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# Temporal trend of polychlorinated dibenzo-p-dioxin/polychlorinated dibenzofuran and dioxin like-polychlorinated biphenyl concentrations in food from Taiwan markets during 2004–2012

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ABSTRACT

The levels of polychlorinated dibenzo-p-dioxin (PCDD) and polychlorinated dibenzofuran (PCDF) or polychlorinated biphenyl (PCB) in foodstuffs have decreased over the past decade in many countries. However, the trend for the levels of these compounds in foodstuffs in Taiwan remains unknown. In this study, we compared the distribution of PCDD/F and PCB in nine foodstuff categories acquired from Taiwan markets from 2004 to 2012. The levels expressed as World Health Organization toxic equivalents (WHO-TEQs) in the different foodstuffs tested were as follows: fish, average 0.463 pg $WHO_{98}$-TEQ/g sample > seafood, 0.163 pg $WHO_{98}$-TEQ/g > eggs, 0.150 pg $WHO_{98}$-TEQ/g > oils, 0.126 pg $WHO_{98}$-TEQ/g > meats, 0.095 pg $WHO_{98}$-TEQ/g > dairy products, 0.054 pg $WHO_{98}$-TEQ/g > cereals, 0.017 pg $WHO_{98}$-TEQ/g > vegetables, 0.013 pg $WHO_{98}$-TEQ/g > fruits, 0.009 pg $WHO_{98}$-TEQ/g. Levels were particularly high in crab (average: 0.6 pg $WHO_{98}$-TEQ/g sample (1.243 pg $WHO_{98}$-TEQ/g sample) and large marine fish (0.6). In Taiwan, a decreasing trend of PCDD/Fs or dioxin-like PCBs (dl-PCBs) was observed in meat, dairy, eggs, and vegetables, whereas an elevated trend was observed in cereals or the levels were nearly equal in fruits and oils at alternative time shift. Dl-PCBs contributed to 60–65% toxicity equivalence levels in fish and seafood, but only to 13–40% in meat and cereal samples. The decreasing trend was consistent with the results in other countries; however, the trends in cereals, fruits, and oils were in contrast to previous results reported in other countries. Cereals and fruits are important crops in southern Taiwan, and the local pollution generated by industries or incinerators

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may seriously affect the distribution of PCDD/Fs and dl-PCBs. To ensure food safety, a risk assessment for residents living in different areas should be adopted for all food categories simultaneously in the future.



## 1. Introduction

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) consist of 75 PCDDs and 135 polychlorinated PCDFs, of which 17 exhibit toxicity. Polychlorinated biphenyls (PCBs) are a group of 209 different chemicals, of which 12 are similar to PCDD/Fs in toxicological characteristics and are referred to as dioxin-like PCBs (dl-PCBs). Therefore, all analytical results for individual PCDD/Fs and dl-PCB congeners of toxicological concern are expressed in terms of the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin toxicity equivalence (TEQ).

Many previous reports have highlighted the importance of dietary exposure to PCDD/Fs and dl-PCB [1,2] in many countries. In the United States, ~50% of the daily PCDD/F intake is attributable to meat and dairy products [3]. In Japan, > 50% of the PCDD/F intake is attributable to fish and shellfish [4], and Mato et al [5] reported that fish and shellfish intake accounted for 45–70% of the total dietary intake of PCDD/Fs and dl-PCB in each age group obtained in their study.

In fish from the southern Baltic Sea, the contribution of dl-PCBs to the total TEQ ranged from 50% to 70% [6], which is lower than the contribution in eggs (average: 71%, range 55–92%) in southwest Germany [7]. In the Adriatic Sea region, dl-PCBs account for > 77% of this intake, followed by PCDDs (15.5%) and PCDFs (13.1%) [8]. In Taiwan, dl-PCBs contributed to 31%, 59%, 36%, 46%, and 13% of the total TEQ in meat and meat products, muscle meat of fish, milk and dairy products, fat and oil, and eggs collected from 14 food groups of animal origin from 11 locations, respectively [9]. The contribution of PCBs to the total TEQs in Taiwan is not as high as the values reported in other countries.

In Greece, the results for the analyses of PCDD/Fs and coplanar PCB levels in 77 milk and dairy products, meat and meat products, fish, vegetable oil, eggs, fruit, vegetables, and rice collected from markets between August 2002 and December 2002 were far below the European Council Regulation (2375/2001/European Council) limits [10]. However, in 2001, the Joint Food and Agriculture Organization/World Health Organization (WHO) Expert Committee on Food Additives defined a provisional tolerable monthly intake (PTMI) of 70 pg TEQ/kg body weight. The sum of the median intake of PCDD/Fs and PCBs exceeded the PTMI in western European countries. Therefore, the European Union (EU) Commission developed a strategy for reducing the accumulation of PCDD/Fs and PCBs in the environment and in the food chain [11]. In Europe, the risk management of PCDD/Fs and PCB measurements were applied after 2001 [12]. In 2012, comprehensive monitoring of PCDD/Fs and PCBs in dietary components in 26 European countries was defined by the European Food Safety Authority. The data revealed a general decreasing trend in dietary exposure to PCDD/Fs and dl-PCBs from 2002 to 2004 and from 2008 to 2010 [11]. Additionally, decreasing trends were also observed in the United States [13], Canada [14], and Sweden [15], and in hens' eggs in Germany [7]; similar trends were also observed for PCDD/F concentrations in marine mussels in China [16]. The daily dose of PCDD/Fs or PCBs in other countries such as France [17], The Netherlands [18], and Spain [19] as well as the EU [20] all showed decreasing trends.

