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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