Science of the Total Environment 615 (2018) 412–421

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# Levels, congener profiles, and dietary intake assessment of polychlorinated dibenzo-*p*-dioxins/dibenzofurans and dioxin-like polychlorinated biphenyls in beef, freshwater fish, and pork marketed in Guangdong Province, China

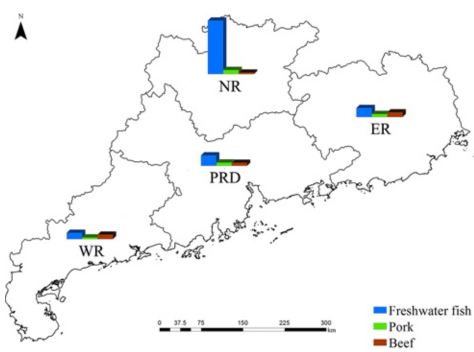
Wei-Liang Wu [a,b,c,1], Xiao-Ling Deng [a,1], Shao-Jun Zhou [a], Hui Liang [a], Xing-Fen Yang [a], Jian Wen [a], Xiao-Ming Li [c,d], Chao-Zheng Zhang [a], Yong-Hui Zhang [a,*], Fei Zou [b,**]

[a] Institute of Nutrition and Food Safety, Guangdong Provincial Center for Disease Control and Prevention, Guangzhou 511430, PR China
[b] School of Public Health, Southern Medical University, Guangzhou 510515, PR China
[c] Guangzhou Punuo Environmental Testing and Technology Service Co. Ltd., Guangzhou 510530, PR China
[d] National Testing Center for Food Quality Supervision (Guangdong), Guangdong Testing Institute of Product Quality Supervision, Foshan 528300, PR China

## HIGHLIGHTS

- The general levels of PCDD/Fs and DL-PCBs in three food groups marketed around Guangdong Province were first reported.
- Different congener profiles were found in three food groups from four different regions.
- Freshwater fish from north region of Guangdong Province were more prone to accumulate PCDD/Fs rather than DL-PCBs.
- Dietary exposure of PCDD/Fs and DL-PCBs had a low contribution to PTMI.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

*Article history:*
Received 28 July 2017
Received in revised form 25 September 2017
Accepted 25 September 2017
Available online 4 October 2017

Editor: Adrian Covaci

*Keywords:*
PCDD/F
DL-PCB

## ABSTRACT

Persistent organic pollutants such as polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (DL-PCBs) consisting of non-*ortho* and mono-*ortho* PCBs are suggested to be very hazardous and have adverse effects on human health. However, their levels and congener profiles in retail foods marketed in Guangdong Province of China have not been elucidated thus far. Thus, in this study, 226 individual samples of beef, freshwater fish, and pork marketed across four regions of Guangdong Province were randomly collected during 2013–2015 to determine their levels of PCDD/Fs and DL-PCBs. The results showed that the total toxic equivalency quantities (TEQs) of most samples were below the maximum limits except for the 26 samples collected from the vicinities of pollution areas. The median total TEQs of these three categories were 0.174, 0.488, and 0.113 pg TEQ/g fw, respectively, which indicated that the contamination status of the studied foods was not serious. For congener profiles, significantly different patterns were observed in three food groups, but with the same major

*Abbreviation:* PCDDs, polychlorinated dibenzo-*p*-dioxins; PCDFs, polychlorinated dibenzofurans; DL-PCBs, dioxin-like polychlorinated biphenyls; TEF, toxic equivalency factors; TEQ, toxic equivalency quantity; POPs, persistent organic pollutants; JECFA, Joint FAO/WHO Expert Committee on Food Additives; TDS, total dietary survey; HA, *n*-hexane; DCM, dichloromethane; HRGC/HRMS, high-resolution gas chromatography-high resolution mass spectrometer; SIM, selected ion monitoring; LOD, limit of detection.

* Correspondence to: Y.-H. Zhang, Guangdong Provincial Center for Disease Control and Prevention, 511430 Guangzhou, PR China. ** Corresponding author.
*E-mail addresses:* zyh@cdcp.org.cn (Y.-H. Zhang), zfei@fimmu.com (F. Zou).
[1] Co first authors who contributed equally to this work.

https://doi.org/10.1016/j.scitotenv.2017.09.273

Foodstuff
Congener profile
Dietary intake

TEQ contributors being 2,3,4,7,8-PeCDF in beef, freshwater fish, and pork. Regional differences of congener profiles in each food group were also found in this study, which might be attributed to the regionally different distributions of PCDD/Fs and DL-PCBs in environment media.
The dietary exposures of four population subgroups (girls, boys, male adults, and female adults) to PCDD/Fs and DL-PCBs via three food groups were estimated to assessed the potential risks. They were all lower than the provisional tolerable monthly intake (PTMI, 70 pg TEQ/kg bw/month) established by Joint FAO/WHO Expert Committee on Food Additive. In these food categories, the exposure to PCDD/Fs and DL-PCBs via freshwater fish was the highest one, which accounted for about 20% of PTMI, indicating that it was the major route to expose dioxin compounds.


## 1. Introduction

Persistent organic pollutants (POPs) are toxic organic chemicals that are resistant to environmental degradation via mechanisms such as photolysis, chemical or biological actions (Eduljee, 2001). Owing to their unique characteristics including extensive release in environmental media, high environmental stability, and severe adverse effects on biota and human health, the emissions of POPs need to be reduced and their production prohibited (Chen et al., 2008; G. Zhang et al., 2007a; J.Q. Zhang et al., 2007b). Among POPs, three closely related families of polyhalogenated aromatic hydrocarbons referred to as dioxins, comprising polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (DL-PCBs), are regarded as the most hazardous ones and put into the initial list of Stockholm Convention. Thus, they have received great attention in the recent decades. PCDDs and PCDFs occur naturally and as the unintentional by-products of incomplete combustion and various industrial processes, in contrast, DL-PCBs mainly result from the wide production and a variety of commercial uses of PCBs before the applications have been banned by most countries since 1970s. These environmental toxicants can bioaccumulate easily through the food chain and ultimately show similar spectrum of adverse biological effects and responses in wild life and humans that include dermal toxicity, reproductive and developmental effects, neurological effects, immunomodulatory, and carcinogenic effects (Chan and Wong, 2013; Costopoulou et al., 2016; Marin et al., 2011; Zhang et al., 2012).

In humans, exposure routes of PCDD/Fs and DL-PCBs from contaminated matters include ingestion, inhalation, and dermal contact (Chan et al., 2013). Dietary intake is regarded as the main pathway of exposure to dioxins, because food ingestion is known to account for >90% of total human exposure (Bocio et al., 2007; Fernandes et al., 2010; Grassi et al., 2010; Li et al., 2007; Wang et al., 2015). Due to their ubiquitous nature, persistence, and lipophilicity, dioxins can easily accumulate at high levels in food of animal origin with high fat content, especially meat and dairy products (Chan et al., 2013). Thus, in many countries and regions, the contamination levels of PCDD/Fs and DL-PCBs in foodstuffs have been extensively investigated, particularly in livestock and poultry products (Fernandes et al., 2010; Hoogenboom et al., 2016; Huwe and Larsen, 2005; Kim et al., 2007; Squadrone et al., 2015), dairy products (Durand et al., 2008; Esposito et al., 2009; Schmid et al., 2003), and aquatic products (Han et al., 2007; Moon and Ok, 2006; Munschy et al., 2008; Sakuraia et al., 2000; C.F. Shen et al., 2009a; H.T. Shen et al., 2009b). These studies showed that the toxic equivalency quantity (TEQ) levels, which are used to estimate the harmful effects of these hazardous chemicals, of certain animal-origin food exceeded the limits set by EU Regulation 1259/2011, indicating that intake of such highly contaminated food may pose hazardous effects for residents. In China, the standard of determination method for PCDD/Fs and DL-PCBs in foodstuffs was implemented in 2007 and revised in 2013, but the limits of PCDD/Fs and DL-PCBs still not set by the relevant department. Therefore, the limits for foodstuffs set by EU were recommended as reference in researches and risk assessments.

In order to estimate the dietary exposure of PCDD/Fs and DL-PCBs through food to reveal the extent of chronic intake, total dietary survey (TDS) has been performed in numerous developed and developing countries in recent years (De Mul et al., 2008; Rauscher-Gabernig et al., 2013; Sasamoto et al., 2006; Sirot et al., 2012; Taioli et al., 2005; Wang et al., 2009; Windal et al., 2010; Zhang et al., 2013; Zhang et al., 2015). The results from the latest Chinese TDS show that a relatively higher exposure is observed in some developed regions such as Zhejiang Province, Guangdong Province, and Shanghai City, suggesting that foods polluted by dioxins may cause by a high environmental burden and pose a threat to the health of local residents (Zhang et al., 2015). Therefore, it is necessary to conduct an in-depth survey to give researchers integral insights on the levels of PCDD/Fs and DL-PCBs in foods marketed in these Chinese developed regions.