Many countries have shown decreasing trends of PCDD/Fs or PCBs in foodstuffs; however, these trends remain unclear in Taiwan. In the present study, we compared the concentrations of PCDD/Fs and PCBs in nine foodstuff categories in Taiwan from the samples obtained from 2004 to 2012. The ratio of PCDD/Fs versus PCBs in alternative foodstuffs can also be used to track contaminant sources in different environmental media.

## 2. Methods

### 2.1. *Food sampling*

Sampling was conducted using a systematic process in a market-basket survey. Firstly, 20 sampling counties in Taiwan and four areas were grouped according to their locations: north, central area, south, and east. Secondly, the sampling quality of each foodstuff was defined in every county of each area. Thirdly, all food samples were purchased from traditional markets or supermarkets in each county around Taiwan from 2004 to 2012. Finally, 1715 foodstuffs were sampled from 2004 to 2012 for 9 years and prepared for PCDD/F and PCB analysis.

All group samples were adequately homogenized and then frozen at −20°C until analysis. For example, 600 g of the pork composite sample was prepared by homogenizing 10 aliquots of 60-g homogenized pork, which was prepared by separating pork samples of 500–1000 g. The investigated samples of fishes: freshwater fish, large and small marine fish, fishery fish (sample size = 304); seafood: crab, mollusca, shellfish, shrimp, and other seafood products (86); meats: beef, pork, mutton, chicken, duck, goose, and other poultry products (485); dairy: milk, milk power, yogurt, butter, cheese, cream (236), eggs (167), cereals (89), fruits (27); and vegetables: beans, mushroom, leafy, and root vegetables (301), and oils (16) were prepared as described above.

### 2.2. *High-resolution gas chromatography/high-resolution mass spectrometry analysis of 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 fishes, seafood, meats, eggs, milk, dairy products, and oil samples, as described previously [21,22]. Analytical procedures were adopted from the United States Environmental Protection Agency (USEPA) Method 1613B [23] and USEPA Method 1668A [24] with minor modifications. Three different extraction procedures (I, II, and III) were applied for various sample matrices. Quality assurance/quality control protocols were established, according to those defined in the USEPA Method 1668A [24], in the laboratory to ensure positive identification and measurement quality. The quality assurance/quality control protocols included mass spectrometry resolution, gas chromatography resolution, calibration verification, ongoing precision and recovery, blank, and internal standard recovery. The analytical laboratory responsible for this analysis, Trace Environmental Pollutant, Research Center of Environmental Trace Toxic Substances, at the National Cheng Kung University in Tainan, Taiwan, was certificated by the Taiwan Accreditation Foundation. The PCDD/Fs/dl-PCBs concentrations were stated as a fat-weight and wet-weight basis (pg WHO-TEQ/g fat, and pg WHO-TEQ/g, wet weight).

### 2.3. *Statistical analysis*

The Excel package and JMP (version 6.1, SAS Institute Inc.) were utilized for data management and statistical analysis. The analyses were conducted to compare total PCDD/F intake for to identify any time trends or geographical differences. For time-trend analysis, PCDD/F and dl-PCB levels were analyzed for 9 years in alternative foodstuffs. To analyze geographical variations, PCDD/Fs and dl-PCBs were analyzed for all four sampling areas in Taiwan.

## 3. Results and discussion

### 3.1. *PCDD/Fs and PCBs concentration in foodstuffs of different countries*

Tables 1 and 2 show the sample size for each location and the PCDD/F plus dl-PCB levels in alternative food categories. Because the sampling food was packed food the sampling location did not affect the analysis. Therefore, some samples were just taken from one location, e.g., packed oils were just taken from the south location. The highest levels were found in fish (average, 0.462 pg $WHO_{98}$-TEQ/g sample) and the levels in the other foods were in the following order: seafood (0.264 pg $WHO_{98}$-TEQ/g sample) > eggs (0.150 pg $WHO_{98}$-TEQ/g sample) > oils (0.126 pg $WHO_{98}$-TEQ/g sample) > meats (0.094 pg $WHO_{98}$-TEQ/g sample) > dairy products (0.055 pg $WHO_{98}$-TEQ/g sample) > cereals (0.017 pg $WHO_{98}$-TEQ/g sample) > vegetables (0.012 pg $WHO_{98}$-TEQ/g sample) > fruits (0.009 pg $WHO_{98}$-TEQ/g sample). Crab (average, 1.243 pg $WHO_{98}$-TEQ/g sample) and large marine fish (0.6 pg $WHO_{98}$-TEQ/g sample) showed very high levels. The TEQ based on the WHO definition of 2005 showed the similar values as TEQ defined in 1998. The ratio of dl-PCB to PCDD/Fs is shown in Figure 1. In fish and seafood, dl-PCBs contributed to 60–65% of the TEQ levels, but only 13–40% of dl-PCBs in meat and cereal samples.

In Taiwan, the estimated monthly intake of PCDD/Fs and dl-PCBs was 44.7 pg WHO-TEQ/kg body weight/month and 39.5 pg WHO-TEQ/kg body weight/mo for male and female adults [9], which was calculated based on the measurements of 14 food groups of animal origin from 11 locations in Taiwan of a total diet study samples. The dl-PCB contributions were 31%, 59%, 36%, 46%, and 13% for meat and meat products, fish,

