($p < 0.001$). For DL-PCB levels in different foodstuffs, the highest levels were observed in marine fish (average, 0.359 pg WHO_{05}-TEQ_{PCB} g^{-1} w.w.), followed by fish and its

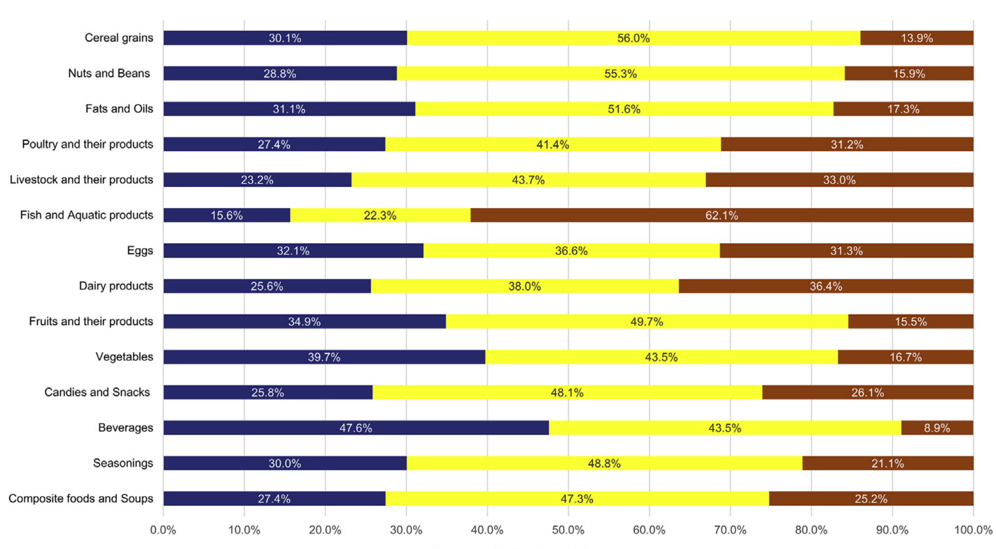

Fig. 2. Percentage of contribution from each food group to the TEQ levels of PCDD/Fs and DL-PCBs. TEQ: toxic equivalent, PCDD/Fs: polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans, DL-PCBs: dioxin-like polychlorinated biphenyls.

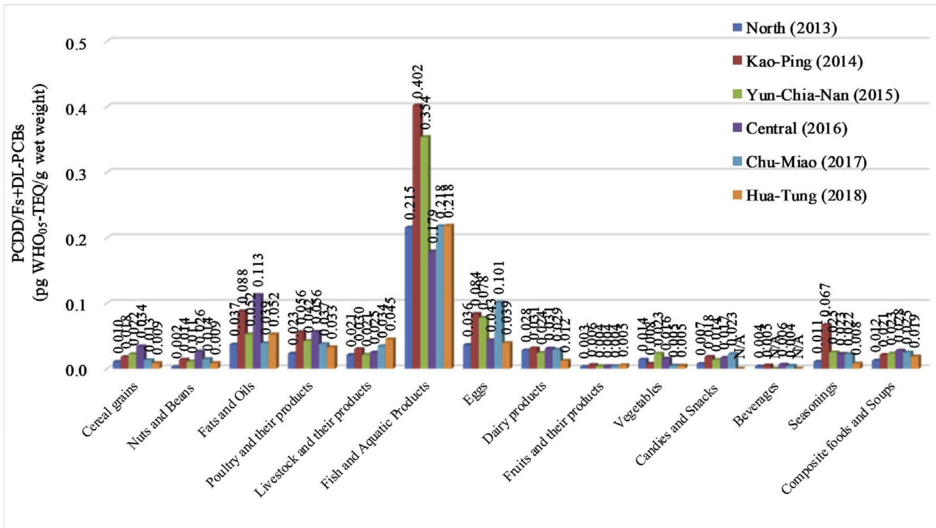

Fig. 3. Total PCDD/F and DL-PCB levels of different food samples in the six AQRs, expressed as pg WHO_{05}-TEQ g^{-1} w.w. PCDD/F: polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans, DL-PCB: dioxin-like polychlorinated biphenyls, AQRs air quality regions, WHO: World Health Organization, TEQ: toxic equivalent.

products (0.155 pg WHO_{05}-TEQ_{PCB} g^{-1} w.w.) > freshwater fish (0.147 pg WHO_{05}-TEQ_{PCB} g^{-1} w.w.) > other aquatic animals and their products (0.116 pg WHO_{05}-TEQ_{PCB} g^{-1} w.w.) > cheese (0.075 pg WHO_{05}-TEQ_{PCB} g^{-1} w.w.) ($p < 0.001$). For total PCDD/Fs + DL-PCBs levels, the highest levels were observed in marine fish (average, 0.477 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.), followed by freshwater fish (0.246 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.) > fish and its products (0.220 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.) > duck eggs (0.211 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.) > cheese (0.194 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.) ($p < 0.001$). These measurements generally had either no or low difference between UB and LB sum values, the greatest difference being 5.07% for vegetables, was within the required 20% (Commission Regulation 589/2014) (Table S6).

The ratio of DL-PCBs to PCDD/Fs is exhibited in Fig. 2. In fish and seafood, DL-PCBs contributed 62.1 ± 19.1% of the TEQ levels, followed by dairy products (36.4 ± 6.8%) and livestock and their products (33 ± 13.2%), whereas in beverage samples the contribution of DL-PCBs was only 8.9 ± 2.2% ($p < 0.001$). The DL-PCB contributions were 33.0 ± 13.2%, 62.1 ± 19.1%, 31.3 ± 13.8%, and 36.4 ± 6.9% for livestock products, fish and aquatic products, eggs, and dairy samples, respectively ($p < 0.001$); these data agree with the current findings indicating that PCBs contribute more than 50% of dioxin-like components from fish and seafood.

PCDD/F + DL-PCB levels among the different sampling periods exhibited a decreasing trend in fish and aquatic products (from 0.384 ± 0.764 to 0.206 ± 0.223 pg WHO_{05}-TEQ g^{-1} w.w.) (p for trend = 0.043), livestock products (from 0.133 ± 0.298 to 0.035 ± 0.043 pg WHO_{05}-TEQ g^{-1} w.w.), eggs (from 0.221 ± 0.373 to 0.056 ± 0.048 pg WHO_{05}-TEQ g^{-1} w.w.) (p for trend = 0.002), and dairy samples (from 0.066 ± 0.075 to 0.024 ± 0.026 pg WHO_{05}-TEQ g^{-1} w.w.) (p for trend = 0.001). The only exceptions to this trend were poultry products, fish, and seafood. All vegetables exhibited levels lower than 0.016 pg WHO_{05}-TEQ g^{-1} w.w. (Fig. S1). We also compared the PCDD/F + DL-PCB levels in milk from 2004 to 2015 with the emission inventory of PCDD/Fs in Taiwan. Both of these revealed a significant decreasing trend after 2006 (p for trend< 0.001), demonstrating the effectiveness of the dioxin reduction strategy implemented by the Taiwan EPA.

3.2. Distribution of PCDD/F + DL-PCB levels and daily intake in AQRs

Figure 3 displays the PCDD/F + DL-PCB levels in the different sampling AQRs. In all AQRs, the PCDD/F + DL-PCB levels were highest in fish and aquatic products, followed by eggs. In addition, a geographic variation of PCDD/F + DL-PCB levels was observed among the different sampling AQRs. The highest PCDD/F + DL-PCB levels were observed in fish and aquatic products in the Kao-Ping AQR (0.402 ± 0.532 pg WHO_{05}-TEQ g^{-1} w.w.), followed by the Yun-Chia-Nan AQR (0.354 ± 0.494). The lowest PCDD/F + DL-PCB levels were encountered in the Central AQR (0.179 ± 0.209)

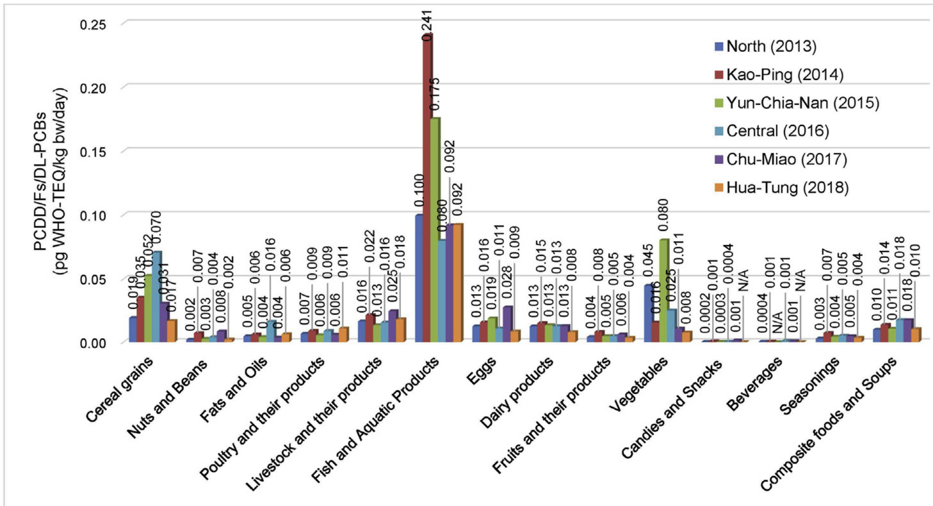

Fig. 4. Distribution of total daily intake of PCDD/Fs and DL-PCBs by Taiwanese according to area and food category. PCDD/Fs: polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans, DL-PCBs: dioxin-like polychlorinated biphenyls.