The occurrences and concentrations of PCDD/Fs and DL-PCBs in food of animal and plant origin marketed in Shenzhen City, one of the developed cities of Guangdong Province, were determined recently by J.Q. Zhang et al. (2007b, 2008). However, their general levels in retail foods available in Guangdong Province have not been reported thus far. Accordingly, one of the objectives of this study was to carry out the first investigation with wide coverage on the general levels and congener profiles of PCDD/Fs and DL-PCBs in three main categories of food including beef, freshwater fish, and pork, sampled from Guangdong Province. Further, we estimated the dietary intakes of PCDD/Fs and DL-PCBs by employing deterministic assessment and evaluated the risk to the health of general and special populations.

## 2. Materials and methods

### 2.1. Chemical reagents and standards

The column packing materials including Silica gel 60 (63–200 μm), Bio-Beads™ S-X3 support (38–75 μm), and Florisil (149–250 μm, pesticide grade) used for column purification were purchased from Merck (Darmstadt, Germany), Bio-Rad Laboratories (Hercules, CA, USA), and Supelco (Bellefonte, PA, USA), respectively. Organic solvents of pesticide grade applied for extraction, elution, and dissolution including *n*-hexane (HA), acetonitrile, acetone, *n*-nonane were obtained from Honeywell Burdick & Jackson (Ulsan, Korea), and dichloromethane (DCM) from Merck Chemicals Co., Ltd. (Shanghai, China). Other chemicals used in the process of pretreatment are anhydrous sodium sulfate, sodium hydroxide, and sulfuric acid. The quantification standard solutions, internal standards, spiking solutions, and injection standards of PCDD/Fs and DL-PCBs were all purchased from Cambridge Isotope Laboratories Co. (Andover, MA, USA).

### 2.2. Sample collection and preparation

#### 2.2.1. Sampling

A total of 226 individual food samples were purchased from local food markets and supermarkets during 2013–2015. These samples were classified into three categories of beef ($n = 71$), freshwater fish ($n = 76$), and pork ($n = 79$). As part of the annual national food contaminants survey in China, 17 sampling sites covering most of the 22 prefectures of Guangdong Province were investigated according to population and economic status, which respectively located in Eastern region of Guangdong Province (ER, $n = 50$), Northern region of Guangdong Province (NR, $n = 59$), Pearl River Delta (PRD, $n = 80$), and Western region of Guangdong Province (WR, $n = 39$). The detail information regarding sampling locations in Guangdong Province and sample sizes in each surveillance site are shown in Supplementary materials (Fig. S1 and Table S1), respectively. Each individual sample of three food groups was sealed into a polyethylene bag after wrapped by aluminum foil and frozen at −40 °C. All the samples were shipped to our joint laboratories for sample preparation and analysis.

#### 2.2.2. Sample preparation

Freshwater fish, including grass carp (*Ctenopharyngodon idellus*), bighead carp (*Aristichthys nobilis*), crucian carp (*Carassius aumtus*), tilapia (*Oreochromis* spp.), catfish (*Silurus asotus*), Chinese perch (*Siniperca chuatsi*), and bass (*Perca fluviatilis*), were prepared as skin-on fillet from which scales, bones, and viscera were removed. Inedible parts from pork, i.e. skinless belly and beef, i.e. beef from rump were removed. Then, each individual sample was washed with deionized water to remove impurities and patted dry with paper towels. Finally, the sample was cut into small pieces, homogenized to form an emulsion with a mechanical blender, and freeze-dried. The sample after lyophilization was preceded with sample purification and determination.

### 2.3. Sample purification and determination

#### 2.3.1. Sample purification

The levels of PCDD/Fs and DL-PCBs in three catalogs of food specimens were determined according to the US EPA method 1613 and US EPA method 1668a with a minor modification. A Soxhlet apparatus was employed for sample extraction. After being spiked with a mixture of $^{13}C_{12}$-labeled PCDD/Fs and DL-PCBs extraction standards in the pretreatment process, the completely freeze-dried food sample was placed into the reflux unit for 16-h extraction using 250 mL HA/DCM mixed-solution (1:1, v/v) as a solvent. The solvent was removed using a rotary evaporator and the lipid content was estimated by a gravimetric method. Then, the lipid fraction was purified to remove the interferences caused by polar substances. Briefly, the fraction was re-dissolved into 100 mL HA and blended with 40 g acid-modified silica gel (40%, g/g) by a magnetic stirrer at ambient temperature for 1 h continuously. After the bulk lipids were removed, the eluate was concentrated and a multi-step cleanup was carried out with column chromatography. The extract was first loaded onto a multilayer silica gel column (from bottom to top: 2 cm anhydrous sodium sulfate, 1 g activated silica gel, 4 g 33% alkaline-modified silica gel, 1 g activated silica gel, 8 g acid-modified silica gel, 2 g activated silica gel, 2 cm anhydrous sodium sulfate) and then eluted with 100 mL HA/DCM mixture (97:3, v/v). The solvent was removed by rotary evaporation and the eluent transferred onto a gel permeation column (Bio-Beads S-X3) for further purification. The target chemicals were then eluted with 50 mL HA/DCM mixture (1:1, v/v) after using 70 mL HA/DCM solvent (1:1, v/v) to clean up the column. Finally, the elution fraction was transferred to a diatomite column for the last cleanup process. DL-PCBs were first eluted with 15 mL HA/DCM mixed-solvent (95:5, v/v) and the fraction of PCDD/Fs was eluted with 40 mL DCM. These two purified eluted fractions containing PCDD/Fs and DL-PCBs were concentrated and transferred to conical vials containing 10 μL of nonane. Before analysis, two concentrates were dried under a gentle nitrogen stream followed by spiking with the corresponding standards.

#### 2.3.2. Analytical method

Quantitative analyses of PCDD/Fs and DL-PCBs were performed using isotope dilution high-resolution gas chromatography-high resolution mass spectrometer (HRGC/HRMS) operating in selected ion monitoring (SIM) mode using an Agilent 7890B (Agilent Technologies, Santa Clara, CA, USA) equipped with a DB-5MS capillary column (60 m × 0.25 mm i.d. × 0.25 μm) and coupled with an HR Auto Spec Premier mass spectrometer (Waters, Manchester, UK). Carrier gas helium was supplied at a constant pressure of 185 kPa. Sample (1 μL) was injected into the injector at 280 °C in the splitless mode. For the PCDD/Fs determination, the oven temperature was programmed as follows: 140 °C for 2 min, then from 140 °C to 220 °C at 8 °C/min, after that from 220 °C to 260 °C at 1.4 °C/min, the last procedure was from 260 °C to 310 °C at 4 °C/min and held at 310 °C for 3 min. For DL-PCBs, the temperature program was 90 °C for 2 min, then 90 °C to 220 °C at 13 °C/min, held for 7 min, after that from 220 °C to 260 °C at 1.4 °C/min, the last procedure was from 260 °C to 300 °C at 5 °C/min. The MS was operated over 10,000 resolutions (10% valley definition) with EI source at 35 V electron energy and 650 μA trap current. The temperatures of transfer line, ion source, and interface were 270 °C, 260 °C and 280 °C, respectively.

In this study, isotope dilution method was used for the quantification of PCDD/F and DL-PCB homologues designated by WHO, and the TEQs of the analyzed PCDD/Fs and DL-PCBs in this study were calculated using the toxic equivalency factors (TEF) 1998 proposed by WHO in 1998 (Van den Berg et al., 1998). The TEQs using TEF 2005 (Van den Berg et al., 2006) are summarized in Tables S2–S4 for reference and comparison. For the total TEQ level calculation, when the analyzed concentrations of congeners were below the limit of detection (LOD), i.e. not detected (N.D.), the values were assumed to be equal to half of LOD in this study for dietary exposure assessment. Additionally, the measurement uncertainties of the analytical method, including total uncertainty and expanded uncertainty, were listed in Table S5.

### 2.4. Quality control and quality assurance

Quality control and quality assurance protocols were set up to ensure the accuracy and precision of identification. The protocols included: (1) a procedural blank and a quantitative control sample for each batch of eight samples; (2) a certificated reference of fish tissue fortified with PCDD/Fs and DL-PCBs (WMF-01, Wellington Laboratory Inc., Guelph, ON, Canada) was used for method verification and quality control; (3) the laboratory testing ability for measurement of PCDD/Fs and DL-PCBs in foodstuff was validated in inter-laboratory trials organized by Norwegian Institute of Public Health. The recovery rates of the labeled $^{13}C_{12}$ surrogates homologues were between 40 and 120% for three food matrices, which conformed to the requirements of the US EPA methods. None of the surrogate recoveries in this study were outside this range and about 75% of our recoveries were in the range of 40%–90%.

The LOD was calculated as three times the signal-to-noise ratio. For PCDD/Fs, LODs ranged from 0.000380 to 0.160 pg/g fresh weight, 'fw', and for DL-PCBs, from 0.00400 to 1.40 pg/g fw.

### 2.5. Dietary exposure assessment

To evaluate the potential risk from food intake, the monthly intakes of PCDD/Fs and DL-PCBs via beef, freshwater fish, and pork were determined by deterministic assessment using the concentration and consumption data to calculate the exposure level for local residents in Guangdong Province, China. Human exposure is generally expressed in terms of bodyweight (TEQ pg/kg bw). The consumption data and average body weights of children and adults were obtained from Nutrition and Health Survey of Guangdong Province 2012 (Zhang and Ma, 2016).

