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Mercury contamination in fish and human hair from Hainan Island, South China Sea: Implication for human exposure

Jin-Ling Liu [a], Xiang-Rong Xu [a,*], Shen Yu [b], Hefa Cheng [c], Jia-Xi Peng [a,f], Yi-Guo Hong [d], Xin-Bin Feng [e]

[a] Key Laboratory of Tropical Marine Bio-resources and Ecology, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, China
[b] Key Laboratory of Urban Environment and Health, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China
[c] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[d] State Key Laboratory of Tropical Oceanography, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, China
[e] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[f] University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Hair has long been recognized as a good biomarker for human exposure to Hg. The mercury concentrations in 14 species of marine fish and hair samples from 177 coastal residents in Hainan, South China Sea were investigated to assess the status of mercury exposure associated with marine fish consumption. Concentrations of total Hg (THg) and methylmercury (MeHg) in the fish muscles were 0.094 ± 0.008 and 0.066 ± 0.006 μg/g ww, respectively, which were far below the limit considered safe for consumption (0.5 μg/g). The average THg concentrations in hair of adults (1.02 ± 0.92 μg/g) were lower than the provisional tolerable weekly intake (PTWI) level of 2.2 μg/g. However, 23.7% of children had a hair THg level exceeding the RfD level of 1 μg/g, indicating a great risk of Hg exposure to children via fish consumption. The concentration of THg in hair was significantly correlated with fish consumption but not with gender-specific fish intake. With higher fish consumption frequency, the fishermen had significantly elevated hair Hg levels compared to the students and the other general public, who had similar hair THg levels but different fish consumption patterns, indicating the existence of other sources of Hg exposure to the residents of Hainan Island.

1. Introduction

Mercury (Hg), which can be present in water, atmosphere, soils, sediments and organisms in elemental, inorganic and organic forms, is of significant ecological and public health concern. It can be emitted from both natural and anthropogenic sources. Elemental Hg is transported via atmosphere and deposited globally (Cheng and Hu, 2010). By both abiotic and biotic processes, inorganic Hg can be transformed into one of the most toxic forms, organic Hg as methylmercury (MeHg), which is a well-documented neurotoxin with adverse neurological and developmental effects on humans and wildlife (Clarkson and Magos, 2006). Hazards of MeHg are often exacerbated by bioaccumulation and biomagnification via food webs in aquatic systems (Kidd et al., 2012).

Besides occupational exposure, fish consumption is generally considered as a main pathway of MeHg exposure to humans (USEPA, 1997; USFDA, 2004; Cheng and Hu, 2012). Mercury level in hair has been widely used as a reliable biomarker for assessing human Hg exposure (NRC, 2000; Lincoln et al., 2011). Generally, the level of Hg in hair is thought to be 150 to 200 folds higher than that in blood (USEPA, 1997; Gill et al., 2002). Moreover, hair Hg represents a time record of the average Hg exposure over the growth period of the hair, based on the typical hair growth rate of 1 – 1.5 cm per month (NRC, 2000). Approximately 80% to 98% of Hg in human hair is MeHg (Dolbec et al., 2001; McDowell et al., 2004) and total Hg (THg) in hair is consistently correlated with MeHg in hair (Li et al., 2010). Previous studies have shown the positive relationship between fish consumption and Hg levels in hair (Díez et al., 2008; Black et al., 2011). It has been found that human hair Hg levels were linearly related to fish consumption quantity (Chien et al., 2010) and consumption of a wide variety of sea fish, in particular, the locally caught fish, could remarkably elevate hair Hg level (Lincoln et al., 2011). Miklavčič et al. (2011) also reported

* Corresponding author. Fax: +86 20 89100753.
E-mail address: xuxr@scsio.ac.cn (X.-R. Xu).

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that THg level in hair was a suitable biomarker of low-level Hg exposure through fish consumption.

