THE TIME TREND OF DIETARY INTAKE OF PCDD/Fs AND DIOXIN-LIKE PCBs FOR GENERAL POPULATION IN CHINA (2000-2007)

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Introduction

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) are ubiquitous, bioaccumulative, toxic and persistent lipophilic chemicals. Due to their severe toxic effects, including carcinogenicity, immunotoxicity and adverse effects on reproduction and development, and bioaccumulation / biomagnification in the food chain, the occurrence of these chemicals in foods has been a matter of great concern in the world. Some studies have shown notable decline of contamination levels in food and dietary exposure to PCDD/Fs and dl-PCBs of general population during the last decade in some developed countries[1-4], which probably results from strict regulations on dioxins emissions in those countries.

In our previous study, dietary intake of PCDD/Fs and dl-PCBs of adult population was estimated from Chinese total diet study (TDS) in 2000[5]. With rapid industrialization, potential health risk associated with various chemical contaminants released from industries may be significant in China. It is urgent to regularly monitor levels of chemical contaminants in various food samples and assess human health risk as well as trends of levels of contamination and dietary exposure. In this study, levels of PCDD/Fs and dl-PCBs were measured in 96 composite food samples collecting in the Chinese total dietary study in 2007, including aquatic foods, meat and meat products, egg and egg products, milk and dairy products, cereals, bean products, potatoes, and vegetables. The time trend of the dietary intake of PCDD/Fs and dl-PCBs was estimated for the Chinese population between 2000 and 2007.

Materials and methods

The Chinese TDS is a national survey to investigate the levels of various chemical contaminants in foods and estimate the dietary intake of these contaminants for general population in China, which has been described elsewhere[5,6]. The food composites approach was used to study the total diet in 12 provinces representing the average dietary patterns of various provinces of China and covering about 50% of the total Chinese population. Food consumption pattern in each provinces involved in TDS was determined by a 3-day household dietary survey which documented all the food consumed by a weighing and recording method. These provinces consist of Heilongjiang (HLJ), Liaoning (LN), Hebei (HeB), Shanxi (ShX), Ningxia (NX), Henan (HeN), Shanghai (ShH), Fujian (FJ), Jiangxi (JX), Guangxi (GX), Hubei (HuB) and Sichuan (SC).

Food samples were collected from local food markets, grocery stores or rural households in each survey site of each province， and then cooked and prepared according to the local habits. The prepared foods were blended to form composites with weights proportional to the average daily consumption for the province. These provincial composites were shipped to the laboratory and frozen at -20℃ until analysis. Eight food groups were selected to investigate the levels of PCDD/Fs and dl-PCBs and estimate the dietary exposure in this study: aquatic foods, meat and meat products, egg and egg products, milk and dairy products, cereals, bean products, potatoes and vegetables.

Approximately 50 g of composite sample was freeze-dried before being blended with silica-gel. After spiking with ^{13}C12-labeled internal standards, samples were extracted with a mixture of n-hexane and dichloromethane (1:1, v/v) using accelerated solvent extractor at 150 ℃and 1500 psi. The bulk lipid was removed by shaking with acid-modified silica-gel, and further cleanup was achieved using a Power Prep instrument with multiple commercial silica-gel columns, alumina columns and carbon columns. Two fractions containing PCDD/Fs and dl-PCBs were collected and concentrated to approximately 20 μL, respectively. Prior to instrumental analysis,

the ^{13} C12-labeled injection standards for PCDD/Fs and dl-PCBs were added to the final extracts, respectively. The analysis of 17 toxic congeners of 2,3,7,8-substituted PCDD/Fs and 12 congeners of dl-PCBs was performed by a gas chromatograph – high resolution mass spectrometer (GC-HRMS, MAT95XP, ThermoFinnigan, Germany) with DB-5MS capillary column (60 m × 0.25 mm i.d. ×0.25μm) using an isotopic dilution method for quantification. Toxic equivalents (TEQ) of the analyzed PCDD/Fs and dl-PCBs were calculated using the recommended toxic equivalency factors (TEF) by WHO in 1998[7].

