Patterns and dietary intake of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans in food products in China

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ABSTRACT

The health risk of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and dioxin-like PCBs (dl-PCBs) to human being should be assessed regularly. To evaluate the contamination levels in various food products in the Chinese market and to assess the dietary exposure of the Chinese population, 11 varieties of food groups totaling 634 samples including beef and mutton, chicken and duck, pork, fish and seafood, milk and dairy products were evaluated. The average concentrations of PCDD/Fs in all groups ranged from 0.291 to 8.468 pg/g whole weight (w.w.). The average toxic equivalency concentrations were from 0.012 pg TEQ/g w.w. for cereal to 0.367 pg TEQ/g fat for marine oil. OCDD and 2,3,7,8-TCDF were the dominant congeners in foodstuffs. The dietary estimated mean intake for the Chinese rural and urban populations were 0.656 and 0.514 pg TEQ/kg body weight/day, respectively, however, the cereal group exposure were higher to the estimate daily intake and contributed 81% for rural and 48% for urban population, followed by fish and seafood which contributed 4% and 16% to the estimate daily intake. The estimated dietary intakes were compared with the toxicological reference values and showed that both rural and urban populations were well below those values.

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Introduction

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are persistent organic pollutants, which are formed and released into the environment during combustion processes or represent unwanted by-products of various industrial processes. Many adverse effects on human health have been reported, including hepato- and immunotoxicity, carcinogenicity, endocrine and reproductive impairments, and embryotoxicity (Van den Berg et al., 1998). Because of their lipophilic characteristic, PCDD/Fs could accumulate in the food chain, such as in aquatic and terrestrial (Domingo and Bocio,
About 90% of human exposure occurs through the ingestion of food, such as meat, dairy products, eggs and egg products, fish and fish products (Gilman et al., 1991; Olanca et al., 2014; Zhang et al., 2008).

Since the World Health Organization (WHO) established a tolerable daily intake (TDI) for PCDD/Fs in the range of 1–4 pg WHO-TEQ/kg body weight (b.w.) (Rolaf van Leeuwen et al., 2000), many countries have started monitoring of PCDD/F levels in food to assess the potential risk to their population (Dashti et al., 2013; Husain et al., 2014; Kim et al., 2013; Loutfy et al., 2007; Olanca et al., 2014; Pizarro-Aranguiz et al., 2015; Rauscher-Gabernig et al., 2013; Vassiliadou et al., 2011). In this present study, concentrations of PCDD/Fs were measured in various media, such as in rivers, lakes, and coastal areas, were reported (Bao et al., 2012), but very limited information about human daily intake (EFSA, 2010, 2012).

In China, the concentrations of PCDD/Fs in the environment media, such as in rivers, lakes, and coastal areas, were reported (Bao et al., 2012), but very limited information about PCDD/F intake through food is available (Labunksa et al., 2015; Li et al., 2007; Pan et al., 2013; Shen et al., 2012; Song et al., 2011; Wang et al., 2015; Zhang et al., 2013). In this present study, concentrations of PCDD/Fs were measured in various food samples collected from Chinese markets from 2011 to 2014. Dietary intakes were estimated for different population groups using the most common foods consumed, and the contributions of different food groups to the total dietary intake were also analysed.

1. Materials and methods

1.1. Chemicals and reagents

Acetone, n-hexane, toluene, ethyl acetate and dichloromethane were purchased from J. T. Baker, Co., Ltd. (USA). The PCDD/F standards including calibration standard solutions, internal surrogate standards (EPA 1613-LCS) and injection standards (EPA 1613-IS) were obtained from Wellington Laboratories (Canada). Silica gel 60 and sulfuric acid were purchased from Merck (Darmstadt, Germany).

