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Hair mercury levels and food consumption in residents from the Pearl River Delta: South China

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ABSTRACT

The Pearl River Delta (PRD) is located in the Southern part of China and is the main region for fish culture in Guangdong Province. In order to assess the potential health risks associated with dietary consumption of mercury, hair samples from 91 urban, town and fishing village residents, 37 species of fish, cereal, vegetables, and meat samples were collected. The average total mercury (THg) and methylmercury (MeHg) concentrations in hair were 1.08 ± 0.94 and 0.58 ± 0.59 μg/g, respectively. Daily Hg intake via fish consumption is significantly correlated with THg and MeHg accumulated in human hair ($r = 0.48$, $p < 0.01$; $r = 0.43$, $p < 0.01$). The estimated daily intake of Hg via different food types showed that both fish and cereal consumption were the two main routes of Hg exposure for residents in the sampling areas. Besides food intake, smoking was also an important source for daily THg intake in the smoke group, contributing 11–18% to EDI of THg.

1. Introduction

Mercury (Hg) is a potent neurotoxin of significant ecological and public health concern. Vapourised elemental Hg emitted into the atmosphere by mining and coal combustion may be over transported very long distances and contaminate water and soil upon deposition. Although Hg is deposited primarily in its inorganic form, it can be converted by microbial activity into a more toxic form: methylmercury (MeHg) under anaerobic conditions (Harmon, King, Gladden, & Newman, 2007). Toxicity of MeHg can be exacerbated by bioaccumulation and biomagnification through food webs in aquatic systems (Cheng et al., 2011; Jaeger, Hop, & Gabrielsen, 2009).

Fish consumption is considered the main source of MeHg exposure in populations that are not occupationally exposed (Johnsson, Sallsten, Schutz, Sjors, & Barregard, 2004). About 75–100% of Hg in fish exists as MeHg (Shao et al., 2011). Chien, Gao, and Lin (2010) revealed that people who consumed greater quantities of fish had higher hair Hg concentrations ($r = 0.32$, $p < 0.0001$). In Kuwait, there were significant differences in MeHg concentrations in fishermen hair between high and low fish consumption groups (Al-Majed & Preston, 2000). In addition, cereals and cereal products are the largest source of dietary Hg intake in China (54%) (Jiang, Shi, & Feng, 2006). A recent study indicated that rice is the main route of MeHg exposure for residents whom seldom consumed fish meals (1.2 g day^{-1}), accounted for 94–96% of the PDI of MeHg in Guizhou province of China (Zhang, Feng, Larssen, Qiu, & Vogt, 2010). The levels of THg in human hair are also influenced by their smoking cigarettes. The concentration of THg is significantly higher for smokers as compared to non-smoking groups (Kowalski & Wiercinski, 2007). Heavy metal (Cd, Cu, Pb, Cr, Zn, Hg) concentrations of tobacco had been determined in 47 brands of cigarettes from five countries. The average THg concentration ranged between 0.02 and 0.11 μg g^{-1}, and 0.04 μg g^{-1} for China (German Müller, 2000).

Mercury concentrations in hair and blood have been widely used as biomarkers for human Hg exposure. The normal ratio of Hg in hair (μg g^{-1}) and blood (ng L^{-1}) is frequently cited as 250:1 (US EPA, 1997). When compared to blood, hair is more widely used as a non-invasive method with higher element concentrations (Salehi & Esmaili-Sari, 2010). Moreover, MeHg can accumulate in hair during growth (1 cm per month), and hair MeHg concentration can possibly reflect longer-term MeHg exposure (Karouna-Renier et al., 2008). It has been noted that MeHg exposure may cause an increased risk of foetal brain damage if the maternal hair Hg concentration exceeds a level of 10–20 μg g^{-1} (WHO, 1990). For instance, the high dose of MeHg exposure in Minamata of Japan and in Iraq during the 1970 caused foetal death, serious birth defects, mental retardation and blindness (NRC, 2000). It has been noted that higher maternal Hg levels (>1.2 μg g^{-1}) are associated with lower offspring cognition (Oken et al., 2005).

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Table 1
Average daily intake of THg and MeHg through major food consumption for population (body weight 58 kg) in PRD.