(p = 0.124). For cereal grains (0.034 ± 0.015), the PCDD/F + DL-PCB levels measured in the Central AQR were significantly higher than in other AQRs ($p < 0.001$).

The total daily intake of PCDD/Fs and DL-PCBs by Taiwanese differed between AQRs (Fig. 4). The highest total daily intake (mainly from fish and aquatic products) was observed in the Kao-Ping AQR at 0.241 pg WHO_{05}-TEQ kg^{-1} b.w. day^{-1}, followed by 0.175 pg WHO_{05}-TEQ kg^{-1} b.w. day^{-1} in the Yun-Chia-Nan AQR. The lowest daily intake of PCDD/Fs + DL-PCBs were observed in the Central AQR (0.080). Cereal grains contributed the second highest exposure dose. The total daily intake of PCDD/Fs + DL-PCBs from cereal grains was higher in the Central (0.070) and Yun-Chia-Nan AQRs (0.052) than in other AQRs. Notably, the highest daily intake of PCDD/Fs + DL-PCBs from vegetables were observed in the Yun-Chia-Nan AQR (0.080), followed by the North AQR (0.045 pg WHO_{05}-TEQ kg^{-1} b.w. day^{-1}).

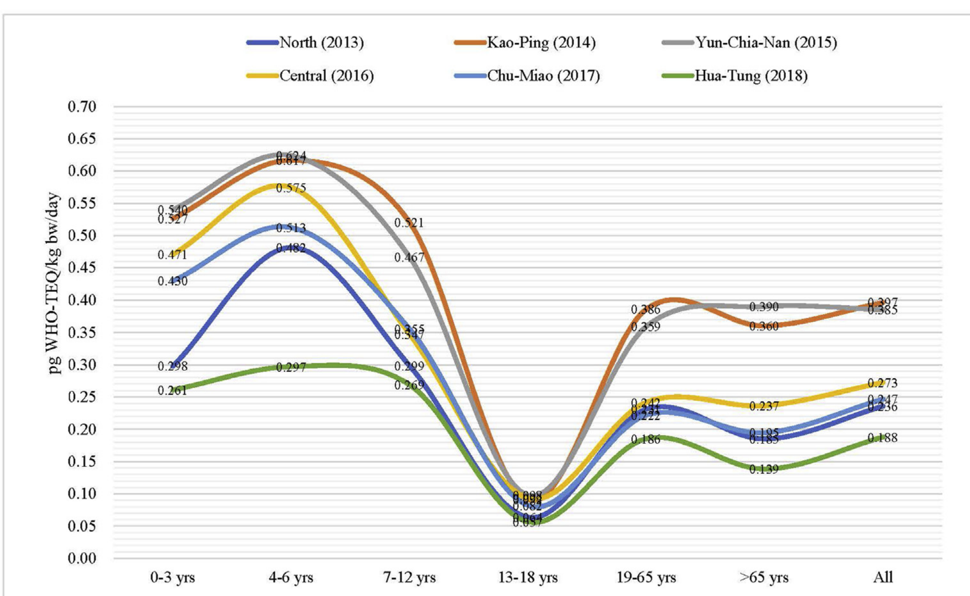

Fig. 5. Distribution of total daily intake of PCDD/Fs and DL-PCBs by Taiwanese in different age groups in 6 AQRs. PCDD/Fs: polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans, DL-PCBs: dioxin-like polychlorinated biphenyls, AQRs: air quality regions.

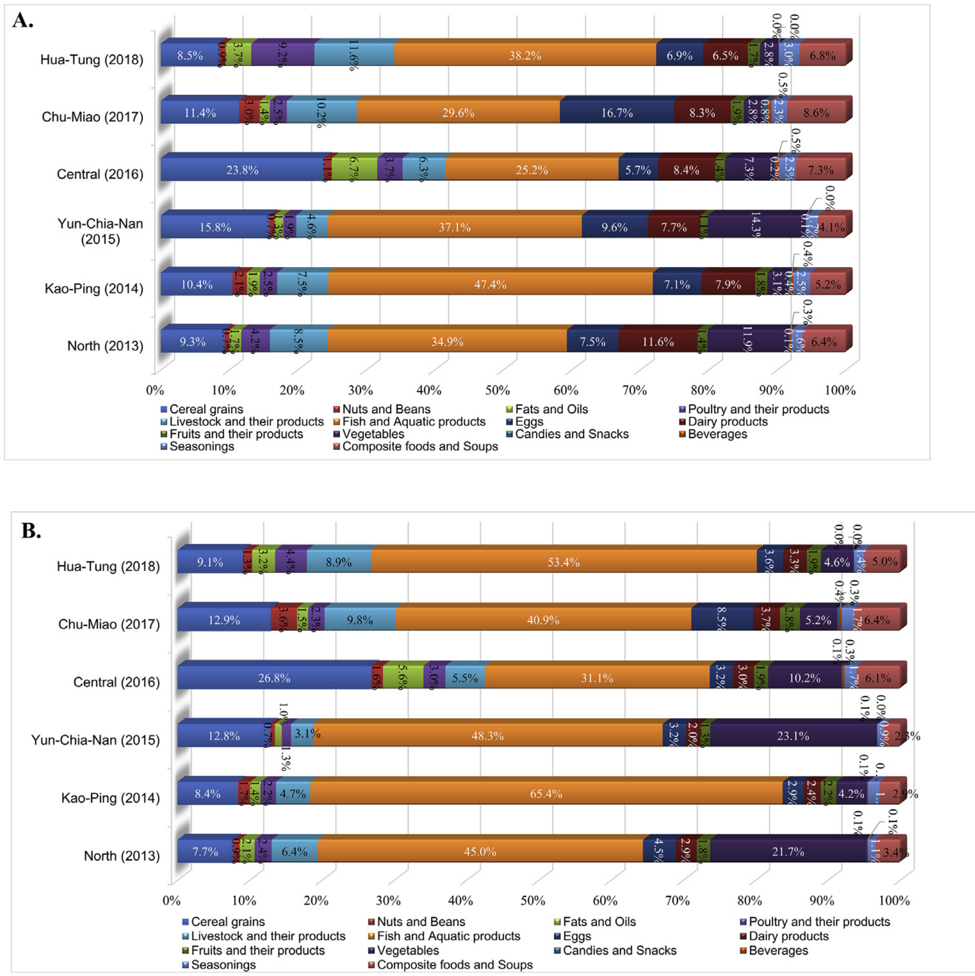

Fig. 6. Percentage of contribution from each food group to total daily intake of PCDD/Fs and DL-PCBs by Taiwanese in six AQRs in (A) < 18 and (B) ≥18 years age groups. PCDD/Fs: polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans, DL-PCBs: dioxin-like polychlorinated biphenyls, AQRs: air quality regions.

The highest daily intake for residents was observed in Kao-Ping (at 0.397 pg WHO_{05}-TEQ kg^{-1} b.w. day^{-1}), followed by Yun-Chia-Nan (0.385), Central (0.273), Chu-Miao (0.247), and North (0.236); the lowest levels were observed in Hua-Tung (0.188). All the lifetime average daily doses (LADDs), which were calculated on the basis of the measurements of 14 food groups from six locations in Taiwan, were below provisional tolerable monthly intake (PTMI) of 70 pg WHO-TEQ kg^{-1} b.w. $month^{-1}$ but higher than the new TWI for PCDD/Fs and DL-PCBs in food, 2 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ kg^{-1} b.w. $week^{-1}$, as published by EFSA's Panel on Contaminants in the Food Chain (CONTAM) [20]. We also calculate the TEQ using another three model, which are TEF 2005 lower bond, TEF 1998 upper bond and TEF 1998 lower bond, respectively. All of them were below the PTMI but higher than the TWI (Fig. S2). The EDI of PCDD/Fs and DL-PCBs by the general population in different AQRs in Taiwan, classified according to age group, is depicted in Fig. 5. In general, children had higher PCDD/F and DL-PCB dietary intakes than adult groups because of their lower body weight. A sharp decrease was observed in the 13–18-year-old group because of their relatively lower fish consumption. In the present survey, none of the age groups exceeded the PTMI.