**Table 1 – The sampling distribution from each location around Taiwan.**

| Food categories | Food items | N | North | South | Center | East |
|---|---|---|---|---|---|---|
| Fish | Fishery products | 82 | 0 | 57 | 25 | 0 |
| | Freshwater fish | 51 | 6 | 25 | 17 | 3 |
| | Marine fish | 171 | 20 | 96 | 30 | 25 |
| Seafood | Crab | 12 | 0 | 4 | 0 | 8 |
| | Seafood products | 28 | 0 | 12 | 16 | 0 |
| | Seafood | 29 | 0 | 12 | 8 | 9 |
| | Shrimp | 21 | 1 | 7 | 12 | 1 |
| Meat | Poultry | 210 | 29 | 104 | 77 | 0 |
| | Livestock | 275 | 52 | 148 | 75 | 0 |
| Dairy | Dairy products | 31 | 0 | 31 | 0 | 0 |
| | Milk | 205 | 34 | 78 | 92 | 1 |
| Eggs | Eggs | 167 | 0 | 72 | 55 | 40 |
| Cereals | Cereals | 89 | 19 | 48 | 22 | 0 |
| Oil | Animal fat | 2 | 0 | 2 | 0 | 0 |
| | Vegetable oil | 14 | 0 | 14 | 0 | 0 |
| Fruits | Fruits | 27 | 0 | 0 | 12 | 15 |
| Vegetables | Bamboo shoot | 3 | 0 | 0 | 0 | 3 |
| | Beans | 21 | 0 | 21 | 0 | 0 |
| | Leafy vegetables | 152 | 39 | 67 | 46 | 0 |
| | Melons | 18 | 7 | 0 | 8 | 3 |
| | Root vegetable | 83 | 0 | 39 | 38 | 6 |
| | Mushroom | 24 | 0 | 24 | 0 | 0 |

**Table 2 – Distribution of polychlorinated dibenzo-p-dioxin/polychlorinated dibenzofuran (PCDD/Fs) and dioxin-like polychlorinated biphenyl (dl-PCBs) in Taiwan food from 2004–2010.**