Results and discussion

Table 1 depicts levels of PCDD/Fs and dl-PCBs, expressed as pg TEQ /g (fresh weight, fw), in selected food composites from 12 provinces in China. The concentrations of total TEQs of PCDD/Fs and dl-PCBs ranges from 0.001 pg TEQ /g to 0.85 pg TEQ /g in all composite samples. The Concentrations of PCDD/Fs and dl-PCBs in animal origin food samples were all higher than that in the plant origin food, and the levels of total TEQs corresponded to aquatic foods, which is coherent with other studies.

Table 1 the concentrations of total TEQs in the food composite samples from China (pg TEQ/g fw).

	HLJ	LN	HeB	ShX	NX	HeN	ShH	FJ	JX	GX	SC	HuB
Aquatic food[a]	0.21	0.31	0.34	0.18	0.12	0.14	0.69	0.32	0.85	0.38	0.25	0.77
Meat and meat products[a]	0.08	0.14	0.27	0.07	0.10	0.11	0.12	0.12	0.19	0.23	0.14	0.08
Egg and egg products[a]	0.15	0.09	0.22	0.09	0.08	0.17	0.14	0.19	0.14	0.12	0.16	0.10
Milk and dairy products[a]	0.10	0.11	0.18	0.09	0.14	0.12	0.16	0.06	0.50	0.19	0.06	0.41
Cereals[b]	0.004	0.001	0.007	0.001	0.002	0.003	0.002	0.016	0.001	0.003	0.005	0.011
Bean products[b]	0.005	0.004	0.003	0.005	0.002	0.003	0.003	0.019	0.003	0.005	0.002	0.004
Potatoes[b]	0.005	0.005	0.009	0.001	0.002	0.005	0.013	0.012	0.013	0.006	0.005	0.007
Vegetables[a,b]	0.003	0.024	0.021	0.001	0.002	0.003	0.002	0.002	0.010	0.033	0.015	0.007

a: ND=LOD
b: ND=0

Estimated dietary intake of PCDD/Fs and dl-PCBs was calculated by multiplying the TEQs of PCDD/Fs and dl-PCBs by the consumption data from the food consumption survey in Chinese TDS. Table 2 shows the estimated dietary intakes of PCDD/Fs and dl-PCBs for adults per day and per month in 12 provinces of China. Large geographical variation of dietary exposure was found among regions in China. Among various provinces, the daily dietary intake of total TEQs varied by a factor of approximately 10 from the minimum of 0.17 pg TEQ /kg.bw in Shanxi to the maximum of 1.55 pg TEQ /kg.bw in Shanghai. The large difference among regions presumably results from variations in contamination levels as well as food consumption values in different regions.

The monthly dietary intakes for adult populations in China were all lower than the provisional tolerable monthly intake (PTMI) of 70 pg TEQ kg-1 bw recommended by Joint FAO/WHO Expert Committee on Food Additives in 2001[8], which indicated the low risk of PCDD/Fs and dl-PCBs to the general populations in China.

The dietary intakes of PCDD/Fs and dl-PCBs from animal origin food have been evaluated in China by TDS 2000[6]. There are slight differences on methodology between TDS 2000 and TDS 2007. In TDS 2000, all the 12 provinces were divided into four regions, including North 1 (HLJ, LN, and HeB), North 2 (ShX, NX, and HN), South 1 (ShH, FJ, and JX), and South 2 (GX, HuB, and SC), and only composite samples of animal origin from the four regions were involved. The dietary exposures of the adults from these four regions in 2007 were also calculated in this study. Fig.1depicts the comparison of dietary intake to PCDD/Fs and dl-PCBs from animal origin food between TDS 2000 and TDS 2007 in China. For the average dietary exposure between these two studies, a slight increase with about 6% was observed from 2000 to 2007. For each region, in North 2 and South 1, there was slight increase with about 13% and 4%, respectively, and in South 2, a notable increase with about 78% was observed. However, in North 1, dietary exposure decreased approximately 27% from 2000 to 2007. Some previous studies have indicated that notable decline of dietary exposure is observed in certain European countries and Japan presumably due to enforcing legislation to reduce exposure to PCDD/Fs and dl-PCBs and strict implementation of control measures[1-4]. Thus, it is very urgent to enforce legislation to reduce dioxin

emission and human exposure to protect the environment and human health in China. Moreover, further studies should be conducted to regularly monitor human dietary exposure to PCDD/Fs and dl-PCBs and clarify the time trend of exposure in China.