3.3. Contribution of each foodstuff to PCDD/Fs + DL-PCBs in AQRs

The percentages of the contribution of each food group to the total dietary intake of $TEQ_{PCDD/F+PCB}$

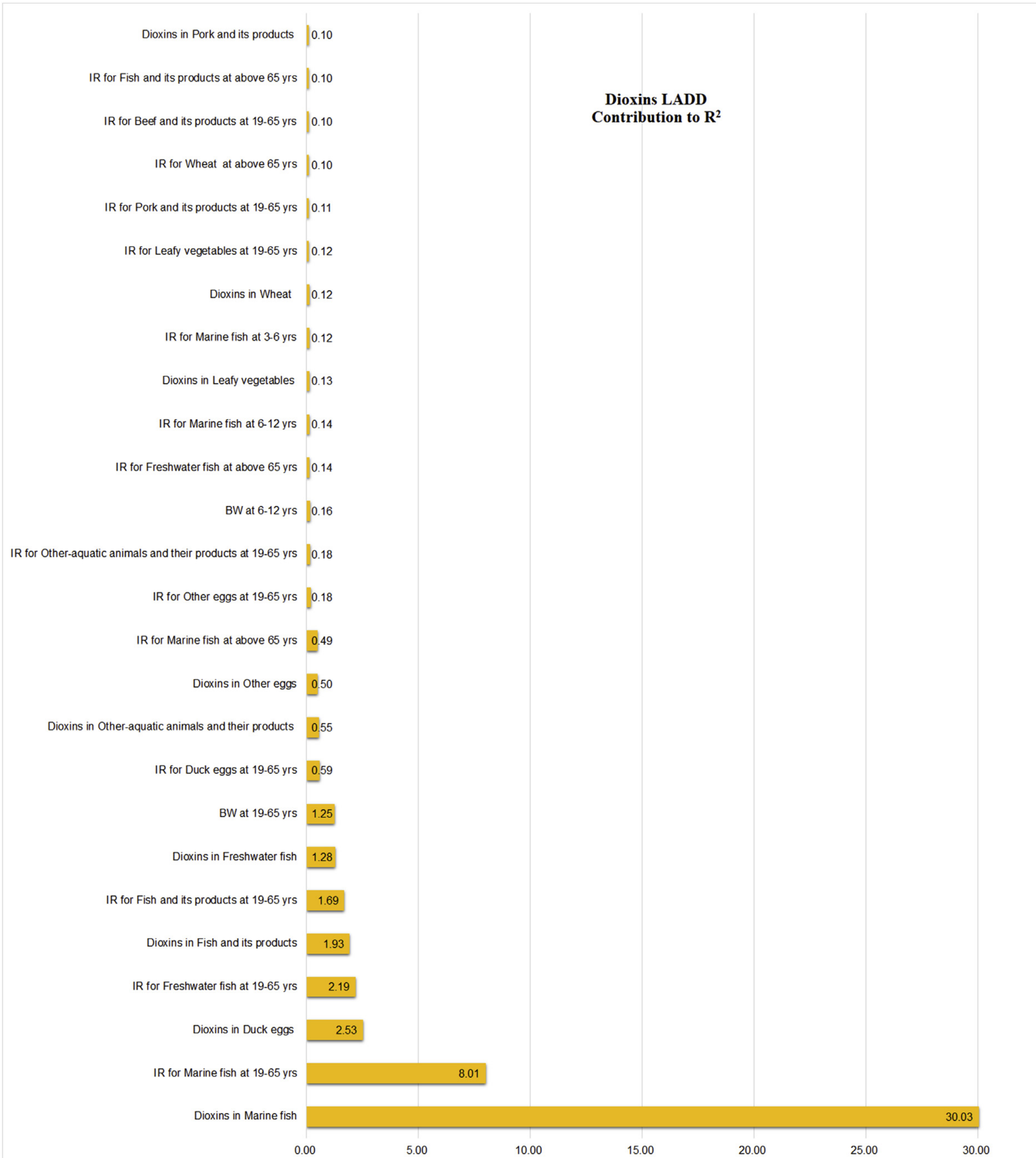

Fig. 7. Sensitivity analysis showing percent contribution to variance for LADD. Note: we neglected variable which contribution was less than 0.1.

in different ambient air dispersion areas and age groups are depicted in Fig. 6A and B. Fish and aquatic products contributed most to the PCDD/Fs and DL-PCBs intake for participants younger than 18 years. Fish and aquatic products are by far (25.2%−47.4%) the main contributor to total exposure to PCDD/Fs and DL-PCBs. The highest contribution of total $TEQ_{PCDD/F+PCB}$ from fish and aquatic products was observed in the Kao-Ping AQR (47.4%), followed by Hua-Tung (38.2%), and Yun-Chia-Nan (37.1%). The lowest contribution was observed in the Central AQR (25.2%). In addition, the major contribution of total $TEQ_{PCDD/F+PCB}$ from cereal grains and vegetables was higher in the Central AQR (31.1%), followed by Yun-Chia-Nan (30.1%) in participants younger than 18 years. The lowest contribution of total $TEQ_{PCDD/F+PCB}$ from cereal grains and vegetable (11.3%) was observed in the Hua-Tung AQR, and the highest contribution from poultry, livestock, and their products (20.8%) was also observed in this area (Fig. 6A).

However, this contribution pattern changed markedly in participants older than 18 years (Fig. 6B). In the Kao-Ping and Hua-Tung AQRs, more than half of total $TEQ_{PCDD/F+PCB}$ exposure was from fish and aquatic products (65.4% and 53.4%, respectively). In the Central and Yun-Chia-Nan AQRs, the contribution of total $TEQ_{PCDD/F+PCB}$ from cereal grains and vegetables were 37% and 35.9%, respectively. These contributions to total $TEQ_{PCDD/F+PCB}$ from low-fat foodstuff (8.5% and 4.4%) notably exceeded the contributions of poultry, livestock, and their products.

Comparing intake estimations between studies from other countries is a difficult task because of the differences in methodologies, the food groups considered, the population groups studied, and the manner results were reported, despite the main factor affecting variability being the dietary habits in the population.

3.4. Monte Carlo sensitivity analysis

Sensitivity analysis was done based on effective variables on risk assessment such as concentration (C) of PCDD/F and DL-PCBs, body weight (BW), and food intake rate (IR). In this study, sensitivity analysis was performed to determine the most effective variable in increasing the carcinogenic risk through dioxins using Monte Carlo simulations. Fig. 7 indicates sensitivity analyses of LADD for exposure to PCDD/F and DL-PCBs (Dioxins) in different food. According to Fig. 7, the concentration of dioxins in Marine fish was the most effective variable in increasing the LADD (contribution to variance was 30.03%). The other effective parameters in increasing the LADD for consumers was IR (Marine fish) and the concentration of dioxins in duck eggs, respectively. Increased body weight (BW) had an inverse relationship with LADD (contribution to variance was −1.25% from 19 to 65 years old).

4. Discussion

4.1. PCDD/Fs and DL-PCBs concentration in foodstuffs of different countries

In this study, we the integrated PCDD/F + DL-PCB levels of 2441 foodstuffs into 14 categories after two sampling stages. For total PCDD/F + DL-PCB levels, the highest levels were observed in marine fish (average, 0.477 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.), followed by freshwater fish (0.246 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.) > fish and its products (0.220 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.) > duck eggs (0.211 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.) > cheese (0.194 pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} w.w.). The levels of PCDD/Fs and DL-PCBs in the present study were lower than those reported by EFSA from samples collected during 1995−2010 from 24 European Union member states, Iceland, and Norway. In studies focusing on fish and seafood, levels reported in Greece (0.49 pg WHO-TEQ g^{-1} w.w.) [21] and France (0.65 pg WHO-TEQ g^{-1} w.w.) [8] were similar to those observed in the current study. For milk products, our measurements were lower than those observed in Belgium (1.74 pg TEQ g^{-1} fat) [22] and Kuwait (2.10 pg BEQ g^{-1} w.w.) [23]. In addition, almost all of the analyzed foodstuffs in this study were under the updated maximum level limit standards of the sum of dioxins, furans, and dioxin-like PCBs set by the European Commission (EN Commission Regulation No 199/2006).