Hainan Island is located in the South China Sea (SCS) and is the second largest island in China. Fishing is the major economic activity for the coastal residents on the island. The coastal residents, especially fishermen in Hainan prefer to capture and consume wild fish from SCS. In our recent study, carnivorous fish were found to have higher levels of THg, MeHg, and MeHg/THg ratios than omnivorous and herbivorous fish. High Hg levels in fish of the SCS were probably related to Hg input from atmospheric deposition and anthropogenic activities (Wang et al., 2013a; Liu et al., 2014). The previous studies have shown that coastal populations commonly exhibited higher levels of Hg in hair than inland populations, most likely as a result of higher fish consumption (Knobeloch et al., 2005; Mahaffey, 2005). Fish (e.g. Bleeker's grouper and Bigeye) from Hainan province were previously found to contain higher levels of MeHg compared to those from other producing areas (Wang et al., 2013b). This study was conducted to investigate the status of Hg exposure associated with fish consumption to the residents in coastal areas of Hainan, SCS. The influence of gender, age, body weight, height, profession of residents on the THg level in human hair was also assessed.

2. Materials and methods

2.1. Questionnaire survey

In March and April 2012, a total of 177 residents (aged 5 – 82) living in coastal area along east Hainan (Fig. 1) were invited to participate in this survey on a voluntary basis. The survey group consisted of 94 males and 83 females. Questionnaire on gender, age, height, weight, profession, hair treatment, type and frequency of fish consumption, and health status was collected for each individual. The detail information could be found in Table 1.

2.2. Sample collection and treatment

The hair samples were taken from several sites of the scalp from each participant using stainless steel scissors, placed and sealed in clean polyethylene bags. Hair samples with length of 3 cm were cut into short segments and sequentially washed with nonionic detergent, acetone, and distilled water, then dried in an oven at 60 °C overnight (Li et al., 2009).

The fish samples, including fourteen marine fish species (n=96), which represented the fish frequently consumed by local population, were purchased from the local fishermen (Table 2). Fish samples were placed in plastic bags and transported to the laboratory in a freezer and then immediately frozen at −20°C until dissection. After weight and length of the fish were measured, the dorsal muscles were taken, homogenized, freeze-dried, crushed and ground into fine powder.

2.3. Analytical procedure and QA/QC

THg levels in the hair and fish samples were measured following the method described in the previous study (Li et al., 2009). Briefly, hair and fish samples were digested by freshly mixed acids of HNO_3/H_2SO_4 (v/v=4:1) in a water bath (95 °C) and subsequently processed with BrCl oxidation, $SnCl_2$ reduction, gold trap, and cold vapor atomic fluorescence spectrometry (CVAFS) determination. MeHg concentrations in the fish muscles were measured as described elsewhere (Liang et al., 1996). Approximately 0.1–0.2 g freeze-dried fish sample was digested with 5 mL KOH solution (20%) for 3 h in water bath (75 ± 3 °C), followed by ethylation, purge and trap onto Tenax traps, isothermal GC separation and CVAFS detection.

Method blanks, blank spikes, matrix spike, certified reference material, and blind duplicates were carried out for quality control of sample analysis. A mean THg concentration of 4.4 ± 0.1 μg/g (n=10) was found for the certified reference material of hair sample (NIES-13) with a certified value of 4.4 ± 0.2 μg/g. The mean values of THg and MeHg determined for the certified reference material of fish samples (TORT-2, NRCC, Canada) were 0.26 ± 0.07 and 0.16 ± 0.012 μg/g, respectively (n=20), which were in good agreement with the certified reference values (0.27 ± 0.06 and 0.152 ± 0.013 μg/g for THg and MeHg, respectively). The relative percentage differences were less than 10% for THg and MeHg in duplicate samples of hair and fish.

Table 1
Demographic characteristics of the surveyed population.