### 2.6. Statistical analysis

All statistical analyses were performed using the IBM SPSS Statistics 23 software package (IBM, Armonk, New York, U.S.A.). All tests were two-tailed, and a level of 0.05 was set for statistical significance.

## 3. Results and discussion

### 3.1. Levels of PCDD/Fs and DL-PCBs in three groups of food

TEF and TEQ have been established for evaluation of the harmful effects of PCDD/Fs and DL-PCBs on environmental matrices and biota

(Van den Berg et al., 1998; Van den Berg et al., 2006). The TEQs of PCDDs (seven congeners), PCDFs (10 congeners), and DL-PCBs (12 congeners) were calculated in each group of food samples sampled from different locations in Guangdong Province, and the median, average, and the range of TEQ levels are shown in Fig. 1 and Table S2, respectively. In summary, the median TEQ levels of PCDDs, PCDFs, and DL-PCBs as well as the total TEQs (PCDD/Fs + DL-PCBs) were significantly different ($p < 0.05$) among the three food groups. The highest median level of total TEQ (0.488 pg TEQ/g fw) was found in freshwater fish, followed by beef and pork with the median total TEQ levels of 0.174 and 0.113 pg TEQ/g fw, respectively. In this study, it is found that 11.50% samples, collected from the vicinities of different contaminated areas, had higher total TEQ levels than the maximum limits set by the EU in all three studied food categories, indicating that the environment might be contaminated by the dioxins release sources and finally caused food contamination. For the median TEQ levels of three homologues, PCDFs were higher than the other two homologues in all three groups of food matrices, which predominantly contributed to the total TEQs (Fig. 1). As shown in Table S2, the concentrations of PCDDs and total TEQs showed large variations in freshwater fish and pork, because these TEQ levels of the samples, collected from the vicinities of typical E-waste disassembling regions, were much higher than the other samples. The food matrices collected from the vicinities of different E-waste disassembling regions in China, including freshwater fish, chicken, and duck, also had higher TEQ levels that exceeded the limits (Chan et al., 2013; Chan and Wong, 2013; Shen et al., 2017; Song et al., 2011; G. Zhang et al., 2007a; J.Q. Zhang et al., 2007b). Additionally, an increase of 10–25% was observed in the average total TEQ levels of the two food categories (freshwater fish and pork) when the TEQ calculation was performed using TEF 2005 instead of TEF 1998, which was in contrast to the results of De Mul et al. (2008) who reported a decrease of 10–25% (Table S2). This is attributable mainly to the extremely high mass concentration levels of OCDD and OCDF congeners in the samples that exceeded the maximum limits.

### 3.1.1. Beef

In this survey, the median concentrations of total TEQ in beef from all four regions were below 0.200 pg TEQ/g fw. The results shown in Fig. 2 indicated that the samples from NR had a higher TEQ value than those from the other three regions did. However, the differences in the TEQs among the four regions were not significant ($p > 0.05$). Our results were similar to the median total TEQ level reported for Zhejiang, China (Shen et al., 2017), but a little higher than the result of Italy (Taioli et al., 2005) (Table 1). The average levels of total TEQ in comparison with the published results are shown in Table S6. The levels in beef marketed from Guangdong Province were higher than the corresponding levels of Taiwan (Chen et al., 2008; Hsu et al., 2007; Lee et al., 2016), Catalonia in Spain (Perelló et al., 2012) and Italy (Taioli et al., 2005). The composition patterns of total TEQ were similar among the samples of the four regions; PCDDs or PCDFs contributed the most, while DL-PCBs contributed <20% of the mean total TEQs (Table S4).

### 3.1.2. Freshwater fish

Although Guangdong Province is one of the coastal regions in China, freshwater fish is also one of the favorite foods of local residents. However, PCDD/Fs and DL-PCBs surveillance for freshwater fish was limited in comparison with that for marine fish (Chan et al., 2013; G. Zhang et al., 2007a; J.Q. Zhang et al., 2007b; Zhang et al., 2008). As shown in Fig. 2, the median TEQ levels of PCDDs and PCDFs, as well as total TEQ in freshwater fish from NR were significantly higher than those observed in PRD, ER and WR ($p < 0.05$), which accounted for the higher contamination in NR. The median total TEQ of the three regions (PRD, ER, and WR) in this survey were comparable to or higher than the values of common fish species including freshwater fish and marine fish from the retail markets of Shenzhen, Guangdong Province (G. Zhang et al., 2007a; J.Q. Zhang et al., 2007b; Zhang et al., 2008) and Zhejiang Province (Shen et al., 2017; Wang et al., 2015), and fish from Italy (Taioli et al., 2005), but lower than the TEQ level of marine fish reported for Valencia in Spain (Marin et al., 2011). However, the median total TEQ of NR samples was much higher than the surveillance results mentioned above (Table 1). The mean total TEQs of four regions in comparison with those of other nations and regions are enumerated in Table S6. The mean levels of total TEQ of ER and WR were close to or higher than the survey results of freshwater fish collected from Taiwan (Chen et al., 2008;) and marine fish sampled from Zhejiang Province, China (Wang et al., 2015), but slightly lower than the values of marine fish marketed in Taiwan (Hsu et al., 2007; Lee et al., 2016). In contrast, the levels of these two regions were lower than the findings in aquatic products reported for the Western nations (Costopoulou et al., 2016; Perelló et al., 2012; Rauscher-Gabernig et al., 2013; Sirot et al., 2012)

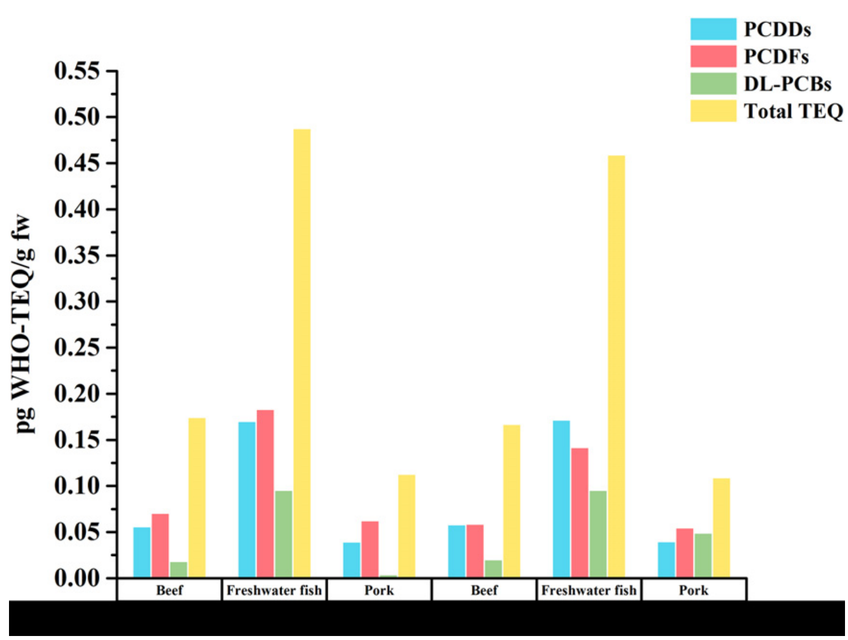



**Fig. 1.** The median levels of PCDDs, PCDFs, DL-PCBs, and total TEQ in the three food categories from four regions of Guangdong Province.