Food item	Intake rate ($g\ day^{-1}$)	THg daily intake ($\mu g\ day^{-1}$)				MeHg daily intake ($\mu g\ day^{-1}$)			
		GZ	NH	SD	ZS	GZ	NH	SD	ZS
Fish	52–341	2.42	3.98	15.2	28.9	1.89	3.54	12.8	23.0
Meat	82–113	0.19	0.50	0.90	0.34	0.12	0.24	0.42	0.14
Vegetable	224–541	0.32	0.44	0.44	0.90	0.01	0.01	0.02	0.02
Cereal	261–357	7.32	3.66	3.22	3.50	2.69	1.92	1.54	1.82
Total	$\mu g\ day^{-1}$	10.3	8.58	19.8	33.6	4.71	5.71	14.8	25.0
	$\mu g\ kg^{-1}\ day^{-1}$	0.18	0.15	0.34	0.58	0.08	0.10	0.25	0.43

The Pearl River Delta (PRD) is located in the Southern part of China and is the main region for fish culture in Guangdong Province. Shao et al. (2011) showed that the fish collected from freshwater fish ponds in PRD contained MeHg ranging from 5.93–76.1 $ng\ g^{-1}$ wet wt, and consumption of carnivorous fish might impose health risk to PRD residents, especially for children. However, assessment results may be influenced by uncertainly factors such as fish ingestion rates and quantities. Besides, personal information such as age, gender, body weight and height, food consumption customs, as well as smoking and drinking habits should also be considered. So far, there is a lack of studies investigating the Hg levels and the main source of Hg exposure in residents of PRD.

The major objectives of the present study were to (1) assess Hg exposure by measuring THg and MeHg in the hair of residents in PRD and to (2) evaluate human exposure to Hg via dietary intake and cigarette smoking, in order to quantify the main source of Hg intake at sampling sites.

2. Materials and methods

2.1. Sample collection

2.1.1. Questionnaire survey

Food consumption pattern such as type and average daily intake of cereal, vegetables, meat, and fish was collected. A questionnaire was used to collect information from volunteers in order to assess their dietary habits. Other information including age, weight, profession, dental fillings, smoking and alcohol drinking habits were also collected.

2.1.2. Hair sample collection

Using clean stainless steel scissors, 30, 20, 20 and 21 human hair samples were collected from four communities in Guangdong province (Fig. S1): Guangzhou (GZ: residential area), Nanhai (NH: town), Shunde (SD: fishing village) and Zhongshan (ZS: fishing village), respectively. Volunteers from Guangzhou and Nanhai included students, teachers, office workers and farmers, aged between 20 and 45 years. In addition, the volunteers from two fish villages in Shunde and Zhongshan,, included fishermen and farmers aged between 24 and 54 years. The hair samples were taken from several sites of the scalp (1–3 cm), placed and sealed in clean polyethylene bag until chemical analysis.

2.1.3. Food and cigarette sample collection

The selection of food items for analyses was based on the results of food consumption survey, as well as their availability in the local markets or grocery stores. Therefore, four major food groups: (1) fish and shellfish, (2) meat, (3) vegetables, and (4) cereals were selected in the present study (Table S1, Supporting Information). Eight species of freshwater fish and three species of shellfish were collected from local fish ponds and fish markets. Twenty species of vegetables, four types of meat and two kinds of cereal samples were collected from local markets. In addition, three popular cigarette brands were bought from tobacco stores. As a general guideline, each type of sample with four replicates each was randomly purchased from three different shops/stalls in each market.

2.2. Hair and food sample analyses

Hair samples were cut into short segments (about 5 mm) and washed successively with acetone and Milli-Q water, and dried in an oven at 60 °C overnight. Fish (dorsal muscles), meat and vegetable samples were freeze-dried, crushed, and ground into powder. Hair, vegetable, meat and rice samples were digested using KOH–methanol/solvent extraction technique for the detection of MeHg (Liang, Bloom, & Horvat, 1994; Liang, Horvat, Cernichiari, Gelein, & Balogh, 1996). 0.1–0.2 g sample was digested with 25% KOH methanol (2.5 mL) in an oven at 70 °C for 3 h. The solution was diluted to 20 mL with methanol after cooling. The solution (30 μl) was added to 40 mL vials with Teflon lined septa caps. Samples were buffered (300 μL) to pH 4.9, ethylated with the addition of $NaBEt_4$ (40 μL), and made up to volume with Milli-Q water, capped, shaken and loaded into the auto sampler of the MeHg analyzer. It was ensured that the vials were absent of air by filling Milli-Q water.

The THg concentration was analysed by the direct mercury analyzer DMA-80 (Milestone, USA) following US EPA Method 7473 (US EPA, 1998). Measurements of MeHg were conducted using the automated modular mercury system from Brooks Rand (MERX, Brooks Rand Labs, USA).

Table 2
Mercury concentrations ($\mu g\ g^{-1}$) in hair of residents from four regions in PRD.

Sampling site	*n*	Mean age (y)	Height (cm)	Weight (kg)	Smoking percent (%)	Fish meals per week	Hg concentration ($\mu g\ g^{-1}$)		
								Mean ± SD	Range
GZ	30	24.5	167	57.2	3.33	1.70	THg	0.39 ± 0.25	0.14–1.11
							MeHg	0.22 ± 0.10	0.03–0.67
NH	20	29.2	163	56.1	20.0	1.95	THg	0.84 ± 0.41	0.25–1.69
							MeHg	0.41 ± 0.19	0.19–0.77
SD	20	40.1	166	60.9	45.0	3.30	THg	1.39 ± 1.43	0.45–7.15
							MeHg	0.86 ± 0.99	0.15–4.64
ZS	21	43.8	165	58.1	52.4	3.67	THg	1.78 ± 0.84	0.55–4.22
							MeHg	0.93 ± 0.46	0.14–2.12

2.3. Cigarette sample determination

Cigarettes of each brand were divided into two parts which were stored in desiccators to prevent from drying. One part of each sample was subjected to determination of THg concentration in tobacco and filter, and the other part was simulated smoking by a syringe with subsequent determination of THg content in ash and filter. THg determination was carried out using the same method as hair and food samples.