4.2. PCDD/F and DL-PCB concentrations in foodstuffs in different AQRs

The large differences among regions presumably results from variations in contamination levels as well as food consumption habits in different AQRs. The highest PCDD/F + DL-PCB levels were observed in fish and aquatic products in the Kao-Ping AQR, followed by the Yun-Chia-Nan AQR. The lowest PCDD/F + DL-PCB levels were observed in the Central AQR. Different fishing types in each region could account for these differences. More deep-sea fishery occurs in the Kao-Ping AQR in comparison with offshore or inshore fishery. Therefore, residents might have more opportunities to consume predatory fish than farmed fish. In terms of cereal grains and vegetables, the PCDD/F + DL-PCB levels measured in the Central and Yun-Chia-Nan AQRs were higher than in other AQRs. These two AQRs have a high density of paddy fields. After harvesting, the rice straw is frequently burned in the open with insufficient time before planting the next crop to remove and dispose of it in a more controlled manner, such as in a furnace or by using another closed burning technique [24]. However, the burning of rice straw in fields may contribute to the emission of harmful air pollutants, such as polycyclic aromatic hydrocarbons, PCDDs, and PCDFs, threatening human health [25−27]. Consequently, numerous studies at

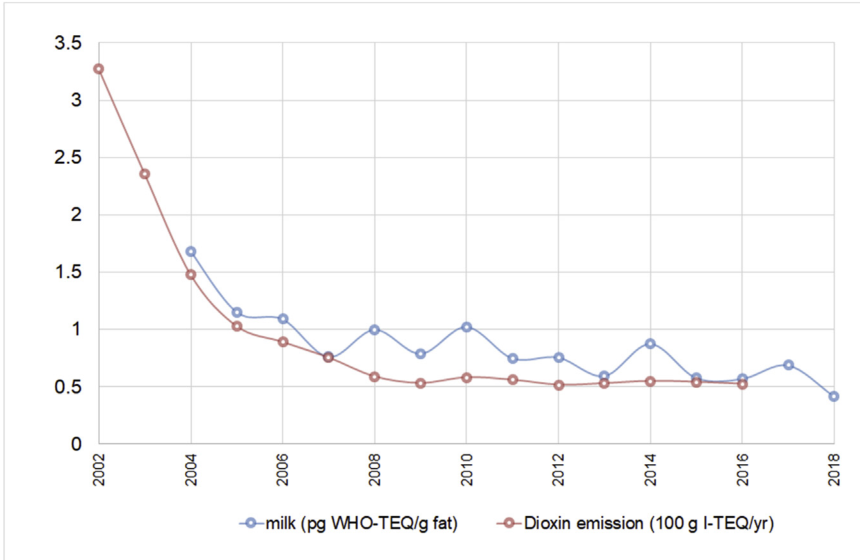

Fig. 8. Comparison of Environmental PCDD/Fs emissions and milk PCDD/Fs level from 2002 to 2016. PCDD/Fs: polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans.

the local and global levels have monitored and estimated air pollutant emissions caused by open rice straw burning [26,28]. These activities also increase the PCDD/F levels in the air and pollute nearby crops. The geographical distribution of dietary exposure in this study was consistent with that of the emission inventory and PCDD/F levels in the air.

4.3. PCDD/F and DL-PCB concentrations in milk samples over past decades in different AQRs

Between 2004 and 2018, we continuously monitored the PCDD/F + DL-PCB levels in milk (Fig. 8). 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 the milk. The mean value for the distribution of PCDD/Fs and DL-PCBs in milk fat in a 2004 survey was 1.68 pg WHO-TEQ g^{-1} fat. Levels were lower than 1 pg WHO-TEQ g^{-1} fat after 2011 (0.42–0.87 pg WHO-TEQ g^{-1} fat), which corresponds to a 48–75% decrease. The downward trend in milk sample mirrors the concomitant downward trend in total dioxin emissions in Taiwan. Several studies have reported a notable decline of contamination levels in food and dietary exposure to PCDD/Fs and DL-PCBs in the general population during the last decade, which probably results from strict regulations on dioxin emissions in some developed countries [22,29–33].

The frequent monitoring of dietary exposure to PCDD/Fs and PCBs since the 1990s revealed a reduction of between 29% and 68% in a period of 10–20 years in developed countries compared with baseline [31,34]. The EDI of total TEQ of PCDD/Fs and DL-PCBs was in a range of 1–2 pg WHO-TEQ kg^{-1} b.w. day^{-1} during 2001–2011. This trend was attributed to the decreasing of PCDD/Fs in meat and dairy products.

4.4. Contribution of food products in different AQRs

The contributions from various food groups vary considerably among AQRs in Taiwan (Fig. 6A and B). Meat and meat products contributed the most to dietary intake in half of the AQRs in this study including Hua-Tung and Chu-Miao AQRs, ranging from 12.7% to 20.8% in the <18 years age group and from 12.1% to 13.3% in the ≥18 years age group. Aquatic foods were the highest contributors in the Yun-Chia-Nan and Kao-Ping AQRs, ranging from 37.1% to 47.4% in the <18 years age group and from 48.3% to 65.4% in the ≥18 years age group. Egg and egg products, and dairy products contributed most in the Chu-Miao and North AQRs, ranging from 19.1% to 25.0% in <18 years age group and from 7.4% to 12.2% in the ≥18 years age group. Notably, although the total percentage of all animal origin food composites were predominant in all AQRs owing to the great amount of consumption, cereals and vegetables made a considerable contribution with ranges of 11.3%–31.3% and 12.6%–37.0% in the <18 and $\geq$ 18 years age group, respectively. Some studies have attributed the notable decline of dietary exposure observed in certain European countries and Japan to the enforcement of legislation to reduce exposure to PCDD/Fs and DL-PCBs and strict implementation of control measures [22,29–33]. In adults as well as in children and teenagers, fish remained the main contributor to exposure to total PCBs (59% and 48%, respectively). Fish was also the major contributor to PCDD/F + DL-PCB exposure (35% and 26%, respectively in both groups), followed by butter (16% and 17%, respectively). Dairy products also appear to be among the main contributors to exposure to PCDD/Fs + DL-PCBs. The cumulated contribution to PCDD/Fs + DL-PCBs of the four related food groups (milk, butter, cheese, and other dairy products) reached 37% in adults and 45% in children and teenagers. The differences in exposure (but not necessarily in contributions) can also be explained by changes in consumption habits between the assessments.

4.5. Distribution of daily intake in different AQRs

Large differences in dietary exposure in different age groups and pattern of contribution of food groups to total exposure were observed among AQRs because of different contamination levels and food habits. Average dietary exposure and even higher consumers of various subgroups were all below the PTMI recommended by JECFA.

However, studies differed in their use of concentration data from various years; lower, middle, or upper-bound estimates; selection of foods and composition of food groups; and calculation methods. Therefore, comparisons should be made cautiously. Adults from the Kao-Ping and Yun-Chia-Nan AQRs exhibited the highest intake of PCDD/Fs and DL-PCBs in total EDI, mainly contributed by fish and aquatic products. Dietary dioxin intakes are calculated by multiplying the dioxin concentration data by the corresponding food consumption data; therefore, both sets of data play a critical part. For the Kao-Ping AQR, the high EDI may be due to the high consumption of fish by adults (83.7 g/day) and the fact that fish exhibited the highest level of contamination among the seafood samples. Several factors could explain the reasons that the LADD of residents in the Kao-Ping AQR was the highest and that the main source of contribution was fish and aquatic products (0.241/0.397 = 60.7%). In this area, the port city of Kaohsiung plays a crucial role in deep-sea fishery. The PCDD/F + DL-PCB levels in fish and aquatic products and the intake of fish for the residents were both the highest compared with other regions.

Fish is the main contributing food group to exposure [35,36]. Domingo and Bocio (2007) reported that some populations who frequently consume high quantities of certain fish species could be significantly increasing their health risks because of exposure to dioxins and PCBs [37]. This phenomenon was also reported in Japan, where dietary intake was highest in fishing areas, followed by farming and urban [38]. In a different study, the mean dioxin concentrations in fishermen, farmers and controls were 161,369, 79,079 and 100,500 pg g fat^{-1}, respectively [39].

Notably, cereal grains and vegetables made a substantial contribution to exposure in the Central and Yun-Chia-Nan AQRs. Furthermore, we also observed more PCDD/Fs in the air in the Yun-Chia-Nan AQR than in other AQRs. When PCDD/Fs are released into the air, they can deposit locally on plants. Moreover, people in this area consume an abundance of cereal grains and vegetables. These phenomena were verified by lower air PCDD/F levels and lower dietary dioxin intake from these crops and plants in the Hua-Tung AQR. Differences in food habits, cuisines, culture and economic levels, and levels of environmental contamination among the various regions in Taiwan could explain the observed geographical variations.

4.6. Daily intake of PCDD/Fs and DL-PCBs in different countries

The daily intake of PCDD/Fs and DL-PCBs in Taiwanese can also be compared with data from other countries. In a recent Swedish market basket study, dioxin (PCDD/Fs and DL-PCBs) intake was 96 pg WHO-TEQ/day or 1.3 pg kg^{-1} b.w. day^{-1} [40]. In addition, PCDD/F and DL-PCB intake estimations for adult populations from other countries (in pg WHO-TEQ kg^{-1} b.w. day^{-1}) was estimated at 0.57 pg in France [8], 0.52 pg in the United Kingdom [41], 0.61 in Belgium [22], 0.3 in Ireland [42], 1.13–1.58 pg in Spain [43], 0.28 pg in Italy [44], 1.06 in Japan [38], 1.36 in China [36], 1.5 in Finland [45] and 0.12–0.52 in Australia [46]. In addition, the total intake (PCDD/Fs + DL-PCBs) for local residents is slightly lower than that estimated for most EU countries by EFSA's CONTAM Panel [11,47] (Table S7). The estimated total intake of the population in this study (total TEQ: 0.188–0.397 pg kg^{-1} b.w. day^{-1}) was below the internationally acceptable intake limits (total TEQ: 2 pg kg^{-1} b.w. day^{-1} set by

SCF). Regarding young people, the estimated intake in the present study is much lower than the values (1.08−2.54 pg kg^{-1} b.w. day^{-1}) reported by EFSA's CONTAM Panel [11,47]. For children, limited choice of food and relatively lower body weight would lead to higher exposure compared with adults. Several articles have revealed significantly higher exposure in children than in adults [30,48,49].