Characteristic	All subjects n=177	Male n=94	Female n=83
Age (years)	5 – 82	5 – 82	5 – 79
Body weight (kg)	15 – 96	20 – 81.5	15 – 96
Height (cm)	95 – 178	100 – 178	95 – 176
Fish consumption (meals/week)			
< 1 (%)	4	5	2
1 – 2 (%)	39	41	36
3 – 5 (%)	34	24	45
> 5 (%)	23	29	17

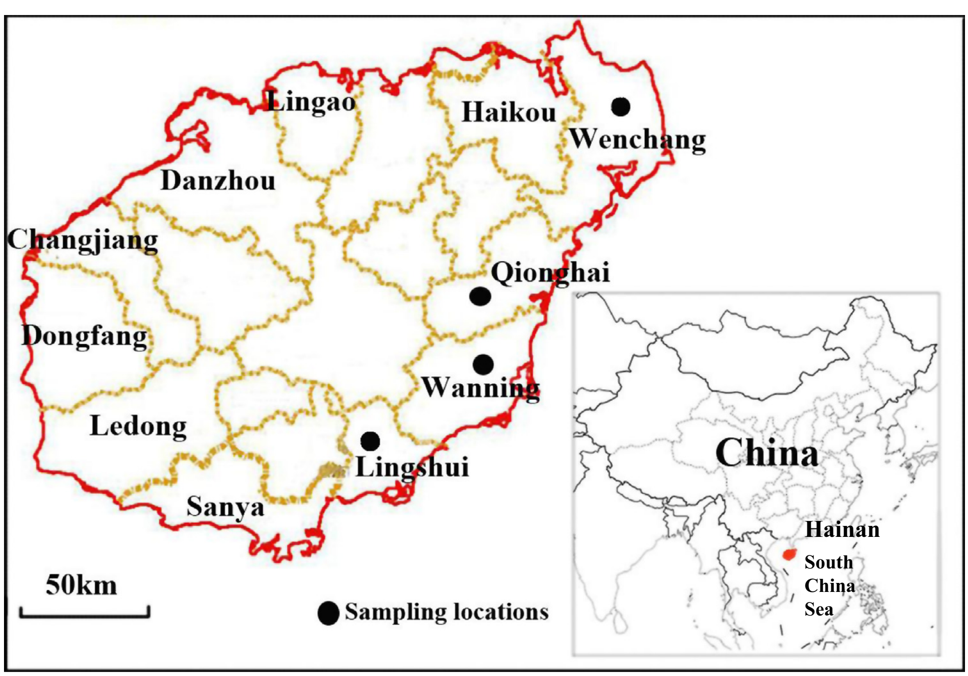

Fig. 1. Study area and sampling locations.

Table 2
Concentrations of THg, MeHg and % of THg as MeHg in fish samples from Hainan, SCS.

Species	English name	*n*	THg ($\mu g\ g^{-1}$)		MeHg ($\mu g\ g^{-1}$)		% MeHg median
			Mean	Range	Mean	Range	
Carnivorous							
Lutjanus erythropterus	Crimson snapper	8	0.081	0.043 – 0.162	0.071	0.030 – 0.154	83
Parargyrops edita	Crimson sea bream	10	0.074	0.052 – 0.088	0.047	0.026 – 0.065	65
Trachurus japonicus	Japanese scad	4	0.081	0.061 – 0.094	0.076	0.059 – 0.086	94
Acanthopagrus berda	Goldsilk seabream	10	0.217	0.021 – 0.293	0.153	0.019 – 0.228	74
Nemipterus virgatus	Golden threadfin bream	4	0.171	0.126 – 0.215	0.128	0.08 – 0.175	73
Scorpaena neglecta	Stonefish	4	0.147	0.085 – 0.209	0.091	0.056 – 0.127	63
Terapon jarbua	Jarbua terapon	4	0.223	0.154 – 0.291	0.165	0.097 – 0.234	71
Epinephelus areolatus	Spotted grouper	4	0.115	0.104 – 0.126	0.060	0.044 – 0.076	51
Cynoglossus robustus	Speckled tongue sole	8	0.071	0.021 – 0.187	0.041	0.014 – 0.097	64
Chorinemus lysan	Largemouthed queenfish	9	0.113	0.068 – 0.152	0.072	0.046 – 0.093	65
Omnivorous							
Ephippus orbis	Orbfish	11	0.043	0.021 – 0.146	0.032	0.016 – 0.093	79
Navodon xanthopterus	Yellowfin filefish	6	0.036	0.014 – 0.061	0.025	0.01 – 0.04	70
Siganus fuscessens	Rabbitfish	4	0.010	0.01 – 0.01	0.009	0.009 – 0.009	91
Herbivorous							
Siganus guttatus	Spinefoot	10	0.015	0.008 – 0.028	0.011	0.004 – 0.022	75
All species		96	0.094	0.008 – 0.293	0.066	0.004 – 0.234	72.7