1.2. Sampling

The sampling of food was based on the main diet consumed by Chinese people. The samples were collected from factories, retail markets and supermarkets from China during 2011–2014 and were classified according to the food type that mentioned of EU Commission Regulation (EC) No 1881/2006. The type and the number of foodstuffs comprised 11 groups and were listed in Table 1. The meat samples were homogenized using a blender and stored at –20°C until analysis. Milk, canned food, fruit juice and fruit jam samples were kept at 4°C. Powdered milk and oil were kept at room temperature until analysis.

1.3. Sample extraction and analysis

Only the edible parts were analysed and each sample was determined individually. Milk samples were frozen-dried and meat samples were completely mixed with diatomite to remove moisture before extraction. Samples were extracted by Accelerated Solvent Extractor (ASE300, Dionex, USA) at 150°C and 1500 psi after spiking with 13C12-labelled internal standards. The extraction solvent was n-hexane and dichloromethane (1:1, V/V). The solvent was evaporated to about 1 mL, and then 44% acid-modified silica-gel was used to remove lipid. Briefly, the concentrate was re-suspended with 100 mL n-hexane and 44% sulfuric acid-modified silica-gel was added, then, shaking to remove lipid roughly. After that, an automated system (Power Prep, Fluid Management Systems, Waltham, MA, USA) was used for further clean-up and PCDD/Fs and dl-PCB fractions were separated. Finally, the sample was concentrated into 20 μL nonane. Before instrumental analysis, the injection standard was added for the recovery calculation of the surrogate standards.

PCDD/Fs were quantified using high resolution gas chromatograph–high resolution mass spectrometer (HRGC-HRMS) (DFS, Thermo, USA/AutoSpec, Waters, USA) with DB-5 MS capillary column (60 m × 0.25 mm i.d. × 0.25 μm). Instrumental conditions were according to Zhang et al. (2008).

1.4. Calculation

The limits of detection (LODs) for Tetra- and Penta-CDDs/Fs were around 0.0006–0.27 pg/g in meat and milk samples, and 0.01–0.69 pg/g in oil, fat and other food products; for Hexa- and Hepta-CDDs/Fs, 0.0006–0.15 pg/g in meat and milk samples, and 0.01–0.73 pg/g in oil, fat and other food products, and for Octa-CDD/F, 0.0007–0.36 in meat and milk samples, and 0.01–0.75 pg/g in oil, fat and other food products.

Considering lower and upper bound values, when the congener was reported as non-detected, the lower bound value was taken as zero and the upper bound value was the Limit of Detection (LOD).

WHO 2005 toxic equivalency factors were used to calculate the TEQ of each sample (Van den Berg et al., 2006).

1.5. Quality assurance and control

A blank sample was carried out for each batch of eight samples. The recoveries of 13C12-labelled internal standards met the requirements of USEPA 1613 method and all were in the range of 60%–120%. Since 2006, the laboratory has participated in international comparisons for the detection of PCDD/Fs and dl-PCBs in food, feed and environment samples.

1.6. Dietary exposure of PCDD/Fs

Food consumption data of the residents were obtained from the China Health and Nutrition Survey (CHNS) (Zhou et al., 2012). The average daily intakes of PCDD/Fs were estimated by multiplying the measured concentrations of PCDD/Fs (expressed in pg TEQ/g whole weight (w.w.)) by the average daily consumption of each type of food (expressed in g/day) and divided by an average adult body weight of 60 kg. The average daily intakes were reported as pg TEQ/kg b.w./day. That is, Daily intake = Measured concentration × Daily consumption / Body weight (60 kg).
**Table 1 – Number of samples from each food category and mean concentrations of PCDD/Fs in different food groups.**