4.7. The percentage contributions of different input parameters to LADD output

We use the sensitivity analysis to evaluate which of the input parameters have a more dominant influence on the uncertainty in the model output and quantifies the contribution that each input factor makes to the variance in the output quantity of interest [50]. From the result, the dioxin levels in marine fish, freshwater fish and fish related products contribute more than 33.2%, and followed by dioxin levels in duck eggs. In addition, marine and freshwater fish consumption rate accounts more than 10.2%. These results represent the major exposure scenario for the general Taiwanese and was consistent with the findings of daily intake in different AQRs. In Taiwan dioxin pollution episode, the poultry eggs were always play an important role. The Taiwan FDA could also give a dietary guide to the consumer for these pollution event promptly.

4.8. Strength and limitations

We have collected food samples from traditional markets or supermarkets in selected towns around Taiwan. In order to get more close to the purchasing habit of Taiwanese, the raw sample was collected in traditional markets and brand sample was collected in supermarkets. Besides, we will also select the towns with high population density. For this sampling strategy, we can achieve the objective of this study and increase the representative of food sampling for the general Taiwanese dietary habit. However, due to the limitation of resource and time, we could not easily investigate the origin place of food production. Moreover, we used a representative NAHSIT dataset on food consumption, which is more appropriate to derive dietary exposure. It uses a multistage, stratified, probability sampling design to select participants represent for Taiwan population of all ages. Face to face interview is conducted in respondents' home or at an appropriate site in each township. Dietary nutrient intakes were assessed by 24-h recall to lower the recall bias. Unfortunately, sources of contamination are difficult to assess because of the random sampling and circulation of food in the market place. Comparing intake estimations between studies from other countries is also challenging because of the differences in methodologies, the food groups considered, the population groups studied, and the manner in which the results are reported, despite the main factor affecting variability being dietary habits in the population.

5. Conclusions

The current results revealed a slightly decreasing trend in the dioxin concentration of these pollutants, demonstrating the effectiveness of the dioxin strategy implemented by the Taiwan EPA. From 2003 to 2007, 19 samples exceeded regulatory standards. After 2008, only one duck egg and one chicken egg were substandard in 2014 and 2017, respectively. This is attributable to dioxin accumulation in ducks and geese that feed in open fields where soil and water sources are more susceptible to pollution by dioxin-containing particles emitted from nearby anthropogenic activity. These results exemplify the effectiveness of contaminative source control of dioxins. However, food originating from counties and administrate districts with higher PCDD/F and DL-PCB contamination risk should be continuously monitored to ensure the safety and hygiene of food.

Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

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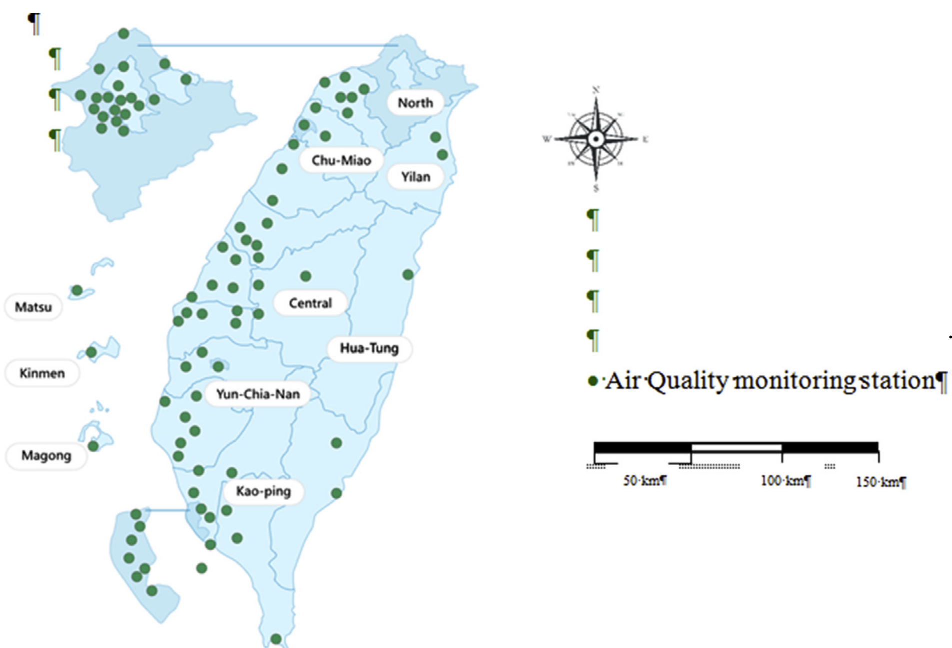
Appendix A.

Supplemental Table 1. Distribution of the different food samples collected from 2004 to 2018.

Food group (No.) area	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	total
Cereals, grains, tubers and roots (No.)	12				28	27	8	8		6	7	10	10	13	9	**138**
area	*1,2,3,4,5,6*				*3,4,5,6*	*3,4,5*	*3,4,6*	*2,3,4*		***1***	***5***	***4***	***3***	***2***	***6***	
Beans and nuts (No.)		3		12		3				2	3	3	3	4	6	**39**
area		*3,4*		*1,2,3,4,5*						***1***	***5***	***4***	***3***	***2***	***6***	
Fats and oils (No.)2	6	13								2	3	4	3	5	5	**41**
area	*1,2,3,4,5,6*									***1***	***5***	***4***	***3***	***2***	***6***	
Poultry and their products (No.)	18	26	21	20	27	23	29	21	19	2	5	15	5	11	5	**247**
area	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*3,4,5*	*3,4,5*	*1,2,3,4,5*	*3,4,5*	*1,3,4,5,6*	***1***	***5***	***4***	***3***	***2***	***6***	
Livestock and their products (No.)	18	41	48	42	25	19	28	16	24	10	10	10	10	23	14	**338**
area	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*3,4,5*	*3,4,5*	*1,3,4,5,6,7*	*3,4,5*	*1,2,3,4,5,6*	***1***	***5***	***4***	***3***	***2***	***6***	
Fish and Aquatic Products (No.)	31	45	44	48	45	51	50	30	39	26	30	26	24	35	22	**546**
area	*1,2,3,4,5*	*1,2,3,4,5,6*	*1,2,3,4,5*	*1,3,5*	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*1,2,3,4,5*	*2,3,4,5,6*	*1,3,5*	***1***	***5***	***4***	***3***	***2***	***6***	
Eggs (No.)	12	30	28	24	15	15	15	16	13	3	5	4	4	4	8	**196**
area	*1,2,3,4,5*	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*1,2,3,4,5,6*	*3,4,5*	*3,4,5*	*3,4,5*	*3,4,5*	*1,3,4,5*	***1***	***5***	***4***	***3***	***2***	***6***	
Dairy (No.)	20	49	46	19	14	15	20	25	25	6	8	10	9	9	9	**284**
area										***1***	***5***	***4***	***3***	***2***	***6***	
Fruits (No.)			15	9						7	7	7	8	9	8	**70**
area			*3,4,5*	*1,2,3,4,5*						***1***	***5***	***4***	***3***	***2***	***6***	
Vegetables (No.)	24	42	45	36	44	39	30	29		12	14	14	13	14	15	**371**
area	*1,3,4*	*1,2,3,4,5,6*	*1,2,3,4,5*	*1,2,3,4,5,6*	*1,2,3,4*	*1,2,3,4*	*2,3,4,5,6*	*1,3,4,5,6*		***1***	***5***	***4***	***3***	***2***	***6***	

Area:1:North;2:Chu-Miao;3:Central;4:Yun-Chia-Nan;5:Kao-Ping;6:Hua-Tung Dairy products, Seasonings, Composite foods and Soups, and Beverages were purchased from different brands based on the market share.



