

TEMPORAL TRENDS AND SPATIAL DISTRIBUTION OF PCDD/FS AND DIOXIN-LIKE PCBS IN TAIWAN FOODSTUFFS

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Introduction:

Polychlorinated dibenzo-*p*-dioxins (PCDD) and dibenzofurans (PCDF) and polychlorinated biphenyls (PCB) are of global concern due to their persistence, bioaccumulation and toxicity¹. More than 90 % of the general human exposure to dioxins and dioxin-like PCBs (dl-PCBs) is estimated to occur through the diet, mainly meat and dairy products, fish and shellfish. Therefore, many national authorities have programmes in place to monitor the food supply.

Materials and methods:

From the year of 2004 to 2012, we had conducted a national survey of PCDD/Fs and dl-PCBs in high-lipid food, cereals, fruits & vegetables and all kinds of processed foods from traditional markets or supermarkets in selected towns around Taiwan, which were regulated by European Commission's standard for dioxins. At this stage, the foods were selected in each town with the greatest quantities. In the following stage during 2013-2018, the background levels of PCDD/Fs and dl-PCBs in selected foods based on the Taiwanese dietary habit derived from the Nutrition and Health Survey in Taiwan (NAHSIT) have been monitored in the Taiwan air quality area in sequence^{2,3}. These areas are North (including Taipei, New Taipei, and Taoyuan City) conducted in 2013, GAO-PING (Kaohsiung and Pingtung City) in 2014, YUN-JIA-NAN (Yunlin, Chiayi, and Tainan City) in 2015, Central section (Taichung, Changhua, and Nantou City) in 2016, ZHU-MAIO (Hsinchu and Miaoli City) in 2017, and HUA-DONG (Hualien and Taitung City) in 2018. Finally, 2441 individual foods in 14 categories have been collected and analyzed. The individual raw food sample for each air quality area was purchased in two or three cities, respectively. In addition, the individual brand sample was also purchased with 3 different brands. Finally, the same matrix of 3 food samples was homogeneously mixed into 1 food sample and then frozen at -20 °C until analysis. We estimated the average daily dose (ADD) of PCDD/Fs/dl-PCBs based on the ingestion rate of foodstuffs by a gender- and age-specified 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 item. The lifetime ADD (LADD) was estimated using Monte Carlo simulation with 10,000 random iterations, and compared with the WHO's tolerable daily intake (TDI) and EFSA's tolerable weekly intake (TWI).

Results and discussion:

The highest PCDD/Fs/dl-PCBs levels based on per gram fat (pg WHO₀₅-TEQ_{PCDD/F+PCB}/g fat) were found in eggs [1.04 (0.191- 20.7)], followed by dairy products [0.959 (0.085- 3.95)], and livestock and their products [0.681 (0.034- 7.81)], and the lowest level was found in Candies and Snacks [0.081 (0.065- 0.110)] (Table 1).

Based on per gram of wet weight (pg WHO₀₅-TEQ_{PCDD/F+PCB}/g w.w), the highest PCDD/Fs/dl-PCBs levels were found in fish and aquatic products [0.343 (0.005- 12.4)], followed by eggs [0.125 (0.011- 2.62)], and oils [0.091 (0.010- 0.305)], and the lowest level was found in fruits and their products [0.005 (0.001- 0.027)].

There was a decreasing trend of PCDD/Fs/dl-PCBs levels in 2013-2018 comparing with the measurement investigated in 2004-2012. As for the comparisons of the PCDD/Fs/dl-PCBs levels between the different sampling areas, the levels of PCDD/Fs/dl-PCBs in livestock products, eggs and milk samples have been gradually decreasing in recent years with the exception of poultry products, fish, and seafood. However, vegetables were all lower than 0.03 pg WHO₀₅-TEQ/g w.w except for the measurement conducted in 2005. The major contribution of total TEQ_{PCDD/F+PCB} were mainly from duck meat and goose meat. The reason is that ducks and geese are often feeding in the open field, and soil and water sources are more susceptible to be polluted by the dioxin-containing particle emitted from the nearby anthropogenic activity, while ducks and geese are also ingested by these particles and cause the dioxin accumulation.