<table>
<thead>
<tr>
<th>Congeners</th>
<th>Detected</th>
<th>Beef and mutton (N = 88)</th>
<th>Chicken and duck (N = 90)</th>
<th>Pork (N = 64)</th>
<th>Fish and seafood (N = 94)</th>
<th>Milk and dairy products (N = 55)</th>
<th>Pig fat (N = 7)</th>
<th>Ruminants and poultry fat (N = 12)</th>
<th>Vegetable Oils and fats (N = 51)</th>
<th>Marine oil (N = 19)</th>
<th>Cereal (N = 23)</th>
<th>Other food products (N = 131)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1,2,3,4,7,8-HpCDD</td>
<td>284</td>
<td>0.068</td>
<td>0.01</td>
<td>0.043</td>
<td>0.028</td>
<td>0.004</td>
<td>0.187</td>
<td>0.278</td>
<td>0.193</td>
<td>0.161</td>
<td>0.040</td>
<td>0.075</td>
</tr>
<tr>
<td>1,2,3,4,7,8-HxCDD</td>
<td>67</td>
<td>0.004</td>
<td>&lt;0.001</td>
<td>0.003</td>
<td>0.003</td>
<td>0.004</td>
<td>0.059</td>
<td>0.018</td>
<td>0.006</td>
<td>0.018</td>
<td>&lt;0.001</td>
<td>0.005</td>
</tr>
<tr>
<td>1,2,3,4,6,7,8-HxCDD</td>
<td>138</td>
<td>0.019</td>
<td>&lt;0.001</td>
<td>0.006</td>
<td>0.008</td>
<td>0.015</td>
<td>0.094</td>
<td>0.057</td>
<td>0.023</td>
<td>0.046</td>
<td>0.003</td>
<td>0.013</td>
</tr>
<tr>
<td>1,2,3,7,8,9-HxCDD</td>
<td>76</td>
<td>0.004</td>
<td>&lt;0.001</td>
<td>&lt;0.001</td>
<td>0.003</td>
<td>0.004</td>
<td>0.063</td>
<td>0.011</td>
<td>0.008</td>
<td>0.024</td>
<td>0.003</td>
<td>0.010</td>
</tr>
<tr>
<td>1,2,3,7,8-PeCDD</td>
<td>122</td>
<td>0.012</td>
<td>0.01</td>
<td>0.002</td>
<td>0.015</td>
<td>0.014</td>
<td>0.071</td>
<td>0.037</td>
<td>0.008</td>
<td>0.077</td>
<td>&lt;0.001</td>
<td>0.018</td>
</tr>
<tr>
<td>2,3,7,8-TCDD</td>
<td>93</td>
<td>0.005</td>
<td>0.01</td>
<td>&lt;0.001</td>
<td>0.007</td>
<td>0.003</td>
<td>0.021</td>
<td>0.006</td>
<td>0.002</td>
<td>0.031</td>
<td>&lt;0.001</td>
<td>0.006</td>
</tr>
<tr>
<td>OCDD</td>
<td>400</td>
<td>0.178</td>
<td>0.10</td>
<td>0.452</td>
<td>0.323</td>
<td>0.177</td>
<td>0.620</td>
<td>0.707</td>
<td>7.186</td>
<td>0.380</td>
<td>0.314</td>
<td>0.287</td>
</tr>
<tr>
<td>1,2,3,4,7,8-HpCDF</td>
<td>401</td>
<td>0.036</td>
<td>0.08</td>
<td>0.023</td>
<td>0.017</td>
<td>0.044</td>
<td>0.166</td>
<td>0.279</td>
<td>0.448</td>
<td>0.087</td>
<td>0.040</td>
<td>0.319</td>
</tr>
<tr>
<td>1,2,3,4,7,8,9-HpCDF</td>
<td>71</td>
<td>0.003</td>
<td>0.03</td>
<td>0.002</td>