Table 2 the estimated dietary intakes of PCDD/Fs and dl-PCBs for adults in 12 provinces of China

	dietary intakes of PCDD/Fs and dl-PCBs	
	pg TEQ/kg bw.day	Pg TEQ/kg bw. month
HLJ	0.47	14.2
LN	0.75	22.4
HeB	0.70	21.1
SHX	0.17	5.0
HeN	0.33	9.8
NX	0.18	5.4
ShH	1.55	46.4
FJ	1.29	38.8
JX	0.46	13.8
HuB	1.07	32.2
SC	0.75	22.6
GX	0.67	20.2
Average	0.70	21.0

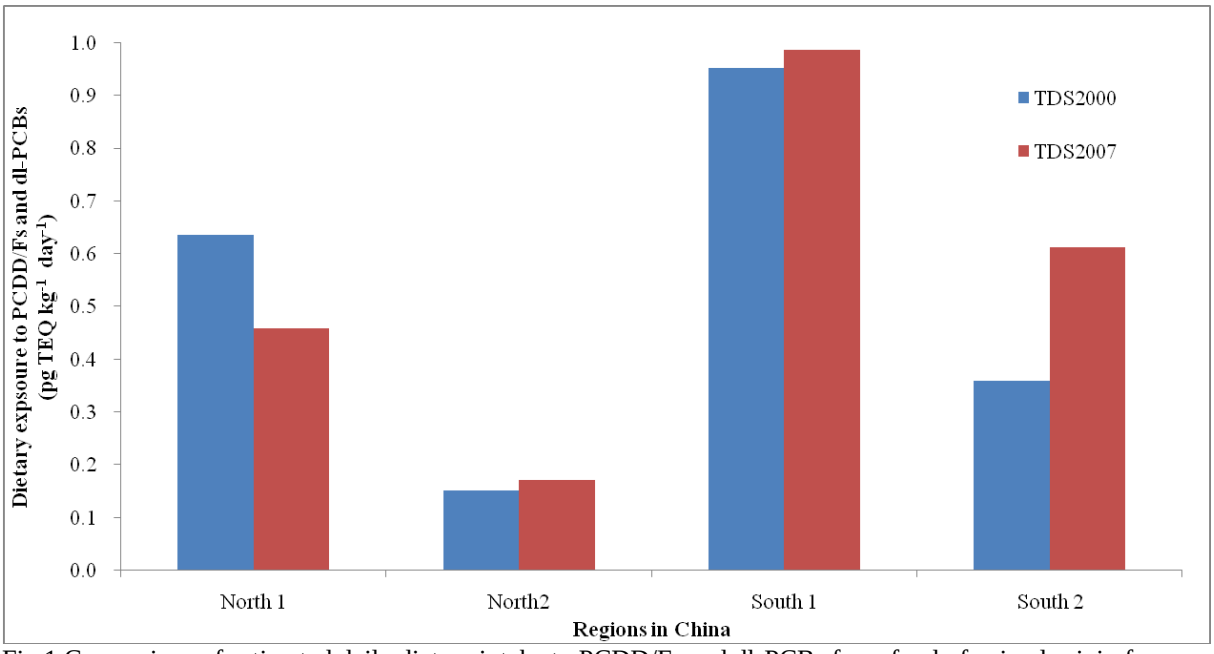

Fig.1 Comparison of estimated daily dietary intake to PCDD/Fs and dl-PCBs from food of animal origin from TDS 2000 and TDS 2007 in China

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