2.4. QA/QC

Quality control system consisted of method blanks, blank spikes, matrix spikes, Certified reference materials (CRM) and blind duplicates. The limits of determination were 0.05 ng g^{-1} for THg in all samples and 0.02 ng g^{-1} for MeHg in both hair and food samples, respectively. The accuracy of Hg analysis was examined by using SRM with each batch of samples (set of 20 samples). The recovery rates varied between 94.6 and 105% for THg and 94.6% and 98.0% for MeHg (Table S2, Supporting Information). Three blank samples were also run with each set. The results showed undetectable levels of THg and MeHg in all the analysed blank samples. In order to check the reproducibility of the analysis, 22% of the samples were analysed in duplicate.

2.5. Calculation of probable daily intake (PDI)

To determine THg and MeHg exposure via food consumption and smoking, the probable daily intake (PDI) values for the general adult population were calculated according to the following formula:

$$\mathrm{PDI_{THg}} = \frac{\sum(C^i_{THg} \times \mathrm{IR}^i)}{\text{body weight}\,(\mathrm{kg})} \quad (1)$$

$$\mathrm{PDI_{MeHg}} = \frac{\sum(C^i_{MeHg} \times \mathrm{IR}^i)}{\text{body weight}\,(\mathrm{kg})} \quad (2)$$

where PDI is given in micrograms per kilogram of body weight (bw) per day; body weight = 58 kg; C is the concentration of exposed medium; IR is intake rate, and i = intake of fish, meat, vegetable, rice and cigarette. The intake rates for different exposure media for the adult populations used were based on the questionnaire survey (Table 1).

THg absorption was calculated using the following formula (Kowalski & Wierciński, 2009):

$$\mathrm{Absorption_{THg}} = [C_{Tobacco} - C_{Ash} - (C^{aftersmoke}_{Filter} - C^{beforesmoke}_{Filter})] \times 80\% \quad (3)$$

where C is the THg concentration; 80% is the percentage of THg retention in the organism via smoke available for absorption in the lung.

2.6. Statistical analyses

The relationship between mercury in hair and PDI of mercury via fish consumption was determined by linear regression. Group difference in Hg concentrations of hair was tested by two-way ANOVA. These statistical analyses were performed using the Statistical Package for Social Sciences (SPSS) 13.0. The level of significance was set at $p < 0.05$.

3. Results and discussion

3.1. THg and MeHg in hair

The mean THg and MeHg concentrations in the hair samples of participants were 1.08 ± 0.94 μg g^{-1} (0.14–7.15) and 0.58 ± 0.59 μg g^{-1} (0.03–4.64), respectively. 39.6% of the population exceeded the safety standard of 1 μg g^{-1} for hair Hg, corresponding to the reference dose (RfD) of 0.1 μg kg^{-1} body weight d^{-1} (US EPA, 1997), and 8.8% of the population had THg levels higher than hair Hg concentration for non-exposed people of 2 μg g^{-1} (Foulke, 1994). None of the data exceeded the WHO tolerance limit of