The highest lifetime average daily dose (LADD) for residents were those living in GAO-PING, which was 0.397 pg WHO₀₅-TEQ_{PCDD/F+PCB}/kg BW/day, followed by YUN-JIA-NAN (0.385), Central section (0.273), ZHU-MAIO (0.247), North (0.236) and the lowest level was found in HUA-DONG (0.188). All of the LADD in the current sampling area were below the TDI (1- 4 pg WHO₀₅-TEQ_{PCDD/F+PCB}/kg BW/day) and TWI (2 pg WHO₀₅-TEQ_{PCDD/F+PCB}/kg BW/week). Moreover, the main contribution of the LADD of PCDD/Fs/dl-PCBs were from fish and aquatic products (29.3-60.7%), followed by livestock and their products (8.2-25.9 %) and cereals grains (3.5-10.0 %) in 6 areas (Figure 1). According to the ambient air monitoring for dioxins from Taiwan air quality area during 2013-2017 derived from Taiwan EPA, the annual data of the air dioxin is highest in the YUN-JIA-NAN area, and the following is GAO-PING, Central section, ZHU-MAIO, North and HUA-DONG area. The pattern of ambient air monitoring for dioxins were comparable to the dietary measurement of dioxins of the residents in each air quality area. There's a number of reasons for the findings that LADD of residents in GAO-PING area was the highest and the main source of contribution is fish and aquatic products (0.241/0.397= 60.7%). Because one port city of this area, Kaohsiung City, has the important town of offshore fishing industry, and PCDD/Fs/dl-PCBs levels in fish and aquatic products and the intake of fish for the residents were both the highest than those in other regions.

From 2003-2007, the measurement of 19 samples were exceeding regulatory standard. After 2008, only one duck and one chicken egg was illegal in 2014 and 2017, respectively. These result shows the effectiveness of contaminative source control of dioxins. However, it is recommended that food originated from the counties and administrate districts with higher risk for PCDD/Fs and dl-PCBs contamination should be continuously monitored to ensure the safety and hygiene of food.

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References:

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

	N	PCDD/Fs+dl-PCBs (pg TEQ/g fat)	N	PCDD/Fs+dl-PCBs (pg TEQ/g wet weight)
Cereal grains	5	0.280 (0.147-0.431)	138	0.016 (0.002-0.050)
Nuts and Beans	24	0.118 (0.016-0.461)	39	0.018 (0.002-0.070)
Fats and Oils	41	0.116 (0.019-0.677)	41	0.091 (0.010-0.305)
Poultry and their products	246	0.651 (0.087-5.90)	247	0.065 (0.007-0.782)
Livestock and their products	332	0.681 (0.034-7.81)	338	0.087 (0.003-2.07)
Fish and Aquatic products	—	—	546	0.343 (0.005-12.4)
Eggs	195	1.04 (0.191-20.7)	196	0.125 (0.011-2.62)
Dairy products	282	0.959 (0.085-3.95)	284	0.043 (0.002-0.505)
Fruits and their products	—	—	70	0.005 (0.001-0.027)
Vegetables	—	—	371	0.011 (0.001-0.295)
Candies and Snacks	3	0.081 (0.065-0.110)	11	0.018 (0.007-0.025)
Beverages	1	0.211	5	0.005 (0.003-0.006)
Seasonings	25	0.242 (0.050-0.667)	41	0.026 (0.002-0.245)
Composite foods and Soups	81	0.229 (0.043-0.955)	114	0.023 (0.004-0.120)