2.4. Statistical analyses

All statistical analyses were performed using SPSS 16.0 for Windows (SPSS Inc., Chicago, IL, USA). Mercury concentrations in the fish and hair samples were tested for normality and were found to be not normally distributed. Therefore, log-transformed data were used for analysis. Differences between the groups were tested for significance using one-way ANOVA tests. Bivariate relationships between different variables were determined using Pearson correlation test. *P*-values less than 0.05 were considered statistically significant. Unless otherwise stated, mercury concentrations are expressed in μg/g wet weight (ww) for fish and μg/g dry weight (dw) for hair.

3. Results and discussion

3.1. Mercury concentrations in fish from coastal areas of Hainan

The determination results of Hg in muscles of the collected fish samples are shown in Table 2. The average THg and MeHg levels in the dietary marine fish muscles were 0.094 ± 0.008 and 0.066 ± 0.006 μg/g, respectively. MeHg constituted large proportions of THg with a median of 72.7% (51 – 94%), which is comparable to the findings of other studies (Ikingura and Akagi, 2003; Carrasco et al., 2011). A significant positive correlation was found between THg and MeHg concentrations in the fish samples (R=0.97, $p<0.01$), which is similar to the observation of Wang et al. (2013b). As expected, species-specific differences were found in THg concentrations ($p<0.01$). Carnivorous fish had higher concentrations of THg than omnivorous and herbivorous fish ($p<0.01$), which is consistent with biomagnification of Hg in aquatic food webs (Cheng and Hu, 2012; Li et al., 2013).

3.2. Risk assessment of Hg exposure due to fish consumption in Hainan residents

To protect human health against MeHg exposure, USFDA (2004) warned women who are planning to conceive, pregnant or breast-feeding, as well as their children to avoid consuming the predatory fish with Hg concentration exceeding 1 μg/g ww. The health guideline in China set the safety limit of MeHg in predatory fish and other fish at 1.0 μg/g and 0.5 μg/g, respectively (SAC, 2012). No observed adversary effect level (NOAEL) of 50 μg/g THg in hair for adult was concluded in an early WHO report (WHO, 1990). In order to protect the developing fetus and children from neurological deficits induced by MeHg, a reference dose (RfD) for MeHg was developed as 0.1 μg/kg body weight/day by USEPA (1997) and a provisional tolerable weekly intake (PTWI) of 1.6 μg/kg body weight/week was established (JECFA, 2003). The intake of MeHg at PTWI corresponds to a hair THg concentration of 2.2 μg/g and RfD corresponds to a hair level of 1.0 μg/g (Yasutake et al., 2004). In this study, PTWI and RfD values with their corresponding hair THg levels were used to estimate the risk of MeHg exposure due to fish consumption for the coastal residents of Hainan Island.