Supplemental Table 2. *The achievement of joined the Interlaboratory Comparison on Dioxins in Food held by Norwegian Institute of Public Health, Oslo, Norway in 2019.*

(1) PCDDs/PCDFs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCDDs/PCDFs	% of Z within ±0.5	% of Z within ±1
Brown meat	fresh weight	0.002	64% (56)	82%
Herring	fresh weight	0.077	66% (61)	90%
Veal	fresh weight	−2.9	8% (49)	31%

(2) Dioxin-like PCBs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCB (NON-ORTHO)	Our Laboratories' Z-scores, TEQ PCB (MONO-ORTHO)	NON-ORTHO % of Z within ±0.5	NON-ORTHO % of Z within ±1	MONO-ORTHO % of Z within ±0.5	MONO-ORTHO % of Z within ±1
Brown meat	fresh weight	0.42	0.38	60%	78%	64%	87%
Herring	fresh weight	0.51	0.43	51%	82%	52%	78%
Veal	fresh weight	0.33	0.49	38%	70%	55%	81%

(3) PCDDs/PCDFs + Dioxin-like PCBs

Sample	Unit	Our Laboratories' Z-scores, TEQ	% of Z within ±0.5	% of Z within ±1
Brown meat	fresh weight	0.18	70%	76%
Herring	fresh weight	0.31	66%	87%
Veal	fresh weight	−0.54	41%	65%

Supplemental Table 2. *The achievement of joined the Interlaboratory Comparison on Dioxins in Food held by Norwegian Institute of Public Health, Oslo, Norway in 2018 (cont'd).*

(1) PCDDs/PCDFs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCDDs/PCDFs	% of Z within ±0.5	% of Z within ±1
Reindeer	fresh weight	0.650	43%	63%
Salmon	fresh weight	0.170	50%	79%
Fish oil	fresh weight	0.047	63%	79%

(2) Dioxin-like PCBs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCB (NON-ORTHO)	Our Laboratories' Z-scores, TEQ PCB (MONO-ORTHO)	NON-ORTHO % of Z within ±0.5	NON-ORTHO % of Z within ±1	MONO-ORTHO % of Z within ±0.5	MONO-ORTHO % of Z within ±1
Reindeer	fresh weight	0.600	0.750	59%	74%	67%	87%
Salmon	fresh weight	0.067	0.340	50%	74%	56%	77%
Fish oil	fresh weight	−0.0094	0.340	68%	82%	73%	97%

Supplemental Table 2. The achievement of joined the Interlaboratory Comparison on Dioxins in Food held by Norwegian Institute of Public Health, Oslo, Norway in 2017 (cont'd).

(1) PCDDs/PCDFs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCDDs/PCDFs	% of Z within ±0.5	% of Z within ±1
Sheep meat	fresh weight	0.400	28%	47.4%
Cod liver	fresh weight	−0.240	57%	77%
Herring	fresh weight	0.260	69%	91%

(2) Dioxin-like PCBs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCB (NON-ORTHO)	Our Laboratories' Z-scores, TEQ PCB (MONO-ORTHO)	NON-ORTHO % of Z within ±0.5	NON-ORTHO % of Z within ±1	MONO-ORTHO % of Z within ±0.5	MONO-ORTHO % of Z within ±1
Sheep meat	fresh weight	0.920	0.051	63%	73%	61%	75%
Cod liver	fresh weight	0.250	0.370	67%	83%	62%	81%
Herring	fresh weight	0.520	0.330	61%	84%	64%	84%

(3) PCDDs/PCDFs + Dioxin-like PCBs

Sample	Unit	Our Laboratories' Z-scores, TEQ	% of Z within ±0.5	% of Z within ±1
Sheep meat	fresh weight	0.680	21%	36%
Cod liver	fresh weight	0.180	61%	78%
Herring	fresh weight	0.390	70%	86%

Supplemental Table 2. The achievement of joined the Interlaboratory Comparison on Dioxins in Food held by Norwegian Institute of Public Health, Oslo, Norway in 2016 (cont'd).

(1) PCDDs/PCDFs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCDDs/PCDFs	% of Z within ±0.5	% of Z within ±1
sheep liver	fresh weight	0.45	28%	47.4%
salmon	fresh weight	0.45	57%	77%
fish oil	fresh weight	−0.00074	69%	91%

(2) Dioxin-like PCBs

Sample	Unit	Our Laboratories' Z-scores, TEQ PCB (NON-ORTHO)	Our Laboratories' Z-scores, TEQ PCB (MONO-ORTHO)	NON-ORTHO % of Z within ±0.5	NON-ORTHO % of Z within ±1	MONO-ORTHO % of Z within ±0.5	MONO-ORTHO % of Z within ±1
sheep liver	fresh weight	−0.53	−1.3	22%	58%	17%	24%
salmon	fresh weight	−0.220	0.790	50%	73%	47%	76%
fish oil	fresh weight	0.41	0.024	57%	77%	72%	86%

Supplemental Table 3. The criteria of recovery of seventeen ^{13}C-labelled 2,3,7,8-substituted internal PCDD/F standards.

Congener	Acceptable Range of Recovery (%)
$^{13}C_{12}$-2,3,7,8-TCDF	35−120
$^{13}C_{12}$-1,2,3,7,8-PeCDF	35−120
$^{13}C_{12}$-2,3,4,7,8-PeCDF	35−120
$^{13}C_{12}$-1,2,3,4,7,8-HxCDF	35−120
$^{13}C_{12}$-1,2,3,6,7,8-HxCDF	35−120
$^{13}C_{12}$-2,3,4,6,7,8-HxCDF	35−120
$^{13}C_{12}$-1,2,3,7,8,9-HxCDF	35−120
$^{13}C_{12}$-1,2,3,4,6,7,8-HpCDF	35−120
$^{13}C_{12}$-1,2,3,4,7,8,9-HpCDF	35−120
$^{13}C_{12}$-2,3,7,8-TCDD	35−120
$^{13}C_{12}$-1,2,3,7,8-PeCDD	35−120
$^{13}C_{12}$-1,2,3,4,7,8-HxCDD	35−120
$^{13}C_{12}$-1,2,3,6,7,8-HxCDD	35−120
$^{13}C_{12}$-1,2,3,4,6,7,8-HpCDD	35−120
$^{13}C_{12}$-OCDD	35−120

Supplemental Table 3. The criteria of recovery of seventeen ^{13}C-labelled internal Dioxin-like PCBs standards (cont'd).

Congener	Acceptable Range of Recovery (%)
$^{13}C_{12}$-3,3′,4,4′-TeCB	26−143
$^{13}C_{12}$-3,4,4′,5-TeCB	26−143
$^{13}C_{12}$-2,3,3′,4,4′-PeCB	26−143
$^{13}C_{12}$-2,3,4,4′,5-PeCB	26−143
$^{13}C_{12}$-2,3′,4,4′,5-PeCB	26−143
$^{13}C_{12}$-2′,3,4,4′,5-PeCB	26−143
$^{13}C_{12}$-3,3′,4,4′,5-PeCB	26−143
$^{13}C_{12}$-2,3,3′,4,4′,5-HxCB	26−143
$^{13}C_{12}$-2,3,3′,4,4′,5′-HxCB	26−143
$^{13}C_{12}$-2,3′,4,4′,5,5′-HxCB	26−143
$^{13}C_{12}$-3,3′,4,4′,5,5′-HxCB	26−143
$^{13}C_{12}$-2,3,3′,4,4′,5,5′-HpCB	26−143

Supplemental Table 4. Matrix-specific Limit of Quantitation (LOQ) of PCDD/Fs in food.

Food group	meat	milk	egg	oil	fish	Vegetables, fruits and plants	feed	soil	air	blood
unit	pg/g fat	pg/g fat	pg/g fat	pg/g fat	pg/g w.w.	pg/g d.w.	pg/g w.w.	pg/g d.w.	pg/Nm^3	pg/g w.w.
2,3,7,8-TCDF	0.021	0.016	0.019	0.016	0.002	0.01	0.003	0.008	0.0004	0.003
1,2,3,7,8-PeCDF	0.01	0.011	0.012	0.01	0.001	0.005	0.001	0.004	0.0012	0.002
2,3,4,7,8-PeCDF	0.008	0.009	0.009	0.008	0.001	0.004	0.001	0.004	0.0012	0.002
1,2,3,4,7,8-HxCDF	0.007	0.008	0.007	0.006	0.001	0.003	0.001	0.005	0.0006	0.002
1,2,3,6,7,8-HxCDF	0.007	0.008	0.007	0.007	0.001	0.003	0.001	0.004	0.0005	0.002
2,3,4,6,7,8-HxCDF	0.007	0.008	0.008	0.007	0.001	0.003	0.001	0.005	0.0006	0.002
1,2,3,7,8,9-HxCDF	0.01	0.011	0.01	0.01	0.001	0.004	0.002	0.008	0.0008	0.003
1,2,3,4,6,7,8-HpCDF	0.007	0.009	0.008	0.009	0.001	0.004	0.002	0.004	0.0008	0.002
1,2,3,4,7,8,9-HpCDF	0.011	0.013	0.012	0.015	0.002	0.008	0.002	0.01	0.0011	0.004
OCDF	0.021	0.028	0.021	0.026	0.003	0.017	0.004	0.016	0.0004	0.008
2,3,7,8-TCDD	0.013	0.014	0.011	0.012	0.001	0.007	0.003	0.015	0.0006	0.004
1,2,3,7,8-PeCDD	0.011	0.01	0.01	0.011	0.001	0.005	0.002	0.004	0.0004	0.003
1,2,3,4,7,8-HxCDD	0.009	0.011	0.009	0.01	0.001	0.005	0.002	0.006	0.0006	0.003
1,2,3,6,7,8-HxCDD	0.01	0.011	0.009	0.01	0.001	0.005	0.002	0.007	0.0005	0.003
1,2,3,7,8,9-HxCDD	0.01	0.012	0.009	0.01	0.001	0.006	0.002	0.006	0.0005	0.003
1,2,3,4,6,7,8-HpCDD	0.012	0.013	0.01	0.014	0.001	0.014	0.002	0.014	0.0003	0.005
OCDD	0.023	0.024	0.021	0.029	0.002	0.015	0.006	0.037	0.0004	0.014
total	0.197	0.216	0.193	0.207	0.021	0.12				
Ref. page	3−4	10	20	16−17	21−22	24−25	27−28	30−31	31	32

Supplemental Table 4. Matrix-specific Limit of Quantitation (LOQ) of dioxin-like PCBs in food (cont'd).