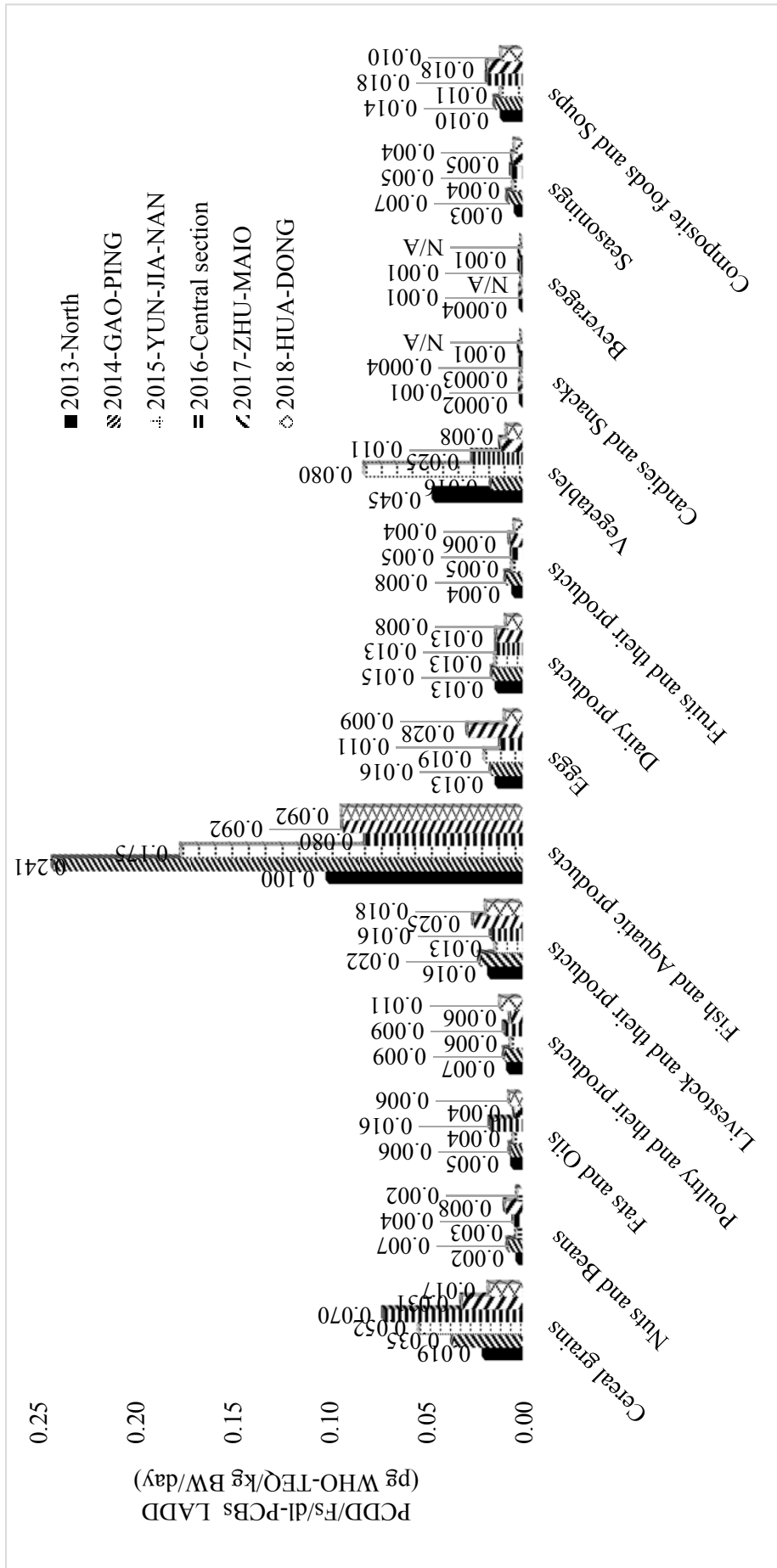


Figure 1. Time trends of PCDD/Fs and dl-PCBs LADD in 6 Taiwan air quality areas.