

MERCURY IN THE SEAFOOD AND HUMAN EXPOSURE IN COASTAL AREA OF GUANGDONG PROVINCE, SOUTH CHINA

PING LI, \dagger XINBIN FENG, $*$ PENG LIANG, \dagger HING MAN CHAN, \dagger HAIYU YAN, \dagger and LAIGUO CHEN§

yState Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, China zCentre for Advanced Research in Environmental Genomics, University of Ottawa, Ottawa, Canada

§Center for Research on Urban Environment, South China Institute of Environmental Sciences, Ministry of Environmental Protection, Guangzhou, China

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Abstract—Consumption of fish and aquatic products is considered to be the main pathway of human exposure to methylmercury (MeHg). To assess human health risk through seafood consumption in a coastal area of Guangdong Province, South China, a total of 518 seafood samples (including fish, shrimp, crabs, and mollusks) were collected from 11 coastal cities for total mercury (THg) analysis. The THg concentrations varied significantly among different seafood groups and ranged from 0.11 to 317 ng/g (with a mean of 37.2 ng/g). The THg concentrations were relatively low compared with fish from Europe and North America, and no samples exceeded the national limit recommended by the Standardization Administration of China $(0.5 \mu g/g$ wet wt). The ecological functional groups and cultured styles have significant effects on THg concentrations in the fish tissue. The median of probable daily intakes (PDIs) of MeHg via seafood consumption ranged from 42.6 to 71.4 ng/kg/d for different age groups, with fish contributing the major portion (84%). The results indicated potential health risks associated with seafood consumption for the general population in the coastal area of Guangdong Province, South China. Environ. Toxicol. Chem. 2013;32:541-547. © 2012 SETAC

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INTRODUCTION

Human exposure to methylmercury (MeHg) occurs mainly through consuming fish. Methylmercury contamination in fish is a worldwide environmental concern, because fish contain high-quality protein and other essential nutrients with known benefits to human health [1]. Fish is an excellent source of omega-3 fatty acids [2], and balancing the risks and benefits has become an increasingly important goal of fish consumption advisories [3]. The MeHg can accumulate to high concentrations in the tissues of fish through the aquatic food web. The MeHG is confirmed to bind to the proteins in fish tissues, which are not removed through any cooking or cleaning processes [4]. Various consumption advisories on total mercury (THg) concentrations in fish recommended by different organizations normally range between 0.3 and 0.5 μ g/g wet weight [5–7]. Using the benchmark dose lower limit (BMDL), the Joint Expert Committee on Food Additives (JECFA) of the Food and Agriculture Organization and the World Health Organization established a provisional tolerable weekly intake (PTWI) for MeHg of $1.6 \mu g/kg$ /week [8]. In 1997, the U.S. Environmental Protection Agency (U.S. EPA) set the daily limit of 0.1μ g/kg/d as a reference dose (RfD) for MeHg [9].

With its rapid economic growth, China is considered to be the largest anthropogenic source of mercury (Hg) emission in the world [10]. It contributed about 33% of the global emissions, with an emission of 635 metric tons (t) in 2005 [10]. Coal combustion and non-ferrous metals smelting are the most important anthropogenic sources [11]. Guangdong is one of the most economically developed areas in China, contributing approximately 11.6% of China's national economic output in 2009 [12]. Meanwhile, it was the second largest province of anthropogenic Hg emission in 1999 (44.2 t) [11]. Serious Hg pollution resulting from industrial activities was observed in the sediments of the Dongjiang River [13] and the Pearl River Estuary [14].

China has been the world's largest producer and exporter of fishery products since 2002. Seafood products including fish, shrimp, crabs, and mollusks are farmed extensively in the coastal region, which has become an important production zone to support exporting activities. The main trade partners include Japan, Korea, Canada, the United States, and the European Union. Guangdong Province was the second largest province of aquatic production, with an output of 7.03 million t in 2009, accounting for 13.8% of the total national production [12]. Because a large quantity of seafood products are produced in and exported from this region, mercury pollution in the seafood should be a concern not only in South China, but also around the globe.

The objective of the present study was to investigate Hg pollution in seafood collected from the coastal area of the Guangdong Province through a comprehensive market basket survey. The data acquired enabled us to perform risk assessments, which could provide critical information to establish seafood consumption advisories. In companion papers, the dietary intake of polybrominated diphenyl ethers (PBDEs) [15], dichlorodiphenyltrichloroethane (DDT) [16], and organochlorine pesticides [17] through seafood consumption and related health risks were examined.