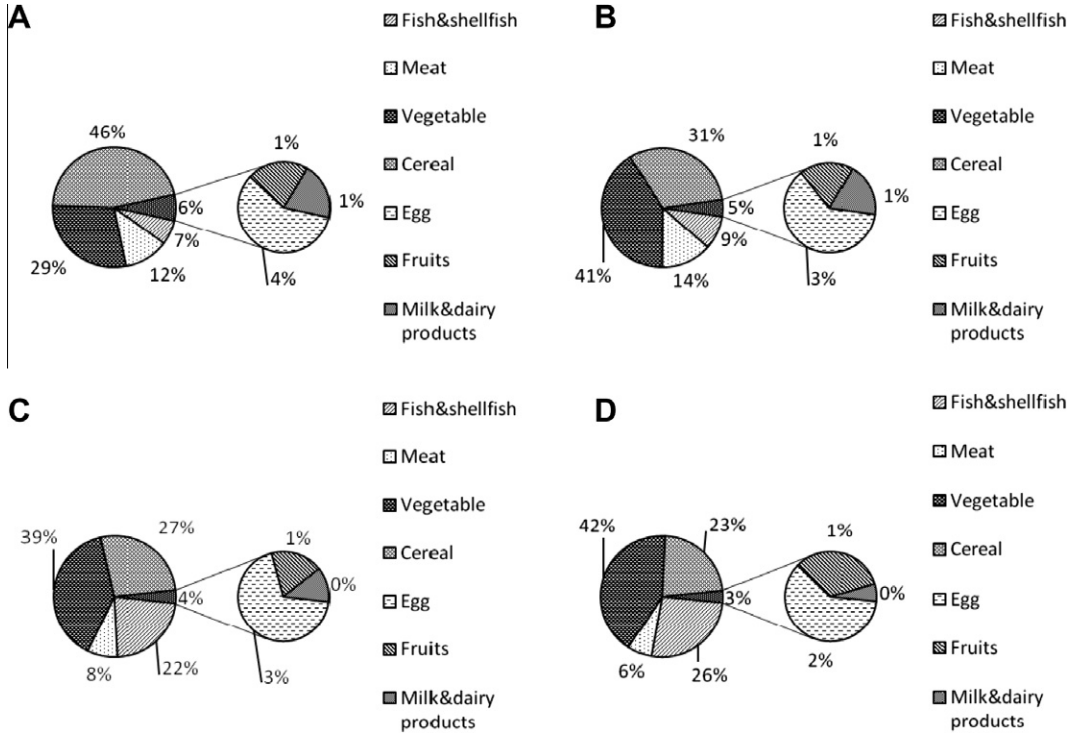

Fig. 1. Contribution to total diet of different food groups in (A) GZ, (B) NH, (C) ZS, and (D) SD.

10 μg g^{-1} (WHO, 1990). MeHg in the hair was significantly correlated with THg in the hair (r^2 = 0.81, p < 0.01, Fig. S2), which is similar to other studies (Feng et al., 2008; McDowell et al., 2004). MeHg in the hair accounted for, on average, 50–60% of THg for the residents in the PRD, similar to the results by Li et al. (2009) and Feng et al. (2008). Most of the previous studies reported that MeHg constituted 70–100% of THg in hair and the major exposure route was through fish consumption (Al-Majed & Preston, 2000; Barbosa, Jardim, Dórea, Fosberg, & Souza, 2001; McDowell et al., 2004). There was no specific Hg contamination source such as gold mining, coal combustion and fluorescent light factory located around the studied area, which may exclude the possibility of elevation of inorganic Hg concentration in hair samples through Hg vapour exposure. This ratio difference might have resulted from the diet of the sampled populations. Apart from fish, cereal, vegetables and meat were also major dietary routes in the present study. The percentages of inorganic Hg as Hg in cereal and vegetables from the PRD were close to 80% and 100%, respectively. Therefore, the inorganic Hg exposure through diet ingestion (for rice and vegetable) was very high, even though the absorbed proportion of inorganic Hg is very low (8%; WHO, 1991). This may also be responsible for the low percentages of Hg as Me-Hg in the hair samples. Table 2 shows the Hg levels in hair samples collected from GZ, NH, SD and ZS. Concentrations of THg and MeHg in hair samples collected from SD and ZS were significantly higher (p < 0.05) than those from GZ and NH. A significant correlation ($y = 0.586x - 0.053$, r = 0.93, p < 0.01) between THg and MeHg concentrations for participants was observed. Compared with studies on Chinese coastal people, the average concentration of THg in the hair in the present study was similar to that found in those of residents in Shanghai (0.5 μg g^{-1}), Xiamen (0.8 μg g^{-1}) and Ningbo (1.0 μg g^{-1}), but lower than that in fishermen from Zhoushan (2.1 μg g^{-1}) (Liu et al., 2008). This may be explained by high consumption rate (241.2 g day^{-1}) of fish for fishermen in Zhoushan.

3.2. Influencing factors

The personal information of the participants is shown in Table 2. The results showed that the participants from GZ (24.5 years of average age) and NH (29.2) were younger than those from SD (40.1) and ZS (43.8). They exhibited a relative narrow range of body weight and height. The percentages of participants who had smoking habit in SD (45.0%) and ZS (53.4%) were higher than those in GZ (3.33%) and NH (20.0%). In addition, the food consumption rates especially for daily fish consumption rates were different (Number of weekly fish meal: GZ 1.7, NH 1.9, SD 3.3, ZS 3.7) among these four areas. None of the participants had dental problem or amalgam fillings. Therefore, the levels of THg and MeHg were evaluated in relation to the following factors.

3.2.1. Age

The hair Hg concentrations in different age groups and statistic differences among all age groups are shown in Fig. S3. Significant positive correlations (r = 0.52, p < 0.01; r = 0.49, p < 0.01) were observed between age and THg (MeHg) levels in hair of residents. The mean concentrations of THg increased with age up to 49 and decreased thereafter. A similar trend was observed in MeHg concentrations. The highest mean values for THg and MeHg were 2.12 ± 1.57 and 1.21 ± 1.01 μg g^{-1} in the age group of 40–49 years, respectively; the lowest values for THg and MeHg were 0.57 ± 0.32 and 0.27 ± 0.18 μg g^{-1} for the age group of 25–29 years.