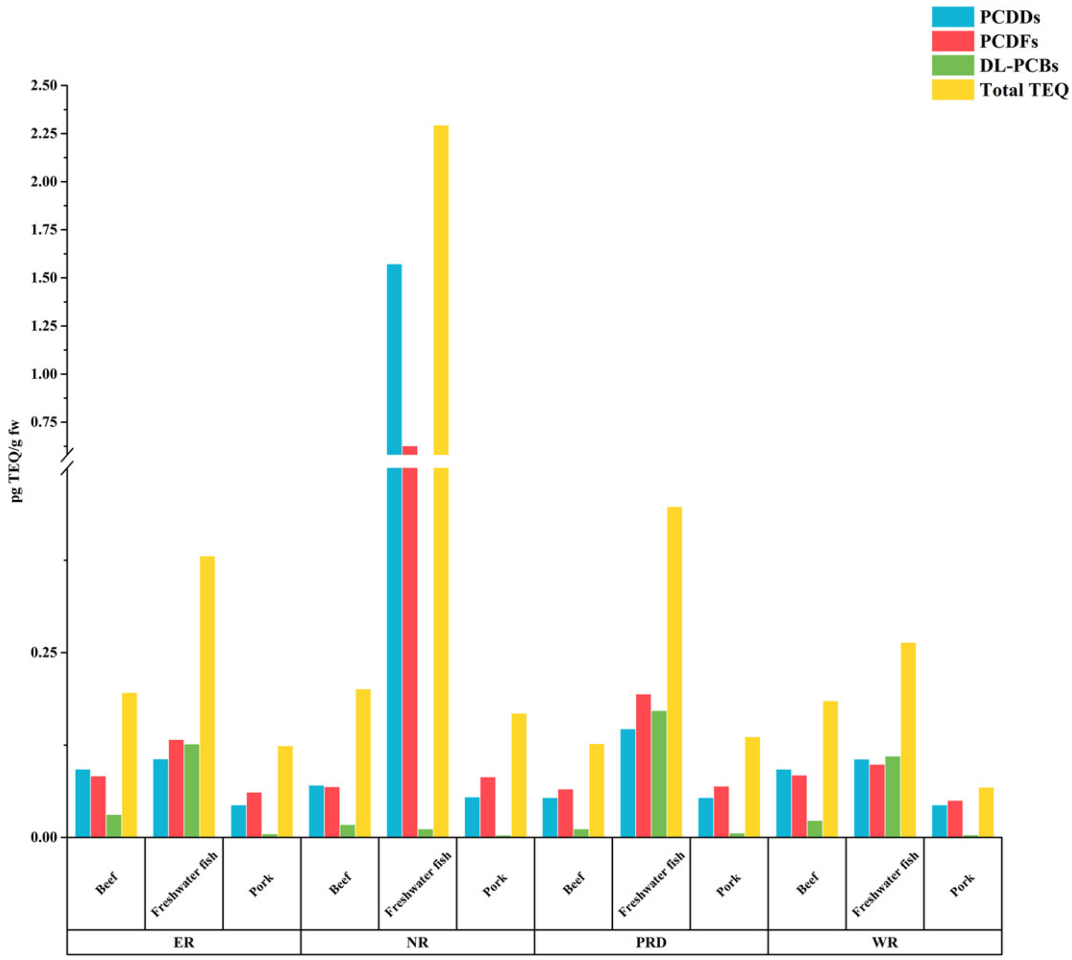



**Fig. 2.** The median levels of PCDDs, PCDFs, DL-PCBs, and total TEQ in the three food categories of Guangdong Province.

expect for Italy (Taioli et al., 2005). However, NR and PRD regions had even higher mean total TEQs (Table S6). It is also important to note that the freshwater samples from NR had an average total TEQ of 4.44 pg TEQ/g fw, which was comparable with or lower than those of the freshwater fish collected from Guiyu (4.64 pg TEQ/g fw) and Taizhou (10.87 pg TEQ/g fw), two of the most well-known E-waste dismantling areas in China (Chan et al., 2013; Song et al., 2011). It is indicated that a part of cultivated freshwater fish samples in NR may contaminated by the dioxin compounds released from the E-waste dismantling sites in this region, but the contamination status of NR was relatively better than that of Taizhou. Previous studies indicated that fish were more prone to accumulating DL-PCBs rather than PCDD/Fs (Costopoulou et al., 2016; Lee et al., 2016; Perelló et al., 2012; Rauscher-Gabernig et al., 2013; C.F. Shen et al., 2009a; H.T. Shen et al.,

**Table 1**
Comparison of the median total TEQ levels of three food groups with the results reported by the published literatures.

| Nation/Region | Food | Year | Median (pg TEQ/g fw) | Reference |
|---|---|---|---|---|
| Guangdong Province, China | Beef (beef from rump) | 2013–2015 | 0.174 | This study |
| Four regions of Guangdong Province | | | 0.196 (ER); 0.201 (NR); 0.128 (PRD); 0.185 (WR) | |
| Zhejiang Province, China | Beef | 2013–2014 | 0.13 | Shen et al., 2017 |
| Italy | Beef | 1999–2001 | 0.037 | Taioli et al., 2005 |
| Guangdong Province, China | Freshwater fish | 2014–2015 | 0.488 | This study |
| Four regions of Guangdong Province | | | 0.381 (ER); 2.29 (NR); 0.448 (PRD); 0.264 (WR) | |
| Shenzhen City, Guangdong Province, China | Fish (freshwater fish and marine fish) | – | 0.26 | G. Zhang et al., 2007a; J.Q. Zhang et al., 2007b |
| | | 2004–2006 | 0.34 | Zhang et al., 2008 |
| Zhoushan, Zhejiang Province, China | Marine fish | 2011 | 0.343 | Wang et al., 2015 |
| Zhejiang Province, China | Freshwater fish | 2014–2015 | 0.17 (herbivorous); 0.28 (omnivorous) | Shen et al., 2017 |
| Italy | Fish | 1999–2001 | 0.245 | Taioli et al., 2005 |
| Valencia, Spain | Marine fish | 2006–2008 | 1.21 | Marin et al., 2011 |
| Guangdong Province, China | Pork (skinless belly) | 2013–2015 | 0.113 | This study |
| Four regions of Guangdong Province | | | 0.124 (ER); 0.168 (NR); 0.136 (PRD); 0.0681(WR) | |
| Zhejiang Province, China | Pork | 2013–2014 | 0.080 | Shen et al., 2017 |
| Italy | Pork meat and derivates | 1999–2001 | 0.022 | Taioli et al., 2005 |

2009b; Sirot et al., 2012; Wang et al., 2015). However, PCDD/Fs TEQs and not DL-PCBs were found to accumulate in the freshwater fish from NR and PRD regions, which was similar with the observations of the survey carried out in Zhejiang Province, China and Ghana (Adu-Kumi et al., 2010; Shen et al., 2012; Shen et al., 2017). This interesting finding may be in relate with the environment matrices contaminated by higher concentration levels of PCDD/Fs than DL-PCBs that release from different processes of E-waste dismantling. Many published literatures pointed out that PCDD/Fs were the major contaminants other than DL-PCBs in soil and sediment samples from ambient regions of different E-waste dismantling sites located in China, Vietnam and Ghana (C.F. Shen et al., 2009a; H.T. Shen et al., 2009b; Suzuki et al., 2016; Tue et al., 2016; Wong et al., 2007).

#### 3.1.3. Pork

The survey also gave great concerns on the levels of PCDDs and DL-PCBs in pork due to the high consumption in China. The median concentration of total TEQ in pork samples from NR was higher than the corresponding ones of the samples from the other three surveillance regions, which were lower than 0.150 pg TEQ/g fw, but the differences were not significant ($p > 0.05$) (Fig. 2). The major contributors to total TEQ were PCDD/Fs, accounting for over 95% of total TEQ. The median total TEQs of NR, ER, and PRD samples were higher than the TEQs of pork collected from Zhejiang Province (Shen et al., 2017) and pork meat and derivates from Italy (Taioli et al., 2005), while the value for WR samples was comparable (Table 1). However, the average total TEQ values of four regions were higher than those reported by many surveys on market retail pork from many regions and countries, such as Taiwan (Chen et al., 2008; Hsu et al., 2007; Lee et al., 2016), Catalonia in Spain (Perelló et al., 2012), and Italy (Taioli et al., 2005) (Table S6).

### 3.2. Congener profiles of three categories of food samples

The congener patterns of PCDD/Fs and DL-PCBs in food matrices are mainly governed by a combination of feed contamination and the environmental pollution of the area where the food is cultivated and produced. The most predominant PCDD/F and DL-PCB congeners in beef were 2,3,4,7,8-PeCDF, 1,2,3,7,8-PeCDD, and PCB126, which contributed to 26.20%, 21.72%, and 12.77% of the total TEQ, respectively (Fig. 3). For freshwater fish, the congener pattern was not completely similar to that of beef, but had the same major contributors including 2,3,4,7,8-PeCDF, PCB126, and 1,2,3,7,8-PeCDD, accounting for 24.67%, 23.55%, and 20.22% of the total TEQ, respectively. For pork, the congener distribution was slightly different from those of the other two groups; the PCDD/F congeners with the highest TEQ levels were 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, and 1,2,3,7,8-PeCDD, accounting for 20.02%, 16.59%, and 15.52% of the total TEQ, respectively, while the DL-PCB congener in pork with the highest TEQ was PCB126, but it only accounted for 3.79% of the total TEQ.

#### 3.2.1. Beef

The congener-specific patterns of PCDD/Fs in beef from four regions had a similar profile and were characterized by the same dominant congeners (2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDD), whereas the congener profiles of DL-PCBs had regional characteristics with the same dominant congener (PCB126) but different distribution of other DL-PCBs. As shown in Fig. 4a and b, the main congeners that contributed to the total TEQs in beef from four regions were penta-CDD/Fs, i.e. 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF, which were similar with the findings of the published papers (Focant et al., 2002; Tsutsumi et al., 2001; Zhang et al., 2008). However, the congener profile of PRD samples was slightly different from those observed in the other three regions and 1,2,3,4,6,7,8-HpCDD was one of the dominant contributors (21.35%) to the total TEQ of the samples. This observation was not in line with the results reported by previous studies in which the dominant contributors to total TEQ in beef were 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF (Focant et al., 2002; Tsutsumi et al., 2001; Zhang et al., 2008). The difference may be caused by many reasons, such as different environment pollution sources of PCDD/Fs in different areas, different feeds used in the cultivation process, and the influence of bio-accumulation.