| Food group | N | TEQ $WHO_{1998}$/g samples | | | TEQ $WHO_{2005}$/g samples | | |
|---|---|---|---|---|---|---|---|
| | | PCDD/Fs | PCBs | PCDD/Fs + dL-PCB | PCDD/Fs | PCBs | PCDD/Fs + dL-PCB |
| Fish | 304 | 0.131 ± 0.3 | 0.332 ± 0.806 | 0.462 ± 1.08 | 0.11 ± 0.249 | 0.299 ± 0.679 | 0.409 ± 0.905 |
| Freshwater fish | 51 | 0.155 ± 0.206 | 0.219 ± 0.367 | 0.374 ± 0.55 | 0.131 ± 0.171 | 0.195 ± 0.313 | 0.325 ± 0.468 |
| Marine fish | 171 | 0.146 ± 0.375 | 0.43 ± 1.027 | 0.576 ± 1.373 | 0.122 ± 0.312 | 0.383 ± 0.858 | 0.505 ± 1.144 |
| Small marine fish | 139 | 0.151 ± 0.385 | 0.42 ± 1.039 | 0.57 ± 1.408 | 0.128 ± 0.324 | 0.37 ± 0.853 | 0.498 ± 1.162 |
| Large marine fish | 32 | 0.125 ± 0.328 | 0.475 ± 0.983 | 0.6 ± 1.225 | 0.098 ± 0.254 | 0.439 ± 0.892 | 0.538 ± 1.082 |
| Fishery products | 82 | 0.084 ± 0.116 | 0.197 ± 0.301 | 0.281 ± 0.399 | 0.072 ± 0.093 | 0.19 ± 0.293 | 0.262 ± 0.371 |
| Seafood | 90 | 0.102 ± 0.198 | 0.162 ± 0.555 | 0.264 ± 0.699 | 0.09 ± 0.176 | 0.138 ± 0.429 | 0.228 ± 0.561 |
| Crab | 12 | 0.444 ± 0.361 | 0.799 ± 1.38 | 1.243 ± 1.6 | 0.396 ± 0.321 | 0.657 ± 1.044 | 1.052 ± 1.239 |
| Mollusca | 6 | 0.025 ± 0.007 | 0.042 ± 0.022 | 0.067 ± 0.024 | 0.022 ± 0.005 | 0.037 ± 0.018 | 0.059 ± 0.02 |
| Shellfish | 22 | 0.053 ± 0.043 | 0.098 ± 0.124 | 0.151 ± 0.16 | 0.046 ± 0.037 | 0.089 ± 0.115 | 0.135 ± 0.146 |
| Shrimp | 21 | 0.057 ± 0.06 | 0.072 ± 0.095 | 0.129 ± 0.129 | 0.049 ± 0.05 | 0.065 ± 0.086 | 0.114 ± 0.112 |
| Seafood products | 29 | 0.046 ± 0.109 | 0.037 ± 0.097 | 0.083 ± 0.205 | 0.041 ± 0.095 | 0.035 ± 0.088 | 0.076 ± 0.182 |
| Meat | 485 | 0.06 ± 0.126 | 0.034 ± 0.079 | 0.094 ± 0.196 | 0.052 ± 0.109 | 0.033 ± 0.077 | 0.085 ± 0.179 |
| Livestock | 275 | 0.067 ± 0.157 | 0.04 ± 0.088 | 0.108 ± 0.242 | 0.059 ± 0.136 | 0.039 ± 0.09 | 0.098 ± 0.223 |
| Beef | 76 | 0.081 ± 0.104 | 0.049 ± 0.072 | 0.13 ± 0.172 | 0.069 ± 0.087 | 0.048 ± 0.071 | 0.117 ± 0.154 |
| Mutton | 65 | 0.147 ± 0.286 | 0.086 ± 0.151 | 0.233 ± 0.432 | 0.129 ± 0.248 | 0.085 ± 0.156 | 0.214 ± 0.399 |
| Livestock products | 78 | 0.017 ± 0.009 | 0.009 ± 0.007 | 0.026 ± 0.013 | 0.016 ± 0.008 | 0.009 ± 0.007 | 0.024 ± 0.013 |
| Pork | 56 | 0.026 ± 0.017 | 0.02 ± 0.025 | 0.046 ± 0.039 | 0.023 ± 0.015 | 0.017 ± 0.021 | 0.039 ± 0.033 |
| Poultry | 210 | 0.05 ± 0.063 | 0.027 ± 0.064 | 0.077 ± 0.109 | 0.044 ± 0.055 | 0.024 ± 0.055 | 0.068 ± 0.096 |
| Chicken | 59 | 0.028 ± 0.039 | 0.01 ± 0.007 | 0.038 ± 0.04 | 0.025 ± 0.033 | 0.009 ± 0.006 | 0.034 ± 0.035 |
| Duck | 60 | 0.04 ± 0.03 | 0.031 ± 0.103 | 0.072 ± 0.118 | 0.035 ± 0.026 | 0.027 ± 0.085 | 0.062 ± 0.098 |
| Goose | 58 | 0.065 ± 0.051 | 0.031 ± 0.024 | 0.097 ± 0.07 | 0.057 ± 0.045 | 0.029 ± 0.022 | 0.086 ± 0.062 |
| Poultry products | 33 | 0.082 ± 0.12 | 0.039 ± 0.074 | 0.121 ± 0.185 | 0.072 ± 0.106 | 0.037 ± 0.071 | 0.109 ± 0.168 |
| Dairy | 236 | 0.035 ± 0.043 | 0.019 ± 0.025 | 0.055 ± 0.066 | 0.03 ± 0.038 | 0.018 ± 0.023 | 0.049 ± 0.059 |
| Milk | 205 | 0.029 ± 0.017 | 0.015 ± 0.009 | 0.045 ± 0.025 | 0.025 ± 0.015 | 0.015 ± 0.009 | 0.04 ± 0.023 |
| Whole fat milk | 181 | 0.029 ± 0.018 | 0.015 ± 0.009 | 0.045 ± 0.026 | 0.025 ± 0.015 | 0.015 ± 0.009 | 0.04 ± 0.023 |
| Powdered milk | 5 | 0.047 ± 0.01 | 0.019 ± 0.011 | 0.065 ± 0.018 | 0.044 ± 0.01 | 0.018 ± 0.011 | 0.062 ± 0.017 |
| Whole fat sheep milk | 19 | 0.024 ± 0.007 | 0.015 ± 0.003 | 0.04 ± 0.009 | 0.021 ± 0.005 | 0.015 ± 0.003 | 0.035 ± 0.008 |
| Dairy products | 31 | 0.075 ± 0.102 | 0.044 ± 0.06 | 0.12 ± 0.157 | 0.066 ± 0.09 | 0.041 ± 0.055 | 0.107 ± 0.142 |
| Yoghurt | 10 | 0.018 ± 0.017 | 0.008 ± 0.007 | 0.026 ± 0.024 | 0.015 ± 0.014 | 0.008 ± 0.007 | 0.023 ± 0.021 |
| Fermented milk | 5 | 0.008 ± 0.004 | 0.004 ± 0.002 | 0.012 ± 0.007 | 0.007 ± 0.004 | 0.004 ± 0.002 | 0.01 ± 0.006 |
| Condensed milk | 3 | 0.062 ± 0.068 | 0.019 ± 0.017 | 0.08 ± 0.084 | 0.051 ± 0.052 | 0.018 ± 0.017 | 0.069 ± 0.069 |
| Butter | 3 | 0.338 ± 0.052 | 0.174 ± 0.019 | 0.512 ± 0.061 | 0.301 ± 0.046 | 0.164 ± 0.009 | 0.464 ± 0.047 |
| Cheese | 7 | 0.081 ± 0.059 | 0.059 ± 0.058 | 0.14 ± 0.101 | 0.073 ± 0.054 | 0.054 ± 0.05 | 0.126 ± 0.089 |
| Cream | 3 | 0.115 ± 0.04 | 0.095 ± 0.049 | 0.21 ± 0.086 | 0.099 ± 0.032 | 0.085 ± 0.041 | 0.184 ± 0.071 |
| Eggs | 167 | 0.108 ± 0.262 | 0.042 ± 0.055 | 0.15 ± 0.29 | 0.092 ± 0.217 | 0.039 ± 0.053 | 0.13 ± 0.246 |
| Chicken eggs | 60 | 0.044 ± 0.02 | 0.014 ± 0.014 | 0.058 ± 0.029 | 0.038 ± 0.017 | 0.013 ± 0.013 | 0.051 ± 0.025 |
| Duck eggs | 62 | 0.177 ± 0.412 | 0.067 ± 0.076 | 0.244 ± 0.448 | 0.15 ± 0.341 | 0.063 ± 0.076 | 0.213 ± 0.379 |
| Egg products | 45 | 0.097 ± 0.106 | 0.044 ± 0.032 | 0.141 ± 0.121 | 0.082 ± 0.089 | 0.039 ± 0.026 | 0.122 ± 0.102 |
| Cereals | 89 | 0.015 ± 0.008 | 0.002 ± 0.001 | 0.017 ± 0.009 | 0.014 ± 0.008 | 0.002 ± 0.001 | 0.016 ± 0.008 |
| Cereals | 47 | 0.014 ± 0.008 | 0.002 ± 0.001 | 0.015 ± 0.008 | 0.012 ± 0.007 | 0.002 ± 0.001 | 0.014 ± 0.007 |
| Cereals products | 42 | 0.017 ± 0.008 | 0.002 ± 0.001 | 0.019 ± 0.009 | 0.016 ± 0.008 | 0.002 ± 0.001 | 0.018 ± 0.008 |
| Oil | 16 | 0.107 ± 0.066 | 0.019 ± 0.015 | 0.126 ± 0.07 | 0.098 ± 0.063 | 0.018 ± 0.014 | 0.116 ± 0.067 |
| Animal fat | 2 | 0.165 | 0.044 | 0.209 | 0.15 | 0.044 | 0.194 |
| Vegetable oil | 14 | 0.099 ± 0.066 | 0.016 ± 0.012 | 0.114 ± 0.067 | 0.091 ± 0.064 | 0.014 ± 0.01 | 0.105 ± 0.064 |
| Peanut oil | 6 | 0.121 ± 0.098 | 0.011 ± 0.005 | 0.132 ± 0.097 | 0.112 ± 0.094 | 0.01 ± 0.005 | 0.122 ± 0.093 |
| Sunflower seed oil | 2 | 0.101 | 0.014 | 0.115 | 0.093 | 0.015 | 0.108 |
| Soybean oil | 2 | 0.079 | 0.009 | 0.088 | 0.072 | 0.009 | 0.081 |
| Grape seed oil | 2 | 0.068 | 0.026 | 0.094 | 0.063 | 0.02 | 0.083 |
| Olive oil | 2 | 0.082 | 0.027 | 0.109 | 0.072 | 0.025 | 0.097 |
| Fruits | 27 | 0.008 ± 0.006 | 0.001 ± 0.001 | 0.009 ± 0.006 | 0.007 ± 0.005 | 0.001 ± 0.001 | 0.008 ± 0.005 |
| Fruit with peels | 12 | 0.009 ± 0.005 | 0.001 ± <0.001 | 0.01 ± 0.006 | 0.008 ± 0.004 | 0.001 ± <0.001 | 0.008 ± 0.005 |
| Fruit without peels | 15 | 0.008 ± 0.006 | 0.001 ± 0.001 | 0.009 ± 0.007 | 0.006 ± 0.005 | 0.001 ± 0.001 | 0.007 ± 0.006 |
| Vegetable | 301 | 0.011 ± 0.02 | 0.002 ± 0.003 | 0.012 ± 0.021 | 0.009 ± 0.02 | 0.002 ± 0.003 | 0.011 ± 0.021 |
| Bamboo shoot | 3 | 0.007 ± 0.004 | 0.001 ± <0.001 | 0.008 ± 0.004 | 0.006 ± 0.003 | 0.001 ± <0.001 | 0.007 ± 0.003 |
| Beans food | 21 | 0.012 ± 0.009 | 0.002 ± 0.002 | 0.014 ± 0.01 | 0.011 ± 0.008 | 0.002 ± 0.002 | 0.013 ± 0.009 |
| Beans | 15 | 0.016 ± 0.008 | 0.002 ± 0.002 | 0.018 ± 0.009 | 0.013 ± 0.007 | 0.002 ± 0.002 | 0.016 ± 0.008 |
| Beans products | 6 | 0.004 ± 0.001 | 0.001 ± <0.001 | 0.005 ± 0.001 | 0.004 ± 0.001 | 0.001 ± <0.001 | 0.005 ± 0.001 |
| Leafy vegetables | 152 | 0.012 ± 0.025 | 0.002 ± 0.002 | 0.014 ± 0.025 | 0.011 ± 0.025 | 0.002 ± 0.002 | 0.013 ± 0.026 |
| Melons | 18 | 0.003 ± 0.002 | <0.001 | 0.003 ± 0.002 | 0.002 ± 0.002 | <0.001 | 0.003 ± 0.002 |
| Mushroom | 24 | 0.009 ± 0.009 | 0.001 ± <0.001 | 0.01 ± 0.009 | 0.008 ± 0.008 | 0.001 ± <0.001 | 0.009 ± 0.009 |
| Root vegetable | 83 | 0.01 ± 0.017 | 0.002 ± 0.005 | 0.012 ± 0.021 | 0.008 ± 0.015 | 0.002 ± 0.005 | 0.01 ± 0.018 |