<td>&lt;0.001</td>
<td>0.005</td>
<td>0.069</td>
<td>0.023</td>
<td>0.007</td>
<td>0.014</td>
<td>&lt;0.001</td>
<td>0.117</td>
</tr>
<tr>
<td>1,2,3,4,7,8-HxCDF</td>
<td>371</td>
<td>0.064</td>
<td>0.01</td>
<td>0.021</td>
<td>0.016</td>
<td>0.082</td>
<td>0.109</td>
<td>0.309</td>
<td>0.159</td>
<td>0.089</td>
<td>0.019</td>
<td>0.375</td>
</tr>
<tr>
<td>1,2,3,7,8,9-HxCDF</td>
<td>272</td>
<td>0.043</td>
<td>0.03</td>
<td>0.012</td>
<td>0.011</td>
<td>0.059</td>
<td>0.083</td>
<td>0.195</td>
<td>0.099</td>
<td>0.093</td>
<td>0.010</td>
<td>0.139</td>
</tr>
<tr>
<td>1,2,3,7,8-PeCDF</td>
<td>38</td>
<td>&lt;0.001</td>
<td>&lt;0.001</td>
<td>&lt;0.001</td>
<td>&lt;0.001</td>
<td>0.001</td>
<td>0.064</td>
<td>&lt;0.001</td>
<td>0.006</td>
<td>0.019</td>
<td>&lt;0.001</td>
<td>0.087</td>
</tr>
<tr>
<td>1,2,3,7,8,9-PeCDF</td>
<td>345</td>
<td>0.007</td>
<td>0.03</td>
<td>0.005</td>
<td>0.041</td>
<td>0.013</td>
<td>0.063</td>
<td>0.132</td>
<td>0.029</td>
<td>0.286</td>
<td>0.014</td>
<td>0.318</td>
</tr>
<tr>
<td>2,3,4,6,7,8-HxCDF</td>
<td>237</td>
<td>0.032</td>
<td>0.01</td>
<td>0.002</td>
<td>0.013</td>
<td>0.044</td>
<td>0.071</td>
<td>0.155</td>
<td>0.034</td>
<td>0.103</td>
<td>0.007</td>
<td>0.108</td>
</tr>
<tr>
<td>2,3,4,7,8-PeCDF</td>
<td>375</td>
<td>0.064</td>
<td>0.03</td>
<td>0.013</td>
<td>0.095</td>
<td>0.091</td>
<td>0.113</td>
<td>0.184</td>
<td>0.068</td>
<td>0.534</td>
<td>0.010</td>
<td>0.151</td>
</tr>
<tr>
<td>2,3,7,8-TCDF</td>
<td>363</td>
<td>0.009</td>
<td>0.08</td>
<td>0.014</td>
<td>0.180</td>
<td>0.023</td>
<td>0.047</td>
<td>0.138</td>
<td>0.054</td>
<td>0.486</td>
<td>0.041</td>
<td>0.386</td>
</tr>
<tr>
<td>OCDF</td>
<td>105</td>
<td>0.003</td>
<td>&lt;0.001</td>
<td>0.007</td>
<td>0.022</td>
<td>0.023</td>
<td>0.219</td>
<td>0.013</td>
<td>0.189</td>
<td>0.051</td>
<td>0.021</td>
<td>0.177</td>
</tr>
<tr>
<td>Sum of PCDFs</td>
<td></td>
<td>0.551</td>
<td>0.42</td>
<td>0.605</td>
<td>0.782</td>
<td>0.626</td>
<td>2.119</td>
<td>2.542</td>
<td>8.468</td>
<td>2.499</td>
<td>0.522</td>
<td>0.291</td>
</tr>
<tr>
<td>Total PCDD/Fs (pg WHO TEQ/g)</td>
<td></td>
<td>0.226</td>
<td>0.343</td>
<td>0.278</td>
<td>0.075</td>
<td>0.255</td>
<td>0.192</td>
<td>0.272</td>
<td>0.073</td>
<td>0.367</td>
<td>0.012</td>
<td>0.089</td>
</tr>
</tbody>
</table>