#### 3.2.2. Freshwater fish

The congener profiles of freshwater fish were more complicated in comparison with those of beef (Fig. 4c and d). Firstly, the isomeric patterns of the samples from PRD and ER had analogous PCDD/Fs and DL-PCBs composition, however, the TEQ levels of most congeners were higher in the samples of PRD. 2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDD were the most prominent congeners in PCDD/Fs homologues, contributing to about 21.18% and 12.82% (PRD), and 19.41% and 22.06% (ER) of the total TEQs, which was in agreement with the results reported by J.Q. Zhang et al. (2007b, 2008). However, the congener distribution of the freshwater fish from WR was different from those of PRD and ER samples, namely, 2,3,7,8-TCDD, 2,3,7,8-TCDF, and 2,3,4,6,7,8-HxCDF were also the major contributors to the TEQ, and this might result from the regionally different distributions of PCDD/Fs in environment media. Comparing the contributions of PCDD/Fs to total TEQs, the freshwater fish samples from these three regions were more likely to accumulate the coplanar congener PCB126, which has the highest toxicity among DL-PCBs, accounting for over one third of the total TEQs. Although the samples from PRD had the same major congeners as those from ER and WR, they primarily accumulated PCB126 (0.1540 pg TEQ/g fw), accounting for 34.88% of the total TEQ. This finding is in accordance with the relevant reports on marine fish and seafood (Costopoulou et al., 2016; Lee et al., 2016; Sasamoto et al., 2006; Tsutsumi et al., 2001). The highest median total TEQ of freshwater fish was for NR samples, in which the congener distributions of PCDD/Fs and DL-PCBs were quite different from those of the other three sampling sites; the more important congeners in this area were the highly-chlorinated PCDD/Fs including 1,2,3,4,6,7,8-HpCDD/F and 1,2,3,6,7,8-HxCDD. The TEQ level of DL-PCBs in the freshwater fish from NR was the lowest; the congeners were not considered important because of their slight contribution to the total TEQ in comparison with PCDD/Fs. The findings for NR samples and those reported by recent studies (Focant et al., 2002; Shen et al., 2017; Song et al., 2011) contradict those of previous studies, which showed that fish are more prone to accumulate DL-PCBs rather than PCDD/Fs (Costopoulou et al., 2016; Lee et al., 2016; Perelló et al., 2012; Rauscher-Gabernig et al., 2013; C.F. Shen et al., 2009a; H.T. Shen et al., 2009b; Sirot et al., 2012; Wang et al., 2015).

#### 3.2.3. Pork

The pork samples from the four different regions significantly accumulated PCDD/Fs rather than DL-PCBs, which is similar with the results reported by Shen et al. (2017). However, the congener patterns of PCDD/Fs were different in each region, while analogous compositions of DL-PCBs were reported for the pork from four survey regions. The results shown in Fig. 4e and f indicated that pork samples from NR were polluted with 1,2,3,4,7,8-HxCDF, which accounted for about 19% of the total TEQ and was different from those reported in the literatures that described 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF to be the predominant congeners in pork (Focant et al., 2002; Tsutsumi et al., 2001; Zhang et al., 2008). The specimens from WR were also characterized by the significant contribution of 1,2,3,4,7,8-HxCDF to total TEQ, whereas the TEQ levels of 1,2,3,4,6,7,8-HxCDF in the WR samples were lower than those reported for NR samples. In contrast, the distributions of PCDD/Fs and DL-PCBs in the samples of PRD and ER were distinguished from those in the other two regions with 2,3,4,7,8-PeCDF being the main congener.

### 3.3. Dietary exposure assessment

Table 2 summarizes the estimated exposures of PCDD/Fs and DL-PCBs via beef, freshwater fish, and pork. Because no specific

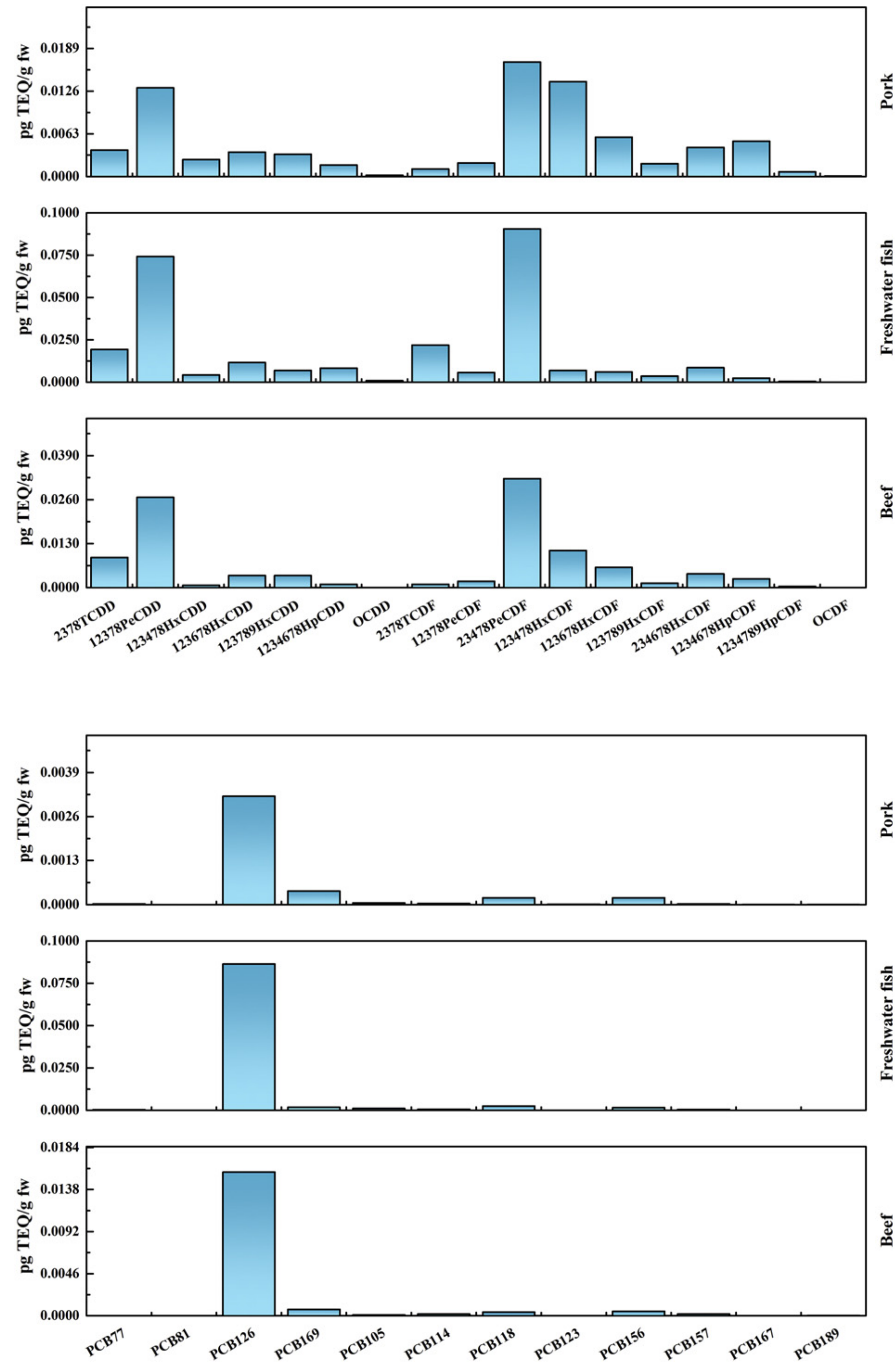



**Fig. 3.** Congener profiles of PCDDs/Fs and DL-PCBs in the three food categories of Guangdong Province.

consumption data for freshwater fish were obtained from the dietary survey, we assumed that the consumption of freshwater fish was equal to the aquatic product (Zhang and Ma, 2016). Risk assessment was carried out by comparing the results with the provisional tolerable monthly intake (PTMI, 70 pg TEQ/kg bw/month) established by the Joint FAO/WHO Expert Committee on Food Additives (JECFA, 2001).

For the children in Guangdong Province, the total exposure to PCDD/Fs and DL-PCBs from three food categories was much lower than the PTMI set by the JECFA, indicating that the consumption of these foods marketed in the Guangdong Province may pose a low threat to children. The dietary exposure for each food group is summarized in Table 2. The results showed that the dietary intake of girls was more than that of boys because of the higher consumption of freshwater fish and the lower body weight. The dietary exposure to PCDD/Fs and DL-PCBs from freshwater fish accounted for about 15%–22% of the PTMI, and was the major contributor in the three food groups. By contrast, the exposure from beef contributed to only about 1% of the PTMI due to the low consumption amount and relatively low concentration of dioxin compounds.