TEQ = toxic equivalency factor; WHO = World Health Organization.

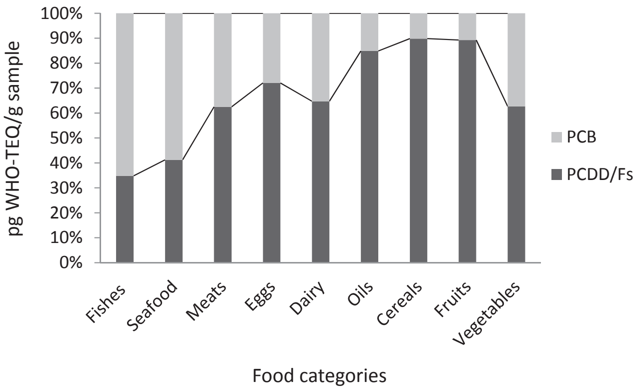


**Figure 1 – Levels of polychlorinated dibenzo-p-dioxin/ polychlorinated dibenzofuran (PCDD/Fs) and polychlorinated biphenyl (PCBs) in alternative food categories. TEQ = toxic equivalency factor; WHO = World Health Organization.**

milk and dairy products, fat and oil, and egg, respectively [9]; these data agrees with the current findings showing that PCBs contributed over 50% of dioxin-like components from fish and seafood.