Food group	meat	milk	eggs	oil	fish	Vegetables, fruits & plants	feed	blood
unit	pg/g fat	pg/g fat	pg/g fat	pg/g fat	pg/g w.w.	pg/g d.w.	pg/g w.w.	pg/g w.w.
3,4,4′,5-TeCB 81	0.023	0.024	0.041	0.037	0.01	0.011	0.007	0.008
3,3′,4,4′-TeCB 77	0.025	0.025	0.043	0.039	0.007	0.012	0.007	0.008
2′,3,4,4′,5-PeCB 123	0.04	0.037	0.054	0.049	0.011	0.015	0.009	0.013
2,3′,4,4′,5-PeCB 118	0.035	0.033	0.049	0.042	0.01	0.013	0.006	0.012
2,3,4,4′,5-PeCB 114	0.031	0.03	0.044	0.038	0.009	0.011	0.006	0.013
2,3,3′,4,4′-PeCB 105	0.03	0.03	0.048	0.038	0.009	0.011	0.007	0.012
3,3′,4,4′,5-PeCB 126	0.029	0.033	0.053	0.033	0.009	0.011	0.007	0.014
2,3′,4,4′,5,5′-HxCB 167	0.039	0.038	0.065	0.049	0.0013	0.015	0.008	0.013
2,3,3′,4,4′,5-HxCB 156	0.019	0.018	0.034	0.022	0.007	0.006	0.005	0.008
2,3,3′,4,4′,5′-HxCB 157	0.017	0.016	0.032	0.021	0.006	0.006	0.006	0.008
3,3′,4,4′,5,5′-HxCB 169	0.014	0.012	0.037	0.017	0.005	0.004	0.004	0.007
2,3,3′,4,4′,5,5′-HpCB 189	0.026	0.019	0.064	0.059	0.102	0,020	0014	0.007
total	0.327	0.314	0.564	0.445	0.102	0.135		
Ref. page	4	9	12	17–18	21	25	28–29	33

Supplemental Table 5. Comparison of reference values for PCDD/Fs in CRM 1954 Whole milk powder in 2019.

	2015 WHO TEF	CRM conc.		Our Lab				Z-score
		value	1 Std Dev	Test Sample	conc. absolute difference	conc. difference in Std Dev	Relative percent difference (RPD)	
Sample weight (g)				5.0468				
Lipid (%)				3.73%				
Congeners		(pg/g sample)		(pg/g sample)	(pg/g sample)		(%)	
2,3,7,8-TCDF	0.100	0.125	0.010	0.143	0.018	1.77	14%	1.77
1,2,3,7,8-PeCDF	0.030	0.132	0.018	0.125	−0.007	−0.40	−5%	−0.40
2,3,4,7,8-PeCDF	0.3	0.347	0.025	0.327	−0.020	−0.80	−6%	−0.80
1,2,3,4,7,8-HxCDF	0.1	0.171	0.015	0.188	0.017	1.15	10%	1.15
1,2,3,6,7,8-HxCDF	0.1	0.186	0.017	0.194	0.008	0.48	4%	0.48
1,2,3,4,6,7,8-HpCDF	0.01	0.407	0.045	0.353	−0.054	−1.21	−13%	−1.21
1,2,3,4,7,8,9-HpCDF	0.01	0.160	0.100	0.139	−0.021	−0.21	−13%	−0.21
OCDF	0.0003	0.094	0.013	0.149	0.054	4.28	58%	4.28
2,3,7,8-TCDD	1	0.162	0.020	0.226	0.064	3.19	39%	3.19
1,2,3,7,8-PeCDD	1	0.240	0.017	0.254	0.014	0.80	6%	0.80
1,2,3,4,7,8-HxCDD	0.1	0.182	0.016	0.244	0.062	3.86	34%	3.86
1,2,3,6,7,8-HxCDD	0.1	0.890	0.140	0.765	−0.125	−0.89	−14%	−0.89
1,2,3,7,8,9-HxCDD	0.1	0.207	0.020	0.212	0.005	0.25	2%	0.25
1,2,3,4,6,7,8-HpCDD	0.01	1.080	0.240	1.175	0.095	0.40	9%	0.40
OCDD	0.0003	4.890	0.850	5.148	0.258	0.30	5%	0.30
SUM TEQ		0.826	0.082	0.886	0.059	0.72	7%	0.72

*absolute difference = Test sample-CRM Certified value; *Relative percent difference=(Test Sample-CRM Certified value)/(CRM Certified value).

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Supplemental Table 5. Comparison of reference values for DL PCB in CRM 1954 Whole milk powder (cont'd).

	2015 WHO TEF	CRM conc.		Our Lab				Z-score
		value	1 Std Dev	Test Sample	conc. **absolute difference**	conc. difference **in Std Dev**	Relative percent difference (RPD)	
Sample weight (g)				5.0468				
Lipid (%)				3.73%				
Congeners		(pg/g sample)		(pg/g sample)	(pg/g sample)		(%)	
3,4,4′,5-TeCB 81	0.0003	0.63	0.028	0.541	−0.089	−3.18	−14%	−3.18
3,3′,4,4′-TeCB 77	0.0001	2.71	0.14	3.458	0.748	5.34	28%	5.34
2′,3,4,4′,5-PeCB 123	0.0000	67.9	3.1	68.136	0.236	0.08	0%	0.08
2,3,4,4′,5-PeCB 114	0.0000	90.5	7.4	61.191	−29.309	−3.96	−32%	−3.96
3,3′,4,4′,5-PeCB 126	0.1000	10.4	1.5	10.248	−0.152	−0.10	−1%	−0.10
3,3′,4,4′,5,5′-HxCB 169	0.0300	9.3	1.2	10.450	1.150	0.96	12%	0.96
SUM TEQ		1.324	0.186	1.343	0.018	0.10	1%	0.10

*absolute difference = Test sample-CRM Certified value.
*Relative percent difference=(Test Sample-CRM Certified value)/(CRM Certified value).

Supplemental Table 6. Difference of PCDD/Fs and DL-PCBs in Taiwan food presented in upper and lower bond.

Food group	N	upper bond	lower bond	difference	RPD (%)
Cereals, grains, tubers and roots					
Rice and its products	65	0.0177	0.0128	0.0049	0.606
Wheat and its products	52	0.0169	0.0108	0.0061	1.088
Carbohydrate's tubers, roots, and their products	21	0.0103	0.0073	0.0030	0.596
Beans and nuts					
Beans	15	0.0337	0.0282	0.0055	1.790
Bean processed products	20	0.0054	0.0039	0.0014	0.883
Nuts and its products	4	0.0200	0.0107	0.0093	0.998
Fats and oils					
Vegetable oils	28	0.0836	0.0752	0.0083	0.206
Animal fats	8	0.1639	0.1636	0.0003	0.002
Others	5	0.0198	0.0173	0.0025	0.170
Poultry and their products					
Chicken and its products	96	0.0314	0.0312	0.0002	0.013
Duck and its products	88	0.0880	0.0879	0.0001	0.003
Goose and its products	63	0.0842	0.0842	0.0001	0.001
Livestock and their products					
Pork and its products	163	0.0315	0.0305	0.0011	0.132
Beef and its products	94	0.1038	0.1036	0.0002	0.020
Mutton and its products	81	0.1793	0.1791	0.0002	0.015
Fish and Aquatic Products					
Freshwater fish	70	0.2463	0.2461	0.0002	0.003
Marine fish	266	0.4774	0.4771	0.0003	0.008
Fish and its products	89	0.2203	0.2200	0.0003	0.011
Other aquatic animals and their products	121	0.1922	0.1917	0.0005	0.026
Food group	**N**	**upper bond**	**lower bond**	**difference**	**RPD (%)**
Eggs					
Chicken eggs	89	0.0524	0.0519	0.0005	0.020
Duck eggs	63	0.2112	0.2111	0.0002	0.002
Other eggs	44	0.1480	0.1477	0.0003	0.008
Dairy					
Whole fat milk	204	0.0373	0.0373	0.00004	0.003
Low fat/fat free milk	6	0.0141	0.0140	0.0001	0.008
Whole fat sheep milk	24	0.0336	0.0336	<0.0001	0.001
Fermented milk	14	0.0193	0.0192	0.0001	0.013
Other milk	10	0.0352	0.0346	0.0006	0.139
Powdered milk	13	0.0468	0.0460	0.0008	0.094
Cheese	13	0.1941	0.1940	0.0001	0.001
Fruits					
Berries	32	0.0059	0.0042	0.0017	0.702