For the adults in Guangdong Province, the total exposure to PCDD/Fs and DL-PCBs via the three food categories was also lower than the PTMI. The dietary exposures for the adults are shown in Table 2. The contributions of the exposures of male and female adults via freshwater fish to PTMI were 20.41% and 20.71%, respectively, which were close to those

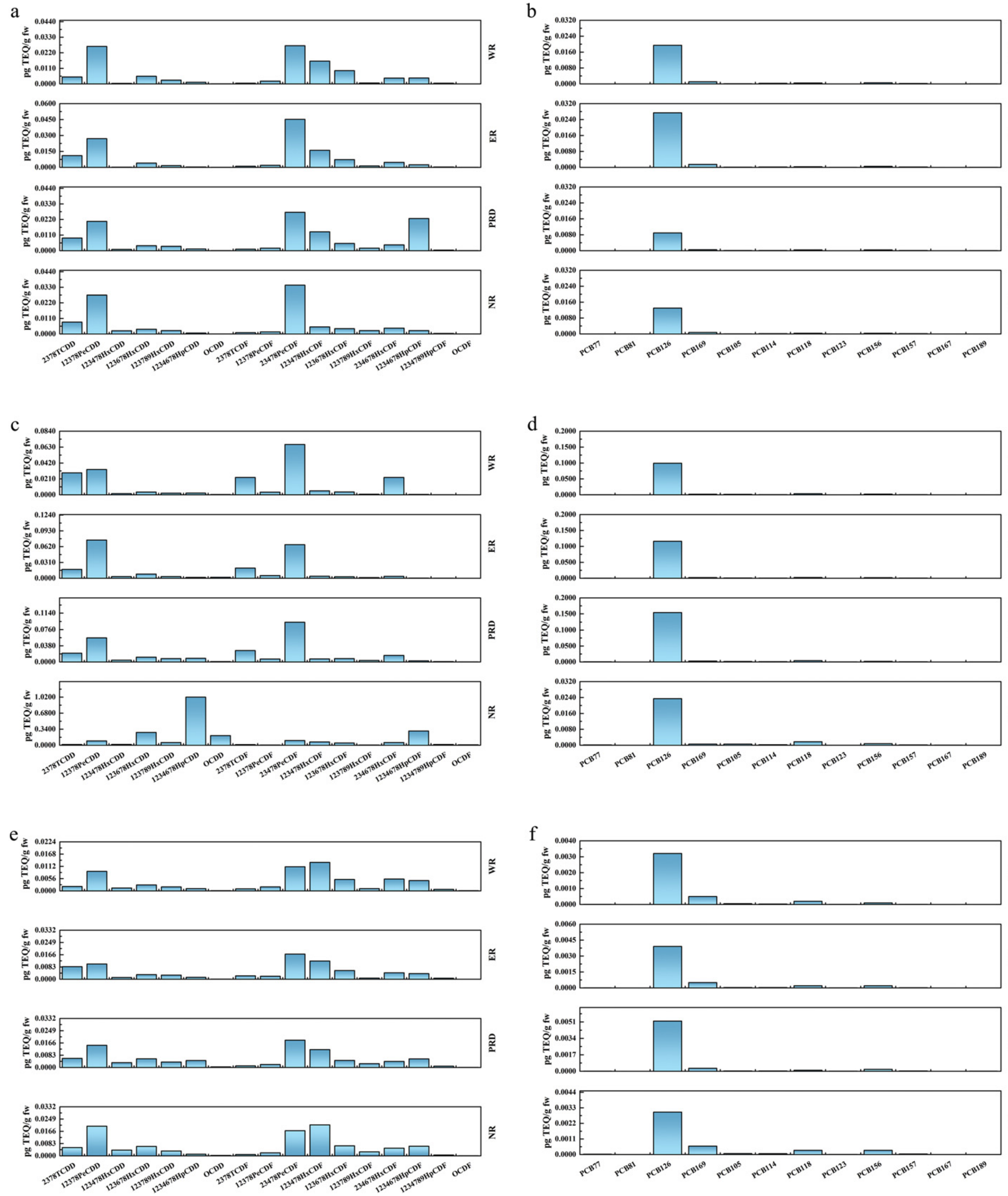



**Fig. 4.** Congener profiles of PCDDs/Fs and DL-PCBs in the three food categories from four regions of Guangdong Province.

reported for children. Male adults consumed more food than female adults did in all the three food categories, resulting in higher total TEQ exposure, which was in contrast with the results for children (Table 2). Children were more likely to be exposed to PCDD/Fs and DL-PCBs than adults, accounting for their low body weights.

However, the preliminary dietary intake of PCDD/Fs and DL-PCBs indicated that local residents in the contaminated regions could be exposed to excessive intake of total TEQ in comparison with PTMI (Table S7) when the TEQ levels of food samples collected from these regions were used for the estimation. This finding suggests that a greater

**Table 2**
Estimated daily intake (pg TEQ/day) and monthly intake (pg TEQ/kg bw/month) of PCDD/Fs and DL-PCBs for children (age 6–18 years) and adult (age 18–70 years) in Guangdong.

| Food group | Concentration (pg TEQ/g fw) | Boys (average body weight: 46.5 kg) | | | Girls (average body weight: 41.1 kg) | | | Male adults (average body weight: 64.5 kg) | | | Female adults (average body weight: 54.6 kg) | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Consumption (g/day) | Daily intake | Monthly intake | Consumption (g/day) | Daily intake | Monthly intake | Consumption (g/day) | Daily intake | Monthly intake | Consumption (g/day) | Daily intake | Monthly intake |
| Beef | 0.174 | 7.3 | 1.27 | 0.82 | 5.1 | 0.89 | 0.65 | 8.6 | 1.50 | 0.70 | 6.7 | 1.17 | 0.64 |
| Freshwater fish | 0.488 | 35.2 | 17.18 | 11.08 | 43.0 | 20.98 | 15.31 | 63.0 | 30.74 | 14.30 | 54.1 | 26.40 | 14.51 |
| Pork | 0.113 | 99.7 | 11.27 | 7.27 | 88.5 | 10.00 | 7.30 | 106.0 | 11.98 | 5.57 | 70.4 | 7.96 | 4.37 |
| Total | | | | 19.17 | | | 23.26 | | | 20.57 | | | 19.52 |

risk for adverse effects on the health of local population of the polluted regions and the dietary intake of PCDD/Fs and DL-PCBs via the local food should be further evaluated.

The dietary exposures of PCDD/Fs and DL-PCBs in this study were compared with the results for the local residents in different regions to understand the dietary intake of PCDD/Fs and DL-PCBs by Guangdong residents. The results indicated that the dietary intake of PCDD/Fs and DL-PCBs via beef was similar to those reported for the Zhejiang Province, China (0.3 pg TEQ/kg bw/month) (Shen et al., 2017), and Taiwan (0.27 pg TEQ/kg bw/month) (Wang et al., 2009), and significantly lower than the levels of western countries, such as Belgium (10.8 pg TEQ/kg bw/month) (Focant et al., 2002) and Egypt (2.4–-5.1 pg TEQ/kg bw/month) (Loutfy et al., 2006). In reference to the dietary intake of PCDD/Fs and DL-PCBs from freshwater fish, our result was comparable to the exposure levels of Western population (Costopoulou et al., 2016; Focant et al., 2002; Loutfy et al., 2006; Perelló et al., 2012) and Shenzhen residents (18.5 pg TEQ/kg bw/month and 14.1 pg TEQ/kg bw/month) (G. Zhang et al., 2007a; J.Q. Zhang et al., 2007b; Zhang et al., 2008), but prominently higher than the exposure levels of Zhejiang Province, China (Shen et al., 2017), Taiwan (Wang et al., 2009), Austria (Rauscher-Gabernig et al., 2013), and France (Sirot et al., 2012). However, the level of exposure of Guangdong residents to PCDD/Fs and DL-PCBs from fish was not high, the dietary intake of fish by the residents of Valencia, Spain was much higher reaching 48.6 pg TEQ/kg bw/month (adult) and 67.8 pg TEQ/kg bw/month (children). For the dietary intake of PCDD/Fs and DL-PCBs from pork, it was higher than the levels reported in Belgium (Focant et al., 2002; Windal et al., 2010), Taiwan (Wang et al., 2009), and the Zhejiang Province of China (Shen et al., 2017).

## 4. Conclusion

As a survey of food contaminants for Guangdong Province to have in-depth insights of the contamination status of PCDD/Fs and DL-PCBs in foodstuffs, this study first reports the general TEQ levels and congener profiles in three food groups including beef, freshwater fish, and pork, and dietary intakes have been evaluated for adults and children.

For contamination level, low concentrations of total TEQs were detected in most food samples. However, it is notable that the samples of three food groups collected from the vicinities of polluted regions had high levels of PCDD/Fs and DL-PCBs that significantly exceeded the MLs set by the EU (Table S7), which indicated that more categories of foodstuffs cultivated or produced from the contamination areas should be intently concerned through a further study for local residents. For congener profile, unique major congeners and congener distributions were observed in different food categories and sampling regions, but the primary reasons caused these results still needs more investigations.

The dietary intakes for adults and children were estimated on basis of the median TEQs levels of three food groups and the corresponding food consumption data, and the results indicated that the exposure to PCDD/Fs and DL-PCBs via the studied foodstuffs did not exceed the PTMI established by JECFA. Exposure estimation for the local residents in dioxins pollution areas did not proceed in this study because the quantities of the foodstuffs collected from these regions were not enough for a valuable estimation. However, the preliminary dietary intakes estimated from the available data indicated that the dietary exposures of PCDD/Fs and DL-PCBs via freshwater fish and pork obtained from the polluted regions were higher than the PTMI (Table S7), which suggested that contaminated foods could have adverse effects on the health of local residents, and therefore, more attention should be paid to the risk posed by these food items on the health of local residents in the further study.