In Vietnamese areas sprayed with Agent Orange, the levels in foods ranged from 3.2 pg WHO-TEQ/g to 8.2 pg WHO-TEQ/g fat [25]. The "high-risk foods" in regards to the samples cultivated locally (e.g., free-range chicken meat and eggs, ducks, freshwater fish, snail, and beef) showed PCDD/F concentrations in the range of 3.8 pg WHO-TEQ/g to 95 pg WHO-TEQ/g, while the concentrations in "low-risk foods" ranged from 0.03 pg WHO-TEQ/g to 6.1 pg WHO-TEQ/g (e.g., caged chicken meat and eggs, seafood, pork, leafy vegetables, fruits, and rice) [26]. All data analyzed from both "high-risk foods" and "low-risk foods" from Vietnam were clearly higher than those in Taiwan. In China, the levels of PCDDs, PCDFs, and dl-PCBs in different fish species varied significantly from 0.002 pg WHO-TEQ/g to 0.078 pg WHO-TEQ/g, from 0.002 pg WHO-TEQ/g to 0.553 pg WHO-TEQ/g, and from 0.003 pg WHO-TEQ/g to 2.059 pg WHO-TEQ/g fresh weight, respectively [27]. Hoogenboom et al [28] showed that the average level of PCDD/Fs and PCBs in Chinese mitten crab, an invasive species from Dutch rivers and lakes, was 43 pg WHO-TEQ/g sample, which was much higher than the values in Taiwan. Meanwhile, PCB levels and their variance were also higher than those in Taiwanese food. However, another study in China showed that the concentrations of PCDD/Fs and dl-PCBs were 0.25 pg $WHO_{98}$-TEQ/g weight and 0.32 pg $WHO_{98}$-TEQ/g weight in fatty fish [29], respectively, which were lower than those in Taiwan. In Spain, from 2006 to 2008, the WHO-TEQ level in 29 PCDD/Fs and dl-PCBs were 6.38 pg WHO-TEQ/g, 1.21 pg WHO-TEQ/g, and 0.90 pg WHO-TEQ/g in fish oil, fish and milk, and fat from dairy products, respectively. Only two fish oil levels were higher than the EU limits of total WHO-TEQ in all analyzed samples [30]. Additionally, the highest mean level for PCDD/Fs and dl-PCBs was 0.32 pg WHO-TEQ/g fat and 0.17 pg WHO-TEQ/g fat in 2012 for 102 raw cow milk samples collected from seven different regions in Chile [31]. The concentrations in milk samples both from Spain and Chile were all lower than the analyzed data in the present study (1.198 pg WHO-TEQ/g fat; data not shown). In Kocaeli, one of the most highly polluted areas in Turkey, the PCDD/F levels in various food samples including local eggs, milks, local meat samples, and local chickens were in the range of 1.16–10.94 pg TEQ/g fat, 0.43–3.29 pg TEQ/g fat, 0.28–1.81 pg TEQ/g fat, and 0.15–2.92 pg TEQ/g fat, respectively [32], which were higher than the findings in the present study. Therefore, except for the PCDD/F and dl-PCB levels in milk, the levels observed in Taiwanese food were lower than those observed in European countries. However, the variation observed in dioxin-like concentrations in Taiwanese food was higher than that in samples from other Asian countries.

### 3.2. *Time trend of PCDD/F and PCB distribution in foodstuffs*

In Taiwan, decreasing trends of PCDD/Fs or dl-PCBs were observed in meats, dairy, eggs, and vegetables, whereas increasing trends were observed in cereals, the levels for which were nearly equal to the levels in fruits and oils by the time. The ratio of PCBs to PCDD/Fs also largely varied in seafood compared to those in fish from 2004 to 2011 (Figures 2A and 2B). For meats (Figure 2C), PCDD/F levels were clearly higher than PCB levels from 2004 to 2012. The first decrease occurred from 2005 to 2008 and then the levels slightly increased in 2009, but decreased again from 2009 to 2012. For dairy (Figure 2D), eggs (Figure 2E), and vegetables (Figure 2F), the levels of PCDD/Fs and PCBs increased from 2004 to 2005, but decreased from 2005 to 2012. For cereals (Figure 2G), the levels increased from 2004 to 2011, whereas levels of PCDD/Fs and PCBs in fruits (Figure 2H) and oils (Figure 2I) were nearly equal during the alternative time shift. Internationally, PCDD/Fs and dl-PCB levels in nearly all samples decreased over time. In 2001, the Joint Food and Agriculture Organization/WHO Expert Committee on Food Additives derived a PTMI of 70 pg WHO-TEQ/kg body weight. The sum of the median intake of PCDD/Fs and PCBs exceeded the PTMI in western European countries. Therefore, the EU Commission developed a strategy for reducing the accumulation of PCDD/Fs and PCBs in the environment and in the food chain [11]. In Europe, the risk management of PCDD/Fs and PCB measurements were defined in 2001 [12]. In 2012, the comprehensive monitoring of PCDD/Fs and PCBs in dietary components in 26 European countries was revealed by the European Food Safety Authority. The data showed general decreases of 16.6% and 79.3% in dietary exposure to PCDD/Fs and dl-PCBs between 2002 and 2004 and 2008 and 2010, respectively. However, the exposed dose above the tolerable weekly intake of 14 pg TEQ/kg body weight ranged from 1.0% to 52.9% [11]. In the United States, the exposures of PCDD/Fs from food and environmental sources continued to decrease [13], as well as PCDD/F levels in the milk samples taken in Canada from 1992 to 2005 [33]. PCDD/F levels in food have also recently shown a decreasing trend in a Swedish market basket from 2005 [15]. The levels in hens' eggs also showed a significant reduction in Belgium [7], and the time trend showed significantly decreased PCDD/F concentrations in marine mussels from 1981 to 2005 at most sites. Even in an e-waste dismantling area of China, the measurements of PCDD/Fs, PCBs, and PBDEs showed a significantly decreasing