These results agreed with the findings of Liu et al. (2008) who found that the total hair Hg increased between twenties and forties, and then gradually decreased with age. Al-Majed and Preston, (2000) also showed that the THg and MeHg decreased in human hair after 45 years of age. However, Buzina et al. (1995) reported that the THg content of hair did not increase after 40 and 35 years of age, respectively. It seems apparent that body burden of Hg increased with age due to gradual accumulation, and cannot be easily excreted from human bodies. However, the reason why Hg content in hair decreased after 45 years old needs further investigation. One possible explanation is due to inclusion of grey hairs from the sample before analysis, as grey hairs have no Hg content (Al-Majed & Preston, 2000).

3.2.2. Smoking habit

In the present study, 27.5% of participants had cigarette smoking habit, and 36% of the smokers were older than 50 years old. Two-way ANOVA test showed there was no interaction between age and smoking habit (p > 0.05). The mean THg and MeHg levels in the hair of the smoking group were 1.76 ± 1.40 and 0.98 ± 0.90 μg g^{-1}, respectively, which were significantly higher (p < 0.05) than the values of 0.82 ± 0.51 and 0.42 ± 0.32 μg g^{-1} for THg and MeHg levels in the hair of non-smoking group. This may be explained by Hg contained in tobacco may enter into human

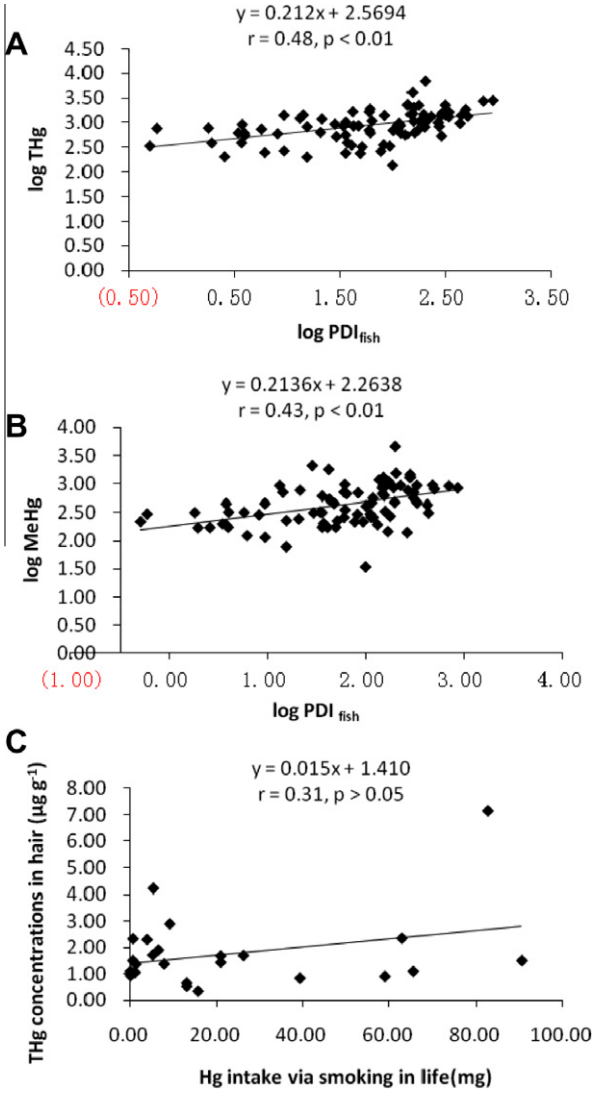

Fig. 2. The relationships between (A) THg concentrations in hair and PDI_{THg} via fish consumption, (B) MeHg concentrations in hair and PDI_{MeHg} via fish consumption, (C) THg concentrations in hair and THg intake via smoking cigarette.

body via smoking (Kowalski & Wierciński, 2009). The present study reported the influence of smoking on the increase of Hg concentration in human body, which was also observed in other studies (Kowalski & Wiercinski, 2007, 2009; Zaborowska & Wiercinski, 1999).

3.2.3. Fish consumption rate

Based on the fish consumption (fish and shellfish) rate, the participants were separated into three groups (low: 0–1 meal per week, median: 2–4 meal per week, high: >4 meal per week) to quantify any differences in hair Hg concentration. No interaction between age and rate of fish consumption was found ($p > 0.05$, two-way ANOVA). 23% of the participants in this study never ate or ate fish less than once a week and the mean values for THg and MeHg were 0.65 ± 0.37 (range: 0.14–1.47 $\mu g\, g^{-1}$) and 0.34 ± 0.23 $\mu g\, g^{-1}$ (range: 0.03–0.93 $\mu g\, g^{-1}$) in their hair, respectively. 37% ate fish 2–4 meals per week and their average hair Hg levels were 0.87 ± 0.75 $\mu g\, g^{-1}$ (range: 0.24–4.22 $\mu g\, g^{-1}$) for THg and 0.41 ± 0.37 $\mu g\, g^{-1}$ (range: 0.12–2.12 $\mu g\, g^{-1}$) for MeHg. 40% ate fish more than 4 times per week, the mean values 1.58 ± 1.16 $\mu g\, g^{-1}$ (range: 0.55–7.15 $\mu g\, g^{-1}$) for THg and 0.90 ± 0.78 $\mu g\, g^{-1}$ (range: 0.14–4.64 $\mu g\, g^{-1}$) for MeHg of hair Hg in this group were significantly higher ($p < 0.05$) than those in median and low fish consumption groups.

