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

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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

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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

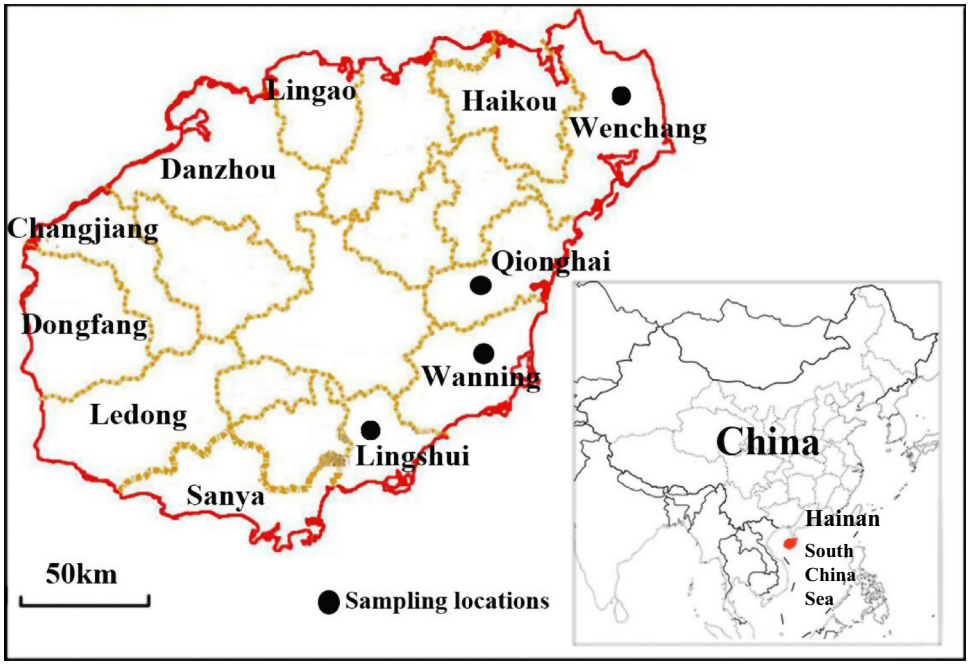

Fig. 1. Study area and sampling locations.