(*continued on next page*)

. *(continued)*

Food group	N	upper bond	lower bond	difference	RPD (%)
Pomaceous fruits	9	0.0039	0.0030	0.0010	0.575
Stone fruits	9	0.0063	0.0057	0.0006	0.633
Melon and fruit	6	0.0025	0.0016	0.0009	0.927
Citrus Fruit	9	0.0049	0.0035	0.0013	0.967
Sugar-cane	5	0.0056	0.0034	0.0022	0.761
Vegetables					
Leafy vegetables	203	0.0143	0.0140	0.0003	0.128
Fruit crops	12	0.0063	0.0060	0.0003	0.120
Bean sprouts	16	0.0070	0.0065	0.0005	0.352
Gourd	25	0.0024	0.0020	0.0004	2.771
Stem vegetables	76	0.0056	0.0050	0.0007	0.413
Mushrooms	32	0.0088	0.0081	0.0007	0.440
Others	7	0.0083	0.0045	0.0038	5.072
Food group	N	upper bond	lower bond	difference	RPD (%)
Seasonings					
Salt	5	0.0150	0.0120	0.0030	0.380
MSG	1	0.0057	0.0049	0.0008	0.168
Soy sauce	18	0.0283	0.0265	0.0018	0.267
Curry sauce	17	0.0279	0.0249	0.0029	0.270
Composite foods and Soups					
Rice	22	0.0091	0.0074	0.0017	0.541
Wheat	90	0.0251	0.0235	0.0016	0.135
Others	2	0.0662	0.0637	0.0025	0.317
Candies and Snacks	11	0.0176	0.0150	0.0027	0.351
Beverages	5	0.0046	0.0041	0.0006	0.154

Unit: pg WHO_{05}-$TEQ_{PCDD/F+PCB}$ g^{-1} wet weight.
Note: PCDDs, polychlorinated dibenzo-*p*-dioxins; PCDFs, polychlorinated dibenzofurans; DL-PCBs, dioxin-like polychlorinated biphenyls.

Supplemental Table 7. *Overview of dietary intake of PCDD/Fs and DL-PCBs (pg total TEQ kg^{-1} bw day^{-1}) obtained from other studies.*

Country	Sampling year	Survey method	WHO-TEF	Adults	Young People	Scenario	Reference
China	2008	TDS	1998	1.36	n.a.	MB	Zhang et al., 2008
France	2012	TDS	1998	0.57	0.89	MB	Sirot et al., 2012
Belgium	2010	24 h/FFQ	2005	0.61	n.a.	MB	Windal et al., 2010
Europe	2012	Monitoring	2005	0.57–1.67	1.08–2.54	ND	EFSA, 2012
Finland	2003	Market basket	1998	1.5	n.a.	ND	Kirivanta et al., 2004
Japan	2008	3 Day Dietary Record	1998	1.06	n.a.	LB	Arisawa et al., 2008
Australia	2011	TDS	1998	0.12–0.52	n.a.	LB-UB	FSANZ, 2011
Spain	2011	24 h	Calux	1.13–1.58	2.04–2.76	LB-UB	Quijano et al., 2017
United Kingdom	2012	TDS	2005	0.52	n.a.	UB	Bramwell et al., 2016
Ireland	2003–10	4 Day Dietary Record	2005	0.3	n.a.	UB	Tlustos et al., 2014
Italy	2013–2016	3 Day Dietary Record	2005	0.9	1.16–1.98	UB	Diletti et al., 2018
Taiwan	2013–2018	24 h	2005	0.172–0.360/0.186–0.386	0.052–0.561/0.057–0.624	LB-UB	This study
Taiwan	2013–2018	24 h	1998	0.190–0.403/0.204–0.429	0.058–0.629 /0.062–0.689	LB-UB	This study

n.a.: no data available in the study.
LB: Lower bound; MB: Medium bound; UB: Upper bound.
European countries included in EFSA, 2012: Iceland, Norway, Hungary, Latvia, Slovakia, Italy, Spain, Cyprus, Belgium, Ireland, Lithuania, Luxembourg, Romania, Bulgaria, Malta, Portugal, Germany, United Kingdom, Denmark, Italy, Norway, Estonia, Austria.
Notes: Total TEQ = sum WHO TEQ PCDD/F + DL-PCB; UB, upper bound (<LOQ = LOQ); MB, medium bound (<LOQ = 0.5LOQ); LB, lower bound (<LOQ = 0); n.a., not available.

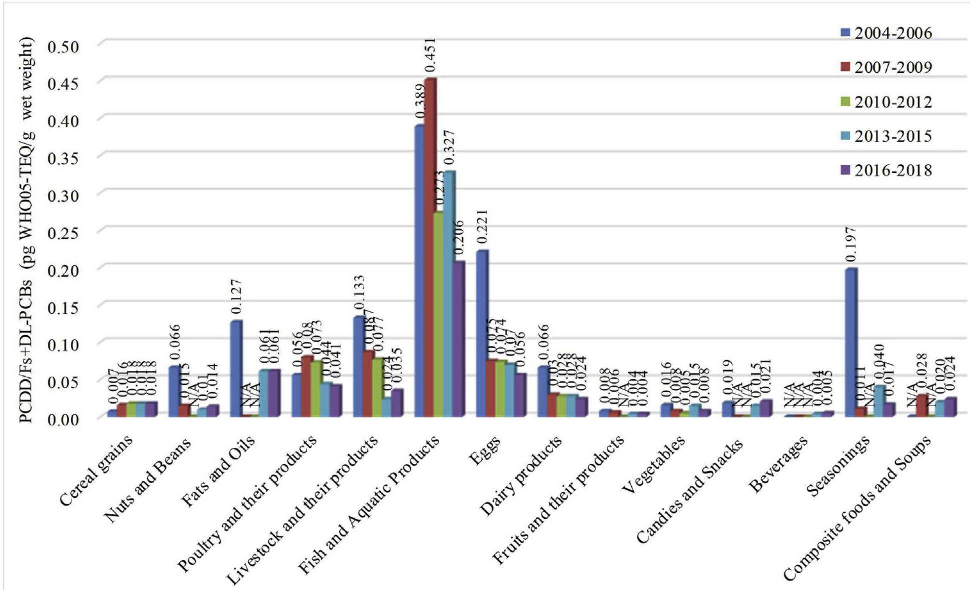

Figure S1. Time trends of the total PCDD/Fs and DL-PCBs levels, expressed as pg WHO_{05}-TEQ g^{-1} w.w. in different food groups. PCDD/F: polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans, DL-PCBs: dioxin-like polychlorinated biphenyls, AQRs air quality regions, WHO: World Health Organization, TEQ: toxic equivalent..

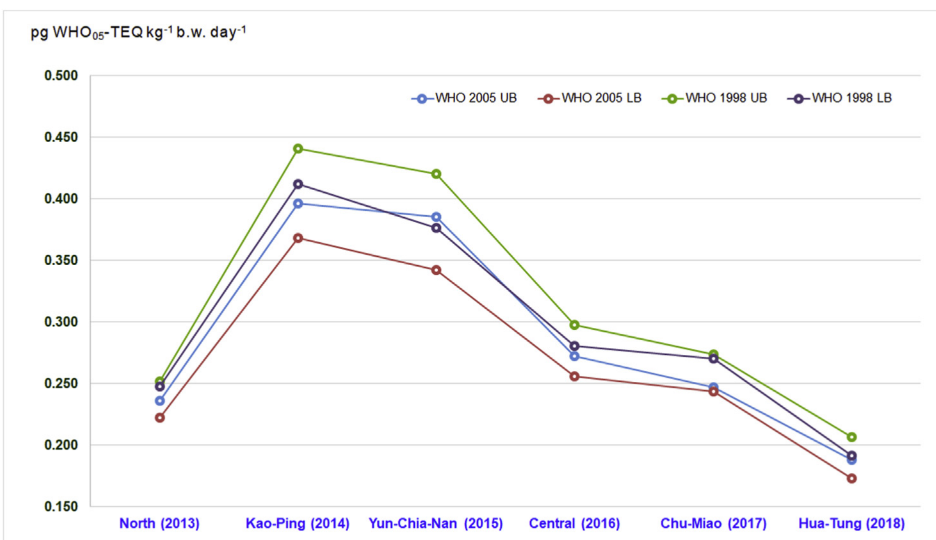

Figure S2. Distribution of total daily intake of PCDD/Fs and DL-PCBs by Taiwanese in 6 AQRs..

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