The relationship between hair THg concentration and rates of fish consumption among different groups are shown in Fig. S4. Previous studies reported that the fish consumption rate significantly affected Hg levels in human hair (Karouna-Renier et al., 2008; Salehi & Esmaili-Sari, 2010). Diez et al. (2008) indicated that people who consumed fish 5–6 meals per week had twice the level of hair Hg than those who consumed less fish (none or less). Salehi and Esmaili-Sari (2010) also noted that the higher the frequency (>4 times per week) of fish intake, the higher the Hg levels (4.93 $\mu g\, g^{-1}$ on average) in maternal hair. In the present study, there was no significant difference ($p > 0.05$) of hair Hg concentrations between medium and low fish consumption rate groups. This may be caused by the actual amount weight of fish consumption. With the same frequency, males may consume more fish per kg body weight at each meal. Therefore, a survey of the food consumption rate and quantity of each participant in our study was also conducted.

3.3. Food consumption survey

The average food consumption rates for the participants in the four sites are shown in Fig. 1. In general, vegetables (38%), cereal (32%), fish and shellfish (16%) and meat (10%) contributed larger proportions, while milk (3%), fruits (1%) and egg (0.5%) contributed to much smaller proportions to the total consumption of Hg. Among the four sampling sites, participants in SD and ZS were fishing village residents, and therefore the fish consumption rates would be higher when compared to GZ and NH (both town residents). The residents of the present study consumed more fish (15%), cereal (19%) and meat (5%) compared to residents in Zhoushan in China (Cheng et al., 2009), but a lower consumption rate of rice consumption (59%) compared to the residents in Guizhou of China (Zhang et al., 2010).

3.3.1. Hg levels in food

Fig. S5 shows the THg and MeHg levels in fish (fish and shellfish), meat, vegetables and cereal collected from the four sampling sites. The mean concentrations of fish and shellfish for THg and MeHg were 59.0 ± 46.4 (range: 2.81–208.5) and 48.7 ± 38.8 (range:

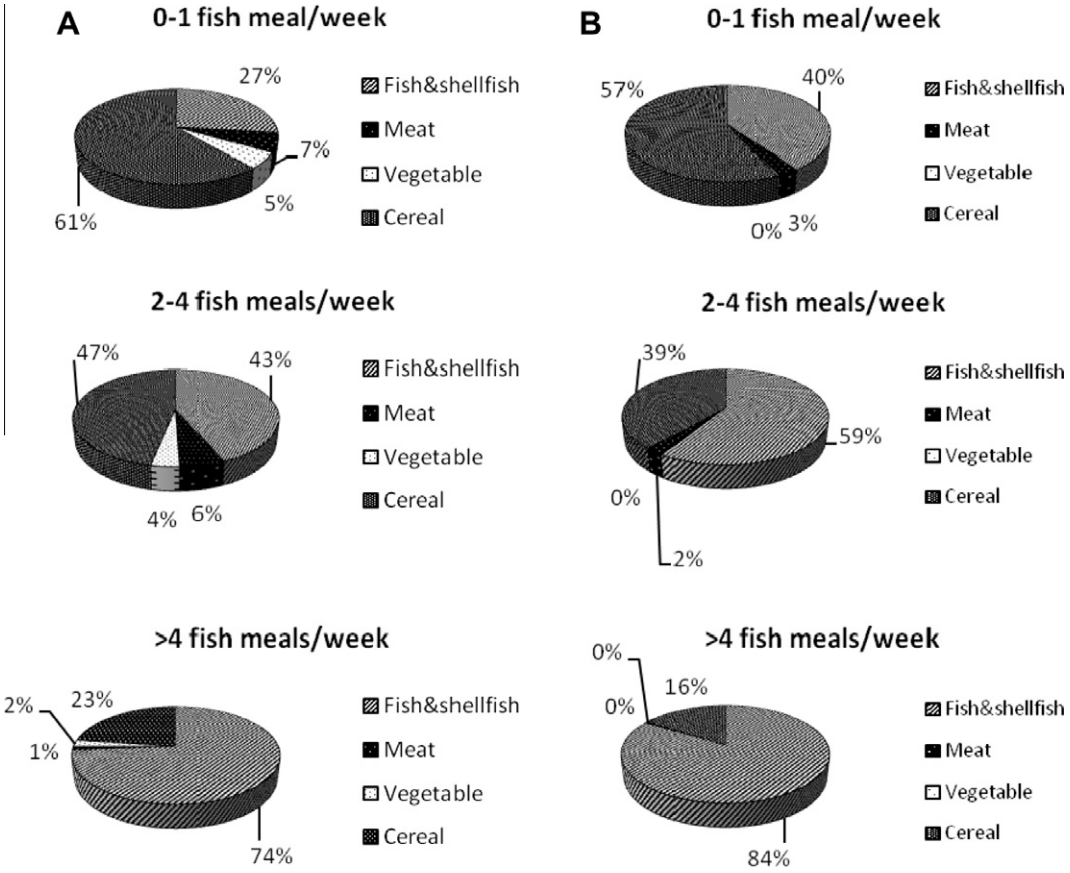

Fig. 3. Percentage of THg intake (A) and MeHg intake (B) from daily dietary intake for participants with different fish consumption rates.

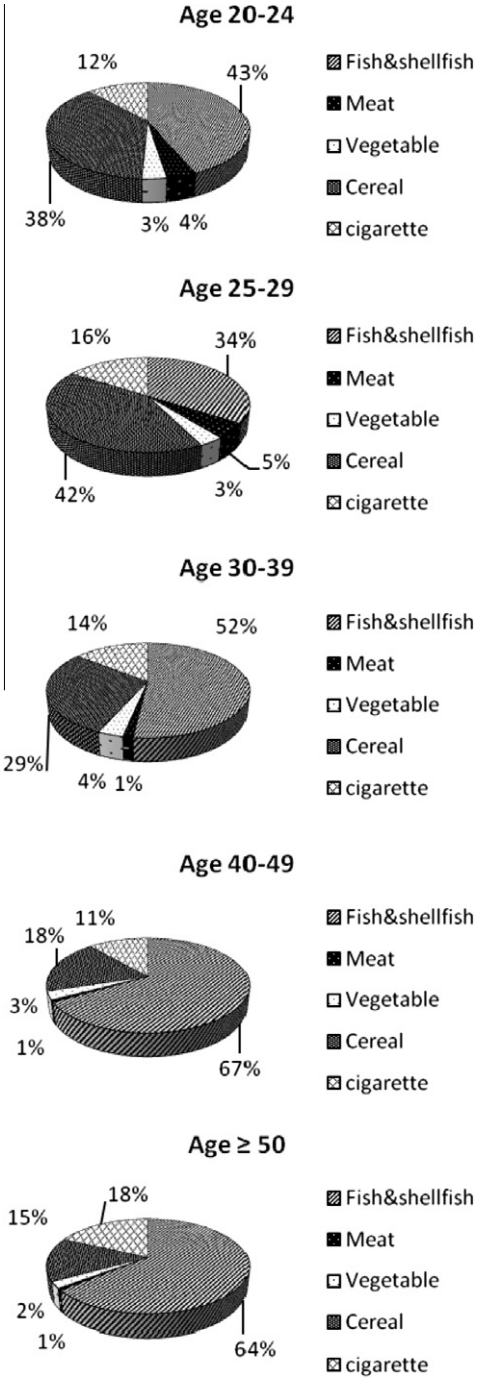

Fig. 4. THg intake via smoking in different age groups.

1.63–287) ng g^{-1} ww, respectively. The highest Hg levels were found in carnivorous fish (such as mandarin fish, largemouth bass and chub), which were 5 times higher than those in crab and mud snail, 10 times higher than those in grass carp. In contrast, the lowest value was detected in shrimp. The THg and MeHg levels in meat (4.85 ± 1.55 ng g^{-1} ww for THg and 2.30 ± 1.02 ng g^{-1} ww for MeHg) and vegetables (1.32 ± 0.57 ng g^{-1} ww for THg and 0.03 ± 0.01 ng g^{-1} ww for MeHg) were lower than those from Zhoushan Island (meat: 29 ng g^{-1} for THg and 16 ng g^{-1} for MeHg, vegetable: 5 ng g^{-1} for THg) (Cheng et al., 2009). The Hg levels in rice and noodle samples are shown in Fig. S5 D, which did not exceed the limits for consumption (0.2 μg g^{-1}) established by the Chinese Standard Agency (GB 2762, 2005), 2005.

3.3.2. Hg levels in cigarette

Fig. S6 indicates the results of absolute THg levels for tobacco in three brands. The mean THg concentration was 35.9 ± 11.2 ng cigarette^{-1}, ranged from 20.9 to 55.9 ng cigarette^{-1}. The guideline of permissible Hg content in cigarette is unavailable. When compared to the results obtained in Poland, the present value was higher (Kowalski & Wierciński, 2009) (2.95–10.2 ng cigarette^{-1}).