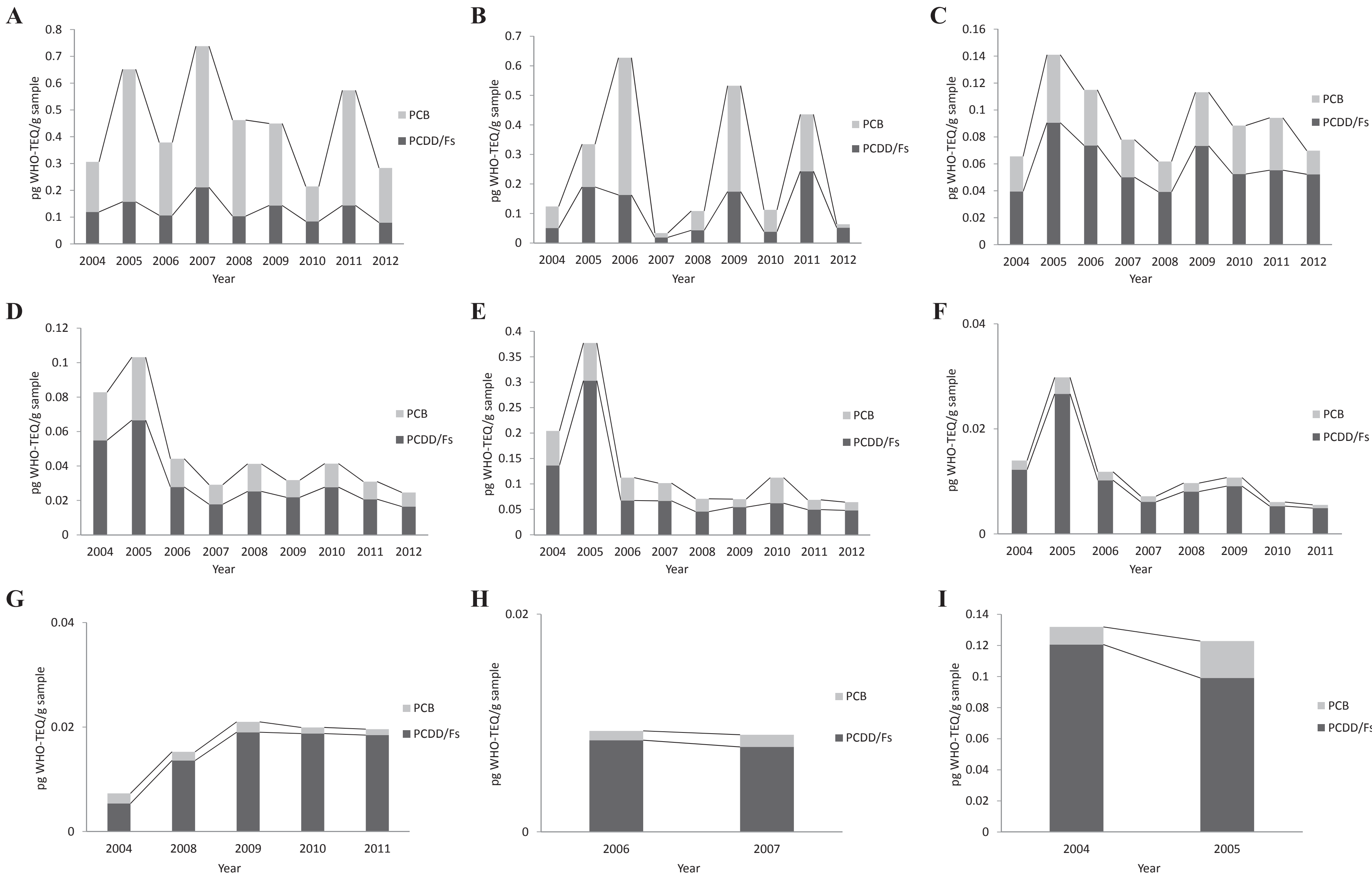


**Figure 2 – Levels of polychlorinated dibenzo-p-dioxin/polychlorinated dibenzofuran (PCDD/Fs) and polychlorinated biphenyl (PCBs) in food by temporal trend. (A) Fishes; (B) seafood; (C) meats; (D) dairy; (E) eggs; (F) vegetables; (G) cereals; (H) fruits; and (I) oils. TEQ = toxic equivalency factor; WHO = World Health Organization.**

temporal trends (2005–2009) in combustion components, including 2,3,4,7,8-PeCDF, 1,2,3,6,7,8HxCDF, and PCB126 [16]. Decreasing trends were observed in dietary intake of PCDD/Fs and PCBs in France [17], The Netherlands [18], Spain [19,34], and Japan [5] compared with their baseline values. However, in human milk, the median values of $WHO_{2005}$-TEQ of PCDD/Fs, dl-PCBs, and total PCBs changed from 1.5 pg/g milk lipid in 1992 to 0.8 pg/g lipid in 2005 [33], whereas a longitudinal increase was observed in PCDD/F and dl-PCB concentrations in northern China [35]. In 2002, the mean concentrations of PCDD/Fs plus dl-PCBs were 4.47 TEQ pg/g fat in human milk from Shijiazhuang and that showed a 39.6% increasing in 2007. The diversity trend was observed in humans and in food samples in China.

Overall, the current study showed decreasing total PCDD/Fs and dl-PCB levels in Taiwan, which were similar to the findings in other countries; although the trends in cereals, fruits, and oils showed elevated levels, their concentrations are very low compared with other categories. Meanwhile, the elevated trends may be because cereals and fruits are sometimes the major cultivated products in southern Taiwan, and PCDD/Fs and PCBs may be polluted in some places with industrial or incinerator areas. Therefore, PCDD/F and dl-PCB distributions should be further monitored.