Concentrations of MeHg in the fish from Hainan were substantially below the limit considered safe for consumption (0.5 g/g ww, SAC, 2012). In this study, the fish intake of 177 persons, including 94 children (51 males and 43 females) and 83 adults (43 males and 40 females), were surveyed. Based on the survey results, the medians of fish consumption frequency and weight were 3, 3, 3, 3.5 meals/week and 53, 52, 55, 60 kg for boys, girls, men, and women, respectively. Assuming a standard fish portion of 100 g (CNS, 2010), with an average MeHg concentration of 0.066 μg/g obtained in this study, the average daily intakes of MeHg would be 0.374, 0.381, 0.360, and 0.385 μg/kg body weight/week for boys, girls, men, and women, respectively. The MeHg intakes of children, including boys and girls, were 3 times higher than the RfD (0.1 μg/kg/week) of MeHg. These results indicate there is a high risk of MeHg exposure via fish consumption for children. In contrast, even if the adults consumed 7 meals of fish each week, the MeHg intakes of adults would be still below the PTWI (1.6 μg/kg/week).

Since children are especially sensitive to MeHg and a high risk of MeHg exposure via fish consumption exists for the children in coastal Hainan, more detailed assessment of fish intake and hair THg levels is needed. In fact, almost one quarter (23.7%) of the children had a hair THg level exceeding the RfD level of 1 μg/g and three children had hair THg levels above 2.2 μg/g. According to the questionnaire, children with THg levels above 1.0 μg/g ate marine fish more than 3 times per week. The ages of these children were 16 and 17 years old, except one child (girl) at the age of 5 years old. Results of Pearson correlation analysis indicate a significantly positive correlation between hair THg levels of children and their fish consumption (R=0.291, $p<0.001$, n=94). Therefore,

Table 3
Concentrations of THg (μg/g) in the hair of coastal residents from Hainan, SCS.

Concentrations of THg in hair of coastal residents	Children 5–17 years		Adults 18–82 years	
	Male	Female	Male	Female
N	51	43	43	40
Mean	0.813	0.794	1.123	0.916
Median	0.679	0.704	0.689	0.545
Range	0.06–4.20	0.06–3.87	0.24–4.81	0.18–3.02
Standard deviation	0.68	0.60	1.03	0.75
95% Confidence interval	0.619–1.007	0.608–0.979	0.803–1.44	0.674–1.158

the dietary consumption of the marine fish is probably a major route of Hg exposure for the children in coastal Hainan, SCS.

3.3. Hair mercury concentrations and influencing factors

The statistics of THg concentrations in the hair of Hainan coastal residents are summarized in Table 3. Overall, the THg concentration in hair ($n=177$) ranged from 0.057 to 4.81 μg/g, with a median of 0.677 μg/g and a mean of 0.907 μg/g. The average THg concentrations in hair of children ($n=94$) were 0.804 ± 0.648 μg/g (geometric mean and range were 0.632 and $0.06-4.20$ μg/g), which were lower than the corresponding values RfD (1.0 μg/g). Meanwhile, the average THg concentrations in hair of adult ($n=83$) were 1.02 ± 0.915 μg/g (geometric mean and range were 0.755 and $0.18-4.81$ μg/g), which were lower than the corresponding values of PTWI (2.2 μg/g) based on the neurotoxicological data.

Compared to other exposed populations in China, the mean concentration of THg in the hair of coastal community in Hainan (0.907 μg/g) was far below that of the inland residents from Wanshan mercury mining area ($3.3-5.5$ μg/g), where artisanal Hg mining activities and rice consumption were main Hg exposure pathways (Li et al., 2009). The level was comparable to the hair Hg concentrations of residents from other coastal areas in China (Shanghai 0.5 μg/g, Xiamen 0.8 μg/g, Ningbo 1.0 μg/g; Liu et al., 2008; Pearl River Delta 1.08 μg/g; Shao et al., 2013), which were mainly related to fish consumption, and higher than those of inland urban residents who had a low fish consumption frequency. For example, concentration of THg in the hair of urban residents from Changchun was 0.448 μg/g (Li et al., 2006). These results demonstrate that fish consumption could be an important Hg exposure route for coastal residents in South China.