3.4. Hg exposure assessment

The common routes of Hg exposure to human included: dust ingestion, dermal absorption, daily air inhalation, drinking water intake and daily dietary intake. However, previous studies reported that dust, dermal, air and water are not the major pathways of Hg exposure when compared to dietary exposure (Horvat et al., 2003; Zhang et al., 2010). Therefore, in the present study, the route of Hg exposure via food consumption was investigated.

The calculated averages of the PDI of THg via food consumption for residents in the four sites around PRD are listed in Table 1. The Hg PDI for the population of SD (0.34 μg kg^{-1} d^{-1} for THg and 0.25 μg kg^{-1} d^{-1} for MeHg) and ZS (0.58 μg kg^{-1} d^{-1} for THg and 0.43 μg kg^{-1} d^{-1} for MeHg) were significantly ($p < 0.05$) higher than those of GZ (0.18 μg kg^{-1} d^{-1} for THg and 0.08 μg kg^{-1} d^{-1} for MeHg) and NH (0.15 μg kg^{-1} d^{-1} for THg and 0.10 μg kg^{-1} d^{-1} for MeHg). The average PDI_{MeHg} in SD and ZS exceeded the PTWI for MeHg of 1.6 μg kg^{-1} $week^{-1}$ (equivalent to 0.23 μg kg^{-1} d^{-1}) (JECFA, 2006). The present study indicated that fish consumption is an important factor of Hg exposure for human. There were significant correlations (THg: $r = 0.48$, $p < 0.01$; MeHg: $r = 0.43$, $p < 0.01$) between Hg in hair and PDI via fish consumption (Fig. 2 A and B). The percentages of estimated THg intake and MeHg intake from daily dietary intake for participants who have different fish consumption rates in PRD are shown in Fig. 3. In the present study, cereal was the main exposure pathway to THg (61%) and MeHg (57%) in the group with a low fish consumption rate. It indicated that rice was the main exposure pathway of Hg for the population who seldom ate fish. These results were in line with the report conducted in Guizhou, China (Zhang et al., 2010). For the group with medium fish consumption rate, both cereal (47% for THg, 39% for MeHg) and fish (43% for THg, 59% for MeHg) were the main source of THg intake. For the group with frequent fish consumption rate, fish and shellfish were the main exposure pathway to THg (74%) and MeHg (84%). The result was consistent with the report conducted in Southern Italy (Diez et al., 2008) and in Taiwan (Chien et al., 2010).

For the participants who had a smoking habit, the mean PDI of THg via smoking cigarette was 0.01 μg kg^{-1} d^{-1}, ranging from 0.002 to 0.024 μg kg^{-1} d^{-1}. There was a positive but not significant ($r = 0.31$, $p > 0.05$) correlation between THg concentration in hair and THg intake via smoking cigarette (Fig. 2C). Fig. 4 shows the contribution from four different foodstuffs and cigarette smoking to the daily intake of THg for smokers in different age groups. The result indicated that besides food consumption, smoking was also an important source for daily THg intake in the smoking group, contributing 11–18% to PDI of THg. Previous studies had found that habitual smoking could facilitate the increase of Hg concentration in human body (Kowalski & Wiercinski, 2007, 2009; Zaborowska & Wierciński, 1999). These results were obtained based on epidemiological approach. However no information was provided on the actual Hg exposure via smoking. A food basket approach was employed in the present study to reveal contribution of smoking to the overall Hg exposure and make comparison with other food. This study is the first to report that smoking could contribute a rather significant proportion of daily THg intake. It indicated that smoking should be considered as a cofactor in Hg exposure. On the other hand, the toxicokinetics of Hg via smoking, such as the

pathway of Hg entering into human body via smoking should be further investigated.

It should be noted that the heterogeneity of the studied population including students, teachers, office workers, farmers and fishermen, could affect the statistical results. Their working environment, possibly related to Hg exposure, were not adjusted in the statistical analyses (two-way ANOVA and regression analyses) due to relative small sample size ($n = 91$) in the present study. Furthermore, the relative small sample size would weaken the statistical power and affect the results. For example, smoking was identified as an important source for daily THg intake in the smoking group, contributing 11–18% to PDI of THg. Whereas, THg concentrations in hair in smoke group were not significant ($r = 0.31$, $p > 0.05$) correlated with THg intake via smoking cigarette, possibly due to the small sample size. The significant difference among the different fish consumption rate groups or between the smoker and non-smoker group were observed, which also needs further investigation to exclude the possibility of false-positive result resulting from small sample size.

4. Conclusion

In the present study, THg and MeHg levels were determined in hair of residents in four areas of PRD. Hair Hg concentrations were mainly dependent on age, fish consumption and smoking habit. The results of Hg PDI calculation showed that fish and cereal consumption were the two main routes of Hg exposure. Besides food intake, smoking was also a major source for daily THg intake in the smoking group.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.foodchem.2012.08.059.

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