**a** pg TEQ/g fat for animal origin food and oil except fish and seafood, pg TEQ/g whole weight for fish and seafood, cereals and other food products. Lower bound values were applied.

**b** Number of samples > LOD of all analysed samples.
2. Results and discussion

2.1. PCDD/Fs concentrations in different food group

Table 1 shows the mean concentrations of 17 PCDD/Fs congeners in each food group. The average mass concentrations of PCDD/Fs ranged from 0.291 pg/g w.w. for other food group to 8.468 pg/g w.w. for vegetable oil and fat. For the other food groups, the average mass concentrations were 2.499 pg/g w.w. for marine oil, 0.782 pg/g w.w. for fish and seafood, 0.551 pg/g w.w. for beef and mutton, 0.626 pg/g w.w. for milk and dairy products and 0.522 pg/g w.w. for cereal. These results were similar or a little lower than previous reports (Loutfy et al., 2007; Marin et al., 2011; Zhang et al., 2008).

The distribution of PCDD/Fs TEQ concentrations in individual food sample and the mean concentration of each food group are presented in Fig. 1. Upper bound values were applied. Almost all the analysed samples were under the maximum limit levels for the sum of PCDD/Fs set by the European Union Commission (EN Commission Regulation (EC) No 1881/2006). Only two chicken (3.170 pg TEQ/g fat, 3.030 pg TEQ/g fat) and one pig fat (1.250 pg TEQ/g fat) samples were found exceed the EU Maximum limit levels. In the survey of Shenzhen (China), three samples exceeded the maximum limit levels and they were pork, beef and egg, respectively (Zhang et al., 2008). Sources of contamination are difficult to assess due to the random sampling and circulation of food in the market place.

The mean concentration of each food group was found obviously lower than Europe monitoring report in 2010 (EFSA, 2010), Belgian in 2010 (Windal et al., 2010), Australia in 2013 (Rauscher-Gabernig et al., 2013), Malaysia in 2014 (Leong et al., 2014), French in 2012 (Sirot et al., 2012) and Valencia (Spain) survey in 2011 (Marin et al., 2011). The results were consistent with the study which involved 152 samples in nineteen provinces in China in 2011, except cereal (Zhang et al., 2015). Cereal presented the lowest PCDD/Fs concentrations in this study due to the low fat content, weak bioaccumulation and bio-amplification, with an average of 0.012 and 0.117 pg TEQ/g w.w. in lower and upper bound values, respectively, but it was almost ten times higher than that in China survey in 2011 (Zhang et al., 2015) and in Shenzhen (China) report (Zhang et al., 2008). This difference is probably due to different sampling strategies (coverage of food type and region) and incomplete data. Therefore, we need to be more comprehensive data to evaluate regional PCDD/Fs levels in various foodstuff.

As one of the main contributors of dietary exposure, the concentrations of PCDD/Fs in fish and seafood were similar to, or lower than those in previous reports in Spain (Marin et al., 2011), Japan (Mato et al., 2007) and Taiwan (Hsu et al., 2007). The concentrations in shrimp, squid and shellfish were found to be much lower than in fish, ranging from 0.070 to 0.300 pg TEQ/g w.w. This result agrees with the report from Catalan, Spain (Bocio et al., 2007).

For milk and dairy products, TEQ concentrations of PCDD/Fs ranged from 0.023 to 2.840 pg TEQ/g fat, with an average of 0.411 pg TEQ/g fat. Similar concentrations were reported in previous studies in other countries, such as Spain (0.540 pg TEQWHO98/g fat) (Marin et al., 2011) and Australia (0.600 pg TEQWHO98/g fat) (Rauscher-Gabernig et al., 2013), and were lower than in Belgium (0.750 pg TEQWHO98/g fat) (Windal et al., 2010) and Taiwan (0.750 pg TEQWHO98/g fat) (Hsu et al., 2007). In infant formulae (N = 32), the concentration ranged from 0.036 to 0.480 pg TEQ/g fat, with average of 0.249 pg TEQ/g fat. This level was clearly lower than the infant formulae available on the EU market (Pandelova et al., 2010).