## Acknowledgements

This study was sponsored for the following foundations: (1) Science and Technology Planning Project of Guangzhou City, China (2017010160523); (2) Science and Technology Planning Project of Guangdong Province, China (2016A040403071); (3) Science and Technology Planning Project of Guangdong Province, China (2015A03040105). The authors kindly thank for these foundations. We also thank for the technical support in determination of PCDD/Fs and DL-PCBs in three food categories from Shenzhen Disease Control and Prevention Center and Guangdong Testing Institute of Product Quality Supervision.

## Appendix A. Supplementary data

Table S1 and Fig. S1 specify the sampling information of the sizes of each category (beef, freshwater fish and pork) in each sampling location of 17 sampling areas in Guangdong Province. The median, average, and range of the TEQ levels of three groups of food samples collected from Guangdong Province are summarized in Table S2. The median and mean TEQ levels (pg TEQ/g fw) of PCDD/Fs and DL-PCBs in three food categories from four regions of Guangdong Province are summarized in Table S3–Table S4, respectively. The measurement uncertainties (pg/g fw) of the analytical method for PCDD/Fs and DL-PCBs in foodstuffs using HRGC-HRMS are listed in Table S5. Table S6 shows the comparison of the mean total TEQ levels of three food groups with the results reported by the published literatures. Table S7 summaries the total TEQ levels of PCDD/Fs and DL-PCBs in the samples exceeded the EU regulations and their contributions to the estimated monthly intake (pg TEQ/kg bw/month) of PCDD/Fs and DL-PCBs for local residents in E-waste dismantling regions. Supplementary data associated with this article can be found in the online version, at doi: https://doi.org/10.1016/j.scitotenv.2017.09.273.

## References

Adu-Kumi, S., Kawano, M., Shiki, Y., Yeboah, P.O., Carboo, D., Pwamang, J., Morita, M., Suzuk, N., 2010. Organochlorine pesticides (OCPs), dioxin-like polychlorinated biphenyls (dl-PCBs), polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzo furans (PCDD/Fs) in edible fish from Lake Volta, Lake Bosumtwi and Weija Lake in Ghana. Chemosphere 81, 675–684.

Bocio, A., Domingo, J.L., Falcó, G., Llobet, J.M., 2007. Concentrations of PCDD/PCDFs and PCBs in fish and seafood from the Catalan (Spain) market: estimated human intake. Environ. Int. 33, 170–175.

Chan, J.K.Y., Wong, M.H., 2013. A review of environmental fate, body burdens, and human health risk assessment of PCDD/Fs at two typical electronic waste recycling sites in China. Sci. Total Environ. 463–464, 1111–1123.

Chan, J.K.Y., Man, Y.B., Xing, G.H., Wu, S.C., Murphy, M.B., Xu, Y., Wong, M.H., 2013. Dietary exposure to polychlorinated dibenzo-*p*-dioxins and dibenzofurans via fish consumption and dioxin-like activity in fish determined by H4IIE-luc bioassay. Sci. Total Environ. 463–464, 1192–1200.

Chen, H.L., Su, H.J., Hsu, J.F., Liao, P.C., Lee, C.C., 2008. High variation of PCDDs, PCDFs, and DL-PCBs ratio in cooked food from the first total diet survey in Taiwan. Chemosphere 70, 673–681.

Costopoulou, D., Vassiliadou, I., Leondiadis, L., 2016. PCDDs, PCDFs and PCBs in farmed fish produced in Greece: levels and human population exposure assessment. Chemosphere 146, 511–518.

De Mul, A., Bakker, M.I., Zeilmaker, M.J., Traag, W.A., van Leeuwen, S.P.J., Hoogenboom, R.L.A.P., Boon, P.E., van Klaveren, J.D., 2008. Dietary exposure to dioxins and DL-PCBs in The Netherlands anno 2004. Regul. Toxicol. Pharmacol. 51, 278–287.

Durand, B., Dufour, B., Fraisse, D., Defour, S., Duhem, K., Le-Barillec, K., 2008. Levels of PCDDs, PCDFs and DL-PCBs in raw cow's milk collected in France in 2006. Chemosphere 70, 689–693.

Eduljee, G.H., 2001. Budget and source inventories: issues and challenges. In: Harrad, S. (Ed.), Persistent Organic Pollutants: Environmental Behaviour and Pathways of Human Exposure. Kluwer Academic Publishers, New York, pp. 1–29.

Esposito, M., Cavallo, S., Serpe, F.P., D'Ambrosio, R., Gallo, P., Colarusso, G., Pellicanò, R., Baldi, L., Guarino, A., Serpe, L., 2009. Levels and congener profiles of polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans and dioxin-like polychlorinated biphenyls in cow's milk collected in Campania, Italy. Chemosphere 77, 1212–1216.

Fernandes, A., Mortimer, D., Rose, M., Gem, M., 2010. Dioxins (PCDD/Fs) and PCBs in offal: occurrence and dietary exposure. Chemosphere 81, 536–540.

Focant, J.F., Eppe, G., Pirard, C., Massart, A.C., André, J.E., De Pauw, E., 2002. Levels and congener distributions of PCDDs, PCDFs and non-*ortho* PCBs in Belgian foodstuffs assessment of dietary intake. Chemosphere 48, 167–179.

Grassi, P., Fattore, E., Generoso, C., Fanelli, R., Arvati, M., Zuccato, E., 2010. Polychlorobiphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) in fruit and vegetables from an industrial area in northern Italy. Chemosphere 79, 292–298.

Han, J.L., Shen, H.T., Tie, X.W., Zhang, W.P., Zhu, G.N., Ren, Y.P., 2007. Polychlorinated dibenzo-*p*-dioxins/furans and polychlorinated biphenyls in fresh fishes from Qiantangjiang River, China. Chemosphere 68, 112–119.

Hoogenboom, R.L.A.P., ten Dam, G., van Bruggen, M., Jeurissen, S.M.F., van Leeuwen, S.P.J., Theelen, R.M.C., Zeilmaker, M.J., 2016. Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and biphenyls (PCBs) in home-produced eggs. Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and biphenyls (PCBs) in home-produced eggs. Chemosphere 150, 311–319.

Hsu, M.S., Hsu, K.Y., Wang, S.M., Chou, U., Chen, S.Y., Huang, N.C., Liao, C.Y., Yu, T.P., Ling, Y.C., 2007. A total diet study to estimate PCDD/Fs and dioxin-like PCBs intake from food in Taiwan. Chemosphere 67, S65–S70.

Huwe, J., Larsen, G., 2005. Polychlorinated dioxins, furans, and biphenyls, and polybrominated diphenyl ethers in a U.S. meat market basket and estimates of dietary intake. Environ. Sci. Technol. 39, 5606–5611.

JECFA, 2001. Evaluation of certain Food Addit. Contam. (Fifty-seventh report of the Joint FAO/WHO Expert Committee on Food Additive). WHO Technical Report Series 909, pp. 139–146.

Kim, M.K., Kim, S.Y., Yun, S.J., Kim, D.G., Chung, G.S., 2007. Background levels and dietary intake of PCDD/Fs in domestic and imported meat in South Korea. Chemosphere 69, 479–484.

Lee, C.C., Lin, H.T., Kao, Y.M., Chang, M.H., Chen, H.L., 2016. Temporal trend of polychlorinated dibenzo-*p*-dioxin/polychlorinated dibenzofuran and dioxin like-polychlorinated biphenyl concentrations in food from Taiwan markets during 2004-2012. J. Food Drug Anal. 24, 644–652.

Li, J.G., Wu, Y.N., Zhang, L., Zhao, Y.F., 2007. Dietary intake of dioxins, furans and dioxin-like polychlorinated biphenyls from foods of animal origin in China. Food Addit. Contam. Part A: Chem. Anal. Control Expo. Risk Assess. 24, 186–193.

Loutfy, N., Fuerhacker, M., Tundo, P., Raccanelli, S., El Dien, A.G., Ahmed, M.T., 2006. Dietary intake of dioxins and dl-PCBs, due to the consumption of dairy products, fish/seafood and meat from Ismailia city, Egypt. Sci. Total Environ. 370, 1–8.

Marin, S., Villalba, P., Diaz-Ferrero, J., Font, G., Yusà, V., 2011. Congener profile, occurrence and estimated dietary intake of dioxins and DL-PCBs in foods marketed in the Region of Valencia (Spain). Chemosphere 82, 1253–1261.

Moon, H.B., Ok, G., 2006. Dietary intake of PCDDs, PCDFs and DL-PCBs, due to the consumption of various marine organisms from Korea. Chemosphere 62, 1142–1152.

Munschy, C., Guiot, N., Héas-Moisan, K., Tixier, C., Tronczyjski, J., 2008. Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in marine mussels from French coasts: levels, patterns and temporal trends from 1981 to 2005. Chemosphere 73, 945–953.

Perelló, G., Gómez-Catalán, J., Castell, V., Llobt, J.M., Domingo, J.L., 2012. Assessment of the temporal trend of the dietary exposure to PCDD/Fs and PCBs in Catalonia, over Spain: health risks. Food Chem. Toxicol. 50, 399–408.

Rauscher-Gabernig, E., Mischek, D., Moche, W., Prean, M., 2013. Dietary intake of dioxins, furans and DL-PCBs in Austria. Food Addit. Contam. Part A: Chem. Anal. Control Expo. Risk Assess. 30, 1770–1779.