### 3.3. *PCDD/Fs and dl-PCBs in different locations*

Among the four areas examined in Taiwan, the concentrations of PCDD/Fs and PCBs in alternative foodstuffs are shown in Figure 3. In fishes, the highest level of PCDD/Fs (Figure 3A) was observed in the north, followed by that in the east, south, and central Taiwan. The highest level of PCBs (Figure 3B) was observed in the east, followed by that in the north, south, and central Taiwan. In seafood, the highest level of PCDD/Fs (Figure 3A) was observed in the east, followed by that in the north, south, and central Taiwan. The highest level of PCBs (Figure 3B) was observed in the south, followed by that in the east, north, and central Taiwan. For meats and eggs, the highest level of PCDD/Fs or PCBs (Figures 3A and 3B) was observed in the south, followed by that in the north and central Taiwan. PCB levels in cereals, fruits, and vegetables were markedly lower than those in other food categories. Due to a notable PCDD/Fs level in ducks (3.660 pg WHO-TEQ/g, fat) obtained from central Taiwan [21], where an electric arc furnace dust treatment plant was suspected of being responsible for the public concern event after due environmental measurements. The PCDD/Fs levels of 0.055 pg WHO-TEQ/g in chicken eggs and 1.003 pg WHO-TEQ/g in duck eggs were utilized to analyze how eating contaminated duck eggs affected serum PCDD/F accumulation in family members of duck-egg farmers in central Taiwan [36]. However, no obviously elevated concentration in eggs obtained from central Taiwan in the present data.

In fish, the highest level of PCDD/Fs and PCBs was found in northern Taiwan, but the results were not consistent in seafood. For meats and eggs, the highest levels of PCDD/Fs or PCBs were observed in the south, followed by those in the north. The data were not consistent with the results of previous studies, which revealed that PCDD/Fs in meat samples, including beef, mutton, duck, and goose, were higher in northern Taiwan than in other areas, and the distribution of dl-PCB levels were also higher in these foods [22]. Taiwanese government forensic scientists have observed high PCDD/Fs and dl-PCB levels in mutton and beef, which are caused by the open burning of industrial and commercial wastes and by the use of polluted animal feed [22]. Additionally, PCB levels in cereals, fruits, and vegetables were markedly lower than the levels of PCDD/Fs, but the trends were the opposite of those for fishery products or meat samples. In fish, seafood, and meats, PCDD/Fs and dl-PCBs showed the lowest levels in central Taiwan. In seafood, the contaminant of PCDD/Fs versus dl-PCBs was not consistent among the three locations. This may be because the source of PCDD/Fs and dl-PCBs differed among the three locations. Meanwhile, the follow up when elevated levels are found seems to lead to a reduction of output from remaining sources, including industrial or incineration emission in the past decade [36].

Therefore, daily exposure may differ among the residents in the three locations. Further, risk assessment for the

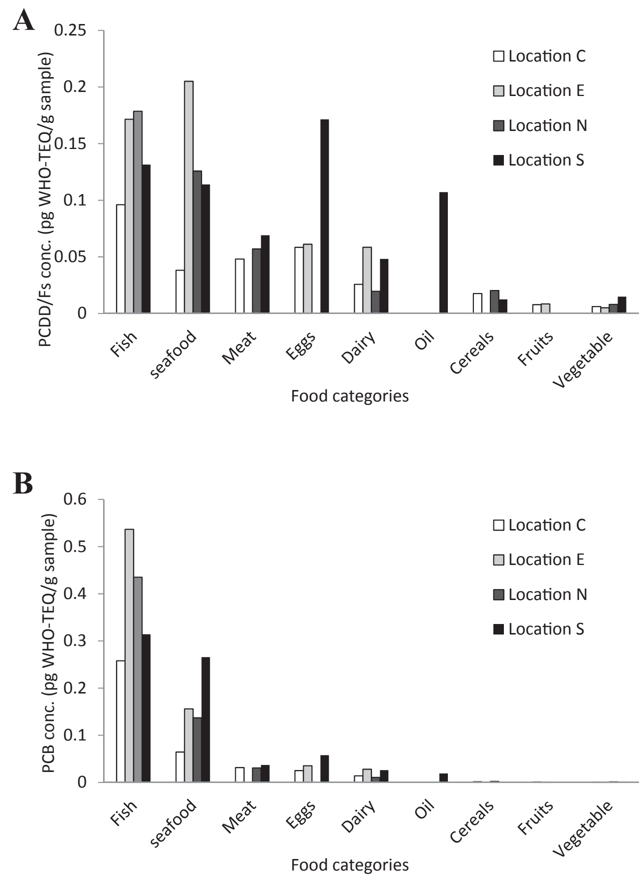


**Figure 3 – Polychlorinated dibenzo-p-dioxin/polychlorinated dibenzofuran (PCDD/Fs) and polychlorinated biphenyl (PCBs) concentrations of each food category in four locations around Taiwan. conc. = concentration; Location C = central Taiwan; Location E = east Taiwan; Location N = north Taiwan; Location S = south Taiwan; TEQ = toxic equivalency factor; WHO = World Health Organization.**

residents living in different areas should be adopted to ensure food safety while considering the overall food category.

Dl-PCBs contributed to 60–65% of the TEQ levels in fish and seafood, but only to 13–40% in meat and cereal samples. In Taiwan, decreasing trends of PCDD/F or dl-PCB levels were observed in meat, dairy, eggs, and vegetables, but cereals showed increasing levels that were similar to those in fruits and oils in the past decade. Cereals and fruits are important cultivars in southern Taiwan, and local pollution may seriously affect PCDD/F and PCB emission. In the future, risk assessment that is specific to different locations should be conducted for residents consuming local food to determine the overall food category simultaneously.

## Conflicts of interest

The authors declare that there are no conflicts of interest.

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