TEQ concentrations of PCDD/Fs in beef and mutton ranged from 0.009 to 2.730 pg TEQ/g fat, with average of 0.483 pg TEQ/g fat. But, the average concentration (0.340 pg TEQ/g fat) in mutton was lower than in beef (0.545 pg TEQ/g fat). This result was consistent with previous reports in Taiwan and Shenzhen (Hsu et al., 2007; Zhang et al., 2008), and different from the...
Belgian report, in which the concentration in beef was two times that in sheep.

At present, more people are consuming supplements, such as vitamins. We also investigated these dietary supplements. In this group (N = 112), concentrations ranged from 0.032 to 1.550 pg TEQ/g w.w., with an average of 0.213 pg TEQ/g w.w. Because many supplements were oil soluble products, such as DHA, vitamin A, D, E, K and coenzyme Q, relatively high PCDD/F levels were found in this foodstuff. It was suggested to eat less this kind of supplements or eat according to the need.

2.2. Homologue and congener profiles of PCDD/Fs in food

Table 1 shows that the congeners with the highest frequency of detection were OCDD for PCDDs, and 1,2,3,4,6,7,8-HpCDF for PCDFs. The congeners with the lowest frequency of detection were 1,2,3,7,8,9-HxCDF and 1,2,3,4,7,8,9-HpCDD, for which 94% and 89% of all the analysed samples were below the limit of detection (LOD), respectively. In Valencia, Spain, the most highly detected congener was OCDD and 2,3,7,8-TCDF, while the less detected congeners were 2,3,7,8-TCDD and 1,2,3,4,7,8,9-HpCDF (Marin et al., 2011). In Europe monitoring report, the most highly detection compounds were OCDD and 2,3,4,7,8-PeCDF, and the less detected congeners were 1,2,3,4,7,8-HxCDF and 1,2,3,4,6,7,8-HpCDF (EFSA, 2010). It was found that OCDD was one of the highest frequency detection compounds in different countries’ reports. In 2006, WHO had changed the TEF of OCDD from 0.0001 to 0.0003 (Van den Berg et al., 2006) and it is also widely detected in environmental samples (Li et al., 2010). So we have to slightly concern about the source and concentration about this PCDD congener. Meanwhile, this result indicated that the main PCDD/F pollutants in foodstuff in every country were different and this may be caused by different contamination sources.

The contribution of the 17 PCDD/F congeners in each food group was also studied and the result is presented in Fig. 2. Overall, OCDD, 2,3,7,8-TCDF, and 2,3,4,7,8-PeCDF were definitely the predominant congeners for foodstuffs and the average contributions to the total PCDD/F contents in all analysed samples were 39%, 9%, 8%, respectively. The results were in agreement with previous studies in China (Chen et al., 2008; Zhang et al., 2007, 2008) and other reports in Europe (EFSA, 2010; Marin et al., 2011; Senthil Kumar et al., 2001). The other five PCDDs (except OCDD) contributed less than 15% to the PCDD/F concentration.

In beef and mutton, OCDD (32%), 1,2,3,4,6,7,8-HpCDF (12%), 1,2,3,4,7,8-HxCDD (12%), and 2,3,4,7,8-PeCDF (12%) were the dominant congeners, together contributing 76% of the total PCDD/Fs. These main contributors were also found in milk and dairy products and a little difference from Chile survey, where the predominant congeners in dairy products were OCDD, 2,3,4,7,8-PeCDF, and 1,2,3,4,6,7,8-HpCDF and the contribution was 26%, 8.8% and 8.4%, respectively (Pizarro-Aranguiz et al., 2015). In fish and seafood, OCDD, 2,3,4,7,8-PeCDF, and 2,3,7,8-TCDF were the important compounds of PCDD/Fs, whose contribution were 41%, 12%, and 23%, respectively. These congeners were also predominant in marine oil. The results in fish and marine oil were consistent with report in Valencia, Spain (Marin et al., 2011) and Zhoushan Fishery, China (Wang et al., 2015). These two sets of data are correspondence with the biological transportation properties of dioxins.