3.3.1. Gender, age, height and weight

For children, no significant difference in hair THg concentrations between girls and boys ($F=0.597$, $p>0.05$, $n=94$) was found. For adults, there was also no significant difference in hair THg concentrations between women and men ($F=0.07$, $p>0.05$, $n=83$). These results suggest there was no gender-specific accumulation of Hg in the hair of adults and children. Similar findings were reported in previous studies (Kosatsky et al., 2000; Olivero et al., 2002). However, other previous studies found that males had higher mean values of Hg concentrations in hair than females in Italy and Spain (Díez et al., 2008, 2011). These gender-related differences might be related to different consumption patterns or hair treatments of males and females (Burger, 2005; Knobeloch et al., 2007; Díez et al., 2011). Besides, difference in the metabolization of MeHg between males and females could also play a role in the gender-related difference (Díez et al., 2008).

No significant correlation was found between height and hair Hg concentration ($R=-0.022$, $p>0.05$), as well as weight and hair Hg concentration ($R=0.094$, $p>0.05$). These results are consistent with those of a previous study on Hg exposure of pregnant women via fish consumption by Salehi and Esmaili-Sari (2010). However,

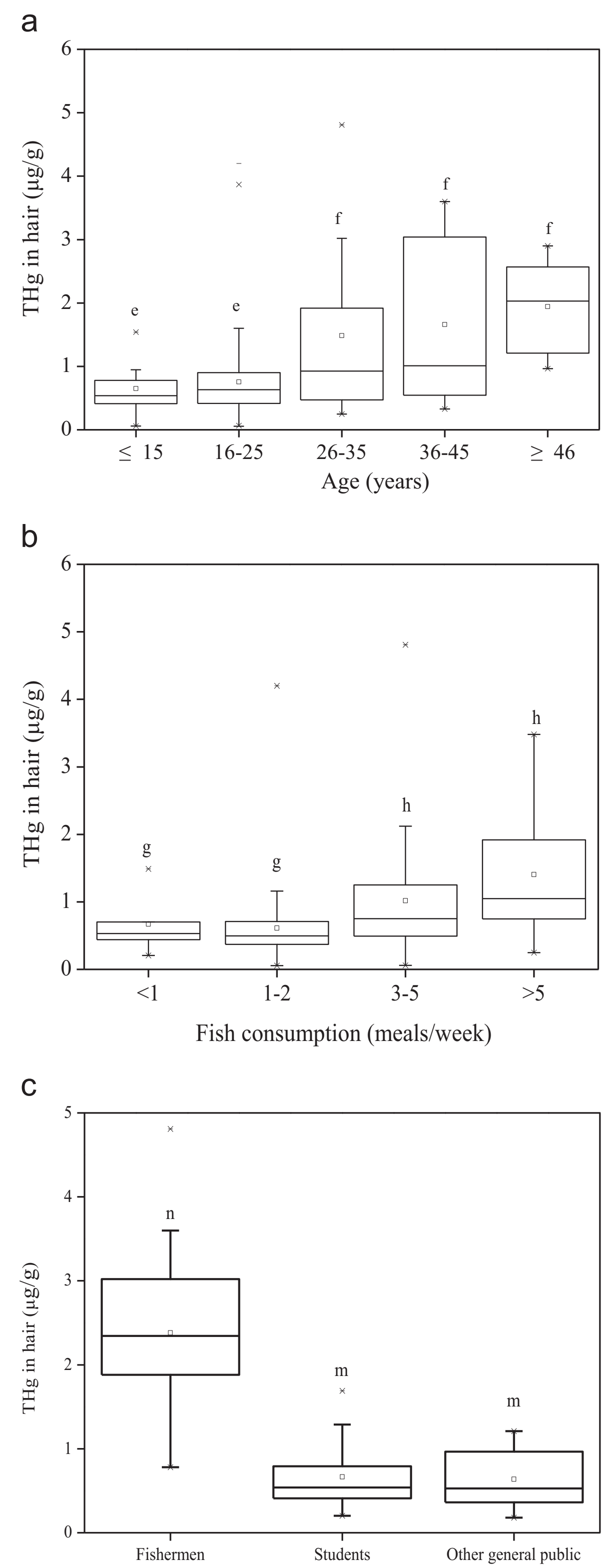

Fig. 2. Concentrations of THg in the human hair as a function of (a) age; (b) fish consumption; (c) profession. The population size for each group is as following: students ($n=42$), fishermen ($n=18$), other general public ($n=23$). Different letters represent statistically significant difference of means ($p<0.05$). Each box represents the interquartile range (25th to 75th percentile), while the horizontal band near the middle of the box is the 50th percentile (the median), and the whiskers represent the 5th and 95th percentiles.

a negative correlation between hair Hg content and body weight was found elsewhere (Babi et al., 2000), whereas a positive correlation between THg concentration in hair and body weight was found by Díez et al. (2008). These findings suggested there was no definite relationship between body weight and hair Hg concentration.