Sakuraia, T., Kima, J.G., Suzuki, N., Matsuoa, T., Li, D.Q., Yao, Y., Masunaga, S., Nakanishi, J., 2000. Polychlorinated dibenzo-*p*-dioxins and dibenzofurans in sediment, soil, fish, shellfish and crab samples from Tokyo Bay area, Japan. Chemosphere 40, 627–640.

Sasamoto, T., Ushio, F., Kikutani, N., Saitoh, Y., Yamaki, Y., Hashimoto, T., Horii, S., Nakagawa, J., Ibe, A., 2006. Estimation of 1999–2004 dietary intake of PCDDs, PCDFs and dioxin-like PCBs by a total diet study in metropolitan Tokyo, Japan. Chemosphere 64, 634–641.

Schmid, P., Gujer, E., Zennegg, M., Studer, C., 2003. Temporal and local trends of PCDD/F levels in cow's milk in Switzerland. Chemosphere 53, 129–136.

Shen, C.F., Chen, Y.X., Huang, S.B., Wang, Z.J., Yu, C.N., Qiao, M., Xu, Y.P., Setty, K., Zhang, J.Y., Zhu, Y.F., Lin, Q., 2009. Dioxin-like compounds in agricultural soils near e-waste recycling sites from Taizhou area, China: chemical and bioanalytical characterization. Environ. Int. 30, 50–55.

Shen, H.T., Yu, C., Ying, Y., Zhao, Y.F., Wu, Y.N., Han, J.L., Xu, Q.Y., 2009. Levels and congener profiles of PCDD/Fs, PCBs and PBDEs in seafood from China. Chemosphere 77, 1206–1211.

Shen, H.T., Ding, G.Q., Wu, Y.N., Pan, G.S., Zhou, X.P., Han, J.L., Li, J.G., Wen, S., 2012. Polychlorinated dibenzo-*p*-dioxins/furans (PCDD/Fs), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) in breast milk from Zhejiang, China. Environ. Int. 42, 84–90.

Shen, H.T., Guan, R.F., Ding, G.Q., Chen, Q., Lou, X.M., Chen, Z.J., Zhang, L., Xing, M.L., Han, J.L., Wu, Y.N., 2017. Polychlorinated dibenzo-*p*-dioxins/furans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in Zhejiang foods (2006–2015): market basket and polluted areas. Sci. Total Environ. 517, 120–127.

Sirot, V., Tard, A., Venisseau, A., Brosseaud, A., Marchand, P., Le Bizec, B., Leblanc, J.C., 2012. Dietary exposure to polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans and polychlorinated biphenyls of the French population: results of the second French Total Diet Study. Chemosphere 88, 492–500.

Song, Y., Wu, N.X., Han, J.L., Shen, H.T., Tan, Y.F., Ding, G.Q., Xiang, J.X., Tao, H., Jin, S.L., 2011. Levels of PCDD/Fs and DL-PCBs on selected foods and estimated dietary intake for the local residents of Luqiao and Yuhang in Zhejiang, China. Chemosphere 85, 329–334.

Squadrone, S., Brizio, P., Nespoli, R., Stella, C., Abete, M.C., 2015. Human dietary exposure and levels of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), dioxin-like polychlorinated biphenyls (DL-PCBs) and non-dioxin-like polychlorinated biphenyls (NDL-PCBs) in free-range eggs close to a secondary aluminum smelter, Northern Italy. Environ. Pollut. 206, 429–436.

Suzuki, G., Someya, M., Matsukami, H., Tue, N.M., Uchida, N., Tuyen, L.H., Viet, P.H., Takahashi, S., Tanabe, S., Brouwer, A., Takigami, H., 2016. Comprehensive evaluation of dioxins and dioxin-like compounds in surface soils and river sediments from e-waste-processing sites in a village in northern Vietnam: heading towards the environmentally sound management of e-waste. Emerg. Contam. 2, 98–108.

Taioli, E., Marabelli, R., Scortichini, G., Migliorati, G., Pedotti, P., Cigliano, A., Caporale, V., 2005. Human exposure to dioxins through diet in Italy. Chemosphere 61, 1672–1676.

Tsutsumi, T., Yanagi, T., Nakamura, M., Kono, Y., Uchibe, H., Iida, T., Hori, T., Nakagawa, R., Tobiishi, K., Matsuda, R., Sasaki, K., Toyoda, M., 2001. Update of daily intake of PCDDs, PCDFs, and dioxin-like PCBs from food in Japan. Chemosphere 45, 1129–1137.

Tue, N.M., Goto, A., Takahashi, S., Itai, T., Asante, K.A., Kunisue, T., Tanabe, S., 2016. Release of chlorinated, brominated and mixed halogenated dioxin-related compounds to soils from open burning of e-waste in Agbogbloshie (Accra, Ghana). J. Hazard. Mater. 302, 151–157.

Van den Berg, M., Birnbaum, L., Bosveld, A.T., Brunstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., van Leeuwen, F.X., Liem, A.K., Nolt, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillitt, D., Tysklind, M., Younes, M., Waern, F., Zacharewski, T., 1998. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. Environ. Health Perspect. 106, 775–792.

Van den Berg, M., Birnbaum, L.S., Denison, M., de Vito, M., Farland, W., Feeley, M., Fiedler, H., Hakansson, H., Hanberg, A., Haws, L., Rose, M., Safe, S., Schrenk, D., Tohyama, C., Tritscher, A., Tuomisto, J., Tysklind, M., Walker, N., Pete, R.E., 2006. The 2005 world health organization reevaluation of human and mammalian toxic equivalency factors for dioxins and dioxin: like compounds. Toxicol. Sci. 93, 223–241.

Wang, I.C., Wu, Y.L., Lin, L.F., Chien, G.P.C., 2009. Human dietary exposure to polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in Taiwan. J. Hazard. Mater. 164, 621–626.

Wang, X.Y., Zhang, H.X., Zhang, L., Zhong, K., Shang, X.H., Zhao, Y.F., Tong, Z.D., Yu, X.W., Li, J.G., Wu, Y.N., 2015. Assessment on dioxin-like compounds intake from various marine fish from Zhoushan Fishery, China. Chemosphere 118, 163–169.

Windal, I., Vandevijvere, S., Maleki, M., Goscinny, S., Vinkx, C., Focant, J.F., Eppe, G., Hanot, V., Van Loco, J., 2010. Dietary intake of PCDD/Fs and dioxin-like PCBs of the Belgian population. Chemosphere 79, 334–340.

Wong, M.H., Wu, S.C., Deng, W.J., Yu, X.Z., Luo, Q., Leung, A.O.W., Wong, C.S.C., Luksemburg, W.J., Wong, A.S., 2007. Export of toxic chemicals–a review of the case of uncontrolled electronic-waste recycling. Environ. Pollut. 149, 131–140.

Zhang, Y.H., Ma, W.J. (Eds.), 2016. Analysis for Ten-years Changes of Dietary Nutrition and Health Status of Guangdong Residents. China Zhijian Publishing House, Beijing.

Zhang, G., Li, X.D., Mai, B.X., Peng, P.A., Ran, Y., Wang, X.M., Zeng, E.Y., Sheng, G.Y., Fu, J.M., 2007. Sources and occurrence of persistent organic pollutants in the Pearl River Delta, South China. In: Li, A., Tanabe, S., Jiang, G., Giesy, J.P., Lam, P.K.S. (Eds.), Developments in Environmental Science. Elsevier, Netherlands, pp. 289–311.

Zhang, J.Q., Zhou, J., Jiang, Y.S., Jiang, J., Zhuang, Z.X., Liu, X.L., Wu, Y.N., 2007. Polychlorinated dibenzo-*p*-dioxin and dibenzo furan concentrations in common fish species in the Pearl River Delta area, China. Chemosphere 66, 199–202.

Zhang, J.Q., Jiang, Y.S., Zhou, J., Fang, D.K., Jiang, J., Liu, G.H., Zhang, H.Y., Xie, J.B., Huang, W., Zhang, J.Z., Li, H., Wang, Z., Pan, L.B., 2008. Concentrations of PCDD/Fs and PCBs in retail foods and an assessment of dietary intake for local population of Shenzhen in China. Environ. Int. 34, 799–803.

Zhang, K., Schnoor, J.L., Zeng, E.Y., 2012. E-waste recycling: where does it go from here? Environ. Sci. Technol. 46, 10861–10867.

Zhang, L., Li, J.G., Liu, X., Zhao, Y.F., Li, X.W., Wen, S., Wu, Y.N., 2013. Dietary intake of PCDD/Fs and dioxin-like PCBs from the Chinese total diet study in 2007. Chemosphere 90, 1625–1630.

Zhang, L., Yin, S.X., Wang, X.Y., Li, J.G., Zhao, Y.F., Li, X.W., Shen, H.T., Wu, Y.N., 2015. Assessment of dietary intake of polychlorinated dibenzo-*p*-dioxins and dibenzofurans and dioxin-like polychlorinated biphenyls from the Chinese Total Diet Study in 2011. Chemosphere 137, 178–184.