The predominant congeners in fats and oils of animal origin were different from those in vegetable oils and fats. For animal origin, 1,2,3,4,6,7,8-HpCDF, OCDD, 1,2,3,4,7,8-HpCDF and 1,2,3,4,7,8-HxCDF were important contributors, while OCDD accounted for 85% of the total PCDD/Fs in vegetable oil and fat. Since the dominators in vegetable oil and fat have lower TEF value than those in animal oil and fat, the vegetable oil and fat samples showed much lower TEQ concentrations.

When considering toxic equivalents, the overall main contributor was 2,3,4,7,8-PeCDF, contributing an average of 32% to the total PCDD/F TEQ (Fig. 3), followed by 1,2,3,7,8-PeCDD (20%) and 2,3,7,8-TCDF (11%). This observation was consistent with many other previous reports (Loutfy et al., 2006; Olanca et al., 2014; Zhang et al., 2008). The high contribution of these congeners reflected their high TEF values (Marin et al., 2011). Although the TEF values for
PCDDs were higher, the contribution of PCDFs was more significant than PCDDs in all groups.

2.3. Foods contributing to daily intake of PCDD/Fs

In this part, when the congener's concentration was under its LOD, the value was assumed to be half of LOD. The average daily intakes of PCDD/Fs by the Chinese population are illustrated in Table 2. The daily intake of PCDD/Fs for rural populations (39.379 pg TEQ/day) was higher than urban populations (30.811 pg TEQ/day). The exposure of per person was 0.656 and 0.514 pg TEQ/kg b.w./day, respectively. This result was lower or at the same level, compared to the reports for Spain (Bocio and Domingo, 2005), Belgium (Focant et al., 2002) and Chile (Pizarro-Aranguiz et al., 2015).

The concentration of PCDD/Fs in cereal was low, but it was the major food for most Chinese, so cereal contributed the most to the total exposure. The proportion of its contribution was 81.4% for rural dietary daily intake and 47.7% for urban daily intake (Table 2). This result was different from previous reports in China (Zhang et al., 2008, 2013, 2015). Since the concentrations of each food group were similar with previous studies in China, the main difference is from food consumption weight. We used general food consumption level to assess exposure. In other studies, more regional and targeted data was applied, such as, for fish and seafood, in our study, the consumption was 14.1 and 39.2 g for rural and urban populations, and the contribution to daily intake was 4% and 16%, respectively, but in Shenzhen, the consumption was 109.1 g, and contribution was 44% (Zhang et al., 2008). The results from Shenzhen were in accordance with other countries, such as French (Sirot et al., 2012), Belgium (Focant et al., 2002), Spain (Marin et al., 2011), Malaysia (Leong et al., 2014) and Japan (Tsutsumi et al., 2001). In Australia, the main contributor was cheese and butter (Rauscher-Gabernig et al., 2013), but milk and dairy products only contributed 2.2% for daily intake.