The hair Hg concentrations stratified in accordance with the age are presented in Fig. 2a, which shows an obvious increase in hair Hg with age. Pearson correlation analysis indicated that there was a statistically significant association between age and hair Hg concentration ($R=0.315$, $p<0.001$, $n=177$), which is consistent with previous studies (Zolfaghari et al., 2007; Chien et al., 2010). It should be noted that a positive correlation was also found between age and fish consumption ($R=0.294$, $p<0.001$, $n=177$). It implies that older people consumed more fish than the younger. Fish consumption is an important factor causing the age effect on the hair Hg level.

3.3.2. *Fish consumption*

Based on their fish consumption rate, the study subjects were separated into four groups (<1 meal/week, 1–2 meals/week, 3–5 meals/week, >5 meals/week) to identify the differences in hair Hg concentration. The percentages of different groups were shown in Table 1. In the past 3 months, 4% of the participants ate fish less than once per week, 73% ate fish between one and five times per week, 23% ate fish more than five times per week. Based on these data, the average diet was estimated to be 3.5 meals of fish per week, which is an indicator of rich fish consumption for the general population in coastal areas of Hainan. As shown in Fig. 2b, there was an obvious increasing trend in mean hair THg concentration with fish consumption frequency. Those who ate fish more than five times per week had almost two times higher hair Hg concentrations than those with less than three times per week ($p<0.05$). Significant differences ($p<0.05$) in hair THg concentrations were found between the four groups and a significant positive correlation ($R=0.401$, $p<0.001$, $n=174$) was also found between the consumption of fish and THg in the whole group. Similarly, Knobeloch et al. (2007) found a positive correlation between monthly fish meals and hair Hg levels in Wisconsin. Díez et al. (2011) also found a significant effect of fish consumption on Hg concentrations in the hair of inhabitants in Spain. The significant effect of fish consumption on Hg levels in hair indicates that dietary Hg exposure could be reflected by hair mercury levels of the populations.

3.3.3. *Fishermen, students, and other general public*

Based on the profession, all of the 83 adults in this study were divided into three groups: fishermen ($n=18$), students ($n=42$), and other general public ($n=23$). Fishermen is the subpopulation predicted to represent a critical population for Hg exposure via fish consumption due to frequent and high quantity of fish consumed (Al-Majed and Preston, 2000; Cheng et al., 2009). Most of the participants in this study were students who consumed relative narrow variety of fish (e.g., crimson sea bream and Japanese scad).