### Table 2 – Estimated values of daily dietary intake of PCDD/Fs for Chinese people.

<table>
<thead>
<tr>
<th>Food group</th>
<th>TEQ conc. a (pg TEQ/g)</th>
<th>Rural CW b (g/day)</th>
<th>Rural Daily intake (pg TEQ/day)</th>
<th>Rural Daily intake (pg TEQ/kg b.w./day)</th>
<th>Percentage of contribution (%)</th>
<th>Urban CW b (g/day)</th>
<th>Urban Daily intake (pg TEQ/day)</th>
<th>Urban Daily intake (pg TEQ/Kg b.w./day)</th>
<th>Percentage of contribution (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beef and mutton</td>
<td>0.08</td>
<td>3.92</td>
<td>0.33</td>
<td>0.01</td>
<td>0.83</td>
<td>10.36</td>
<td>0.87</td>
<td>0.01</td>
<td>2.83</td>
</tr>
<tr>
<td>Chicken and duck</td>
<td>0.04</td>
<td>11.42</td>
<td>0.51</td>
<td>0.01</td>
<td>1.28</td>
<td>27.97</td>
<td>1.24</td>
<td>0.02</td>
<td>4.05</td>
</tr>
<tr>
<td>Pork</td>
<td>0.03</td>
<td>39.45</td>
<td>1.28</td>
<td>0.02</td>
<td>3.22</td>
<td>56.79</td>
<td>1.84</td>
<td>0.03</td>
<td>6.01</td>
</tr>
<tr>
<td>Fish and seafood</td>
<td>0.12</td>
<td>14.11</td>
<td>1.72</td>
<td>0.03</td>
<td>4.34</td>
<td>39.18</td>
<td>4.77</td>
<td>0.08</td>
<td>15.59</td>
</tr>
<tr>
<td>Milk and dairy products</td>
<td>0.09</td>
<td>9.73</td>
<td>0.85</td>
<td>0.01</td>
<td>2.15</td>
<td>49.59</td>
<td>4.35</td>
<td>0.07</td>
<td>14.21</td>
</tr>
<tr>
<td>Vegetable oil</td>
<td>0.13</td>
<td>15.12</td>
<td>1.97</td>
<td>0.03</td>
<td>4.96</td>
<td>24.2</td>
<td>3.15</td>
<td>0.05</td>
<td>10.28</td>
</tr>
<tr>
<td>Animal oil</td>
<td>0.23</td>
<td>2.16</td>
<td>0.50</td>
<td>0.008</td>
<td>1.25</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Cereal</td>
<td>0.07</td>
<td>495.89</td>
<td>32.24</td>
<td>0.54</td>
<td>81.40</td>
<td>224.66</td>
<td>14.60</td>
<td>0.24</td>
<td>47.73</td>
</tr>
<tr>
<td>Total</td>
<td>–</td>
<td>–</td>
<td>39.38</td>
<td>0.66</td>
<td>–</td>
<td>30.81</td>
<td>0.51</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

a TEQ concentrations of each food group, and it was calculated for TEQWHO and the non-detected congener concentrations were equal to 1/2 LOD.

b CW: consumption weight.

Fig. 3 – Contribution of PCDD/F congeners to the total TEQ concentration of all analysed food samples.
rural and 14.2% for urban populations in our study, and were also much lower than in Tarragona, Spain (20.1%) (Bocio and Domingo, 2005).

In this present study, meat and meat products, including beef, mutton, chicken, duck and pork, had little contribution to the total dietary exposure and the contribution was 5.3% for rural and 12.9% for urban populations, while in Tarragona, Spain, this group was 8.2% (Bocio and Domingo, 2005) and in Australia, it was 12% for women and 18% for men (Rauscher-Gabernig et al., 2013). The total contribution of vegetable oil and animal oil was 6.2% and 10.3% for rural and urban populations, respectively, and was lower than Tarragona, Spain (Bocio and Domingo, 2005).

The important finding of this study is that the daily intake of PCDD/Fs by the Chinese was found to be lower than expected and lower than some of the most developed countries. This can be attributed to the fact that food of animal origin represents a less significant part of the diet in China, especially for rural populations (Zhou et al., 2012). High levels of PCDD/Fs found in marine oil and fish and seafood point to the existing sources of environmental contamination. Therefore, it is important to proactively monitor and clean up the environment, especially water sources and soil, to avoid adverse effects on human health as consumption of animal products increases in China.

3. Conclusions

The results in this study showed that there is little health risk from PCDD/Fs in food in China. Moreover, the overall daily intake in China was lower than in many developed countries. These results show general exposures of the population in China. Due to the limited samples and differences in eating habits in China, a further study should be carried out to determine PCDD/Fs as well as dl-PCB contamination levels in food and to obtain detailed information of human exposures in different areas.

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