As shown in Fig. 2c, 11% of the fishermen had hair THg level lower than 1 μg/g, 61% had hair THg between 1 and 3 μg/g, while the rest had hair THg between 3 and 5 μg/g. The hair THg concentration of fishermen (mean 2.38 μg/g; median 2.35 μg/g) in Hainan was approximately four times higher than that of Italian fishermen (Naples 0.6 μg/g of mean; Díez et al., 2008), and two times higher than the median of high fish-consuming recreational anglers in the USA (Louisiana 0.81 μg/g of median; Lincoln et al., 2011) and Canada (Montreal 0.82 μg/g of median; Kosatsky et al., 2000). This may be explained by the high fish consumption frequency (mean: 6.5 meals/week) of fishermen in Hainan. Nonetheless, the THg levels in hair of the fishermen in this study were still far below the NOAEL (50 μg/g) (WHO, 1990). It should be noted that the fisherman who consumed relatively large amount of fish were not at risk in Hainan Island, although other factors such as demographic characteristics, hair growth rate, nutritional status, toxicokinetics, and exogenous contamination, could not be excluded. Meanwhile, the THg levels in hair of the fishermen were close to the corresponding level of PTWI (2.2 μg/g) based on the ecotoxicological data, indicating a potential risk of the fisherman who consumed larger amount of fish.

Compared to the students and other general public, fishermen in this study had three to four times greater THg contents in hair (Fig. 2c), indicating the most severe Hg exposure. This could be explained by several factors. Firstly, demographic differences might contribute to this difference. In this study, the fishermen group consisted of 9 men (21–70 years old) and 9 women (31–82 years old) who lived in Qionghai. Students group consisted of 27 males (18–19 years old) and 15 females (18–19 years old), while the other general public group consisted of 7 males (18–35 years old) and 16 females (20–79 years old). Secondly, fish consumption frequency of different groups was largely different. The mean fish consumption rates were 2.5, 6.5, and 4.13 meals per week for students, fishermen, and the other general public, respectively. Thus the high frequency of fish consumption could be an important factor contributing to the high Hg exposure of the fishermen. Thirdly, the fish consumption patterns of fishermen differed considerably from the general public. In general, fishermen consumed a wide variety of fish, particularly locally caught fish which might exhibit comparatively high MeHg concentrations (Lincoln et al., 2011). These results supported the findings of earlier studies, which showed that hair Hg level of subjects with no occupational exposure was mainly dependent on fish consumption (Al-Majed and Preston, 2000; Olivero et al., 2002; Cheng et al., 2009; Lincoln et al., 2011).

The other two groups, students and other general public had comparable concentrations of THg levels in hair (students 0.75 μg/g; the other general public 0.64 μg/g; $p>0.05$), but significant difference in fish consumption rates (students 2.92 meals/week; the general public 4.13 meals/week; $p<0.05$). Consumption rate would not explain the hair Hg level alone. Amount of fish each meal will be one of the major issues as well. In a recent study, Shao et al. (2013) found that besides fish, cereal consumption and smoking were important sources for Hg intake to the residents from the Pearl River Delta. Therefore, other potential routes of Hg exposure exist for residents in Hainan, which need further study.

4. Conclusions

Concentrations of THg and MeHg in the fish muscles from Hainan Island, SCS, were far below the limit considered safe for consumption (0.5 μg/g). The hair THg concentrations of coastal residents in Hainan were significantly related to age and fish consumption. The average THg concentrations in hair of children and adults were lower than the corresponding values of RfD and PTWI, respectively. However, almost one quarter of the children had a hair THg level that exceeded the RfD level and the estimated MeHg intakes of children were above the RfD level. It implied a significant risk of MeHg exposure to children via fish consumption, rather than adults in Hainan Island. The local fishermen had significantly elevated hair Hg levels compared to students and the other general public, due to their higher fish consumption frequency. The students and the other general public had comparable THg levels in their hair but different fish consumption patterns, suggesting that other factors and/or other Hg exposure sources also played important roles.

Acknowledgments

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