MATERIALS AND METHODS

Sample collection

Seafood samples were collected based on a market basket survey. A total of 518 samples—including 13 species of fish, six species of shrimp, two species of crabs, and 14 species of

^{*} To whom correspondence may be addressed

⁽fengxinbin@vip.skleg.cn).

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Table 1. Basic characteristics of the seafood samples

^a Omnivorous fish.

b Herbivorous fish.

 \degree Carnivorous fish.

THg = total mercury; FWA = fish from freshwater aquaculture; $MC =$ fish from mariculture; OF = fish from ocean fishery.

mollusks (Table 1)—were collected in June and October 2005. The samples were collected from local fishery product markets in 11 coastal cities of Guangdong, South China, including three western cities (Zhanjiang, Maoming, and Yangjiang), six cities from the Pearl River Delta (Dongguan, Foshan, Guangzhou, Jiangmen, Zhuhai, and Zhongshan), and two eastern cities (Shantou and Shanwei) (Fig. 1). The production of aquatic products in these 11 cities accounts for about 80% of the production of aquatic products for all of Guangdong Province. To ensure the appropriate representation in the sampling, the biggest aquatic product markets in each city were selected for the present study. Seafood sample types were selected based on geographical distribution, commercial availability, seafood production, and consuming habits. Approximately 30 individuals for each fish species and 10 individuals of shrimp, crabs, and mollusks were randomly acquired in local fish markets to ensure sufficient statistical power for data analysis. Immediately following collection of the samples, approximately 20 g (wet wt) of muscle or soft tissue were taken and stored in polyethylene bags, kept in ice, and returned to the laboratory. The seafood samples were stored at -20° C until analysis. The fish sampling strictly followed the U.S. EPA Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories [18].

Analytical method and quality control

About 0.5 to 1 g of the fresh seafood samples were digested in a water bath (95 $^{\circ}$ C) with a fresh mixed acid of HNO₃:H₂SO₄ $(4:1 \text{ v/v})$ for THg analysis and determined by SnCl₂ reduction, purge, gold trap, and cold vapor atomic fluorescence spectrometry [19]. The seafood samples were analyzed individually for THg concentrations. Quality control consisted of method blanks, certified reference material, and blind duplicates. The limit of determination was 0.013 ng/g for THg in the seafood samples. The average THg concentration of the certified reference material (National Research Council Canada TORT-2) was 0.27 ± 0.01 μ g/g, which was comparable with the certified value of $0.27 \pm 0.03 \,\mu\text{g/g}$. The relative standard deviation was lower than 10% for THg in duplicate samples.

Dietary intake and risk assessment

A questionnaire-based dietary survey was conducted in the 11 sampling cities (Table 2). Dietary data were collected through face-to-face interviews with 1,527 local residents, and 12 food categories (fish, shrimp, crab, mollusks, meat, egg, vegetables, beans, milk, fruit, rice, and wheat products) were included in the survey. Information on the age, gender, and body weight of each interviewee also was collected. All

Fig. 1. Spatial distribution of the sampling sites in Guangdong, South China.

participants were required to sign a consent form. Ethics approval for the present study was obtained from the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences.

Because MeHg constitutes the major portion of THg in fish tissue in China, generally about 80% (50–87%, with a mean of 68% [20]; 66.3–80.8% [21]; 75–100% [22]; 58.6–83.6%, with a mean of 73.8% [23]), we assumed that 80% of THg in the fish samples was in the form of MeHg. For the invertebrates (shrimp, crabs, mollusks), we hypothesized the percentage of THg as MeHg was 50%, because the proportion of MeHg to THg was averaged at 43% in mollusks along the French coast (with a range of 11–88%) [24].

To estimate MeHg intake from seafood consumption, the probable daily intake (PDI) for different age groups was calculated according to the following formula

$$
PDI = \sum (C_i \times IR_i)/bw
$$

where PDI is given in nanogram per kilogram of body weight per day (ng/kg/d); i is fish from freshwater aquaculture (FWA), mariculture (MC), ocean fishery (OF), shrimp and crab (S&C),

Table 2. Daily seafood intake for different age groups in coastal area of Guangdong, South China (g/d)

Age		$6 - 17$		18-44		$45 - 59$		> 60	
Gender	М	F	M	F	М	F	М	F	
n	204	194	495	490	48	39	28	29	
FWA МC OF S&C Mo Total	25.0 24.5 17.5 53.7 26.8 148	24.8 17.5 12.5 33.2 17.5 106	37.5 24.5 17.5 34.1 19.3 133	24.8 17.5 17.5 31.0 17.5 108	37.5 24.5 17.5 26.3 17.5 123	62.5 25.0 12.5 42.5 19.8 162	41.3 24.8 17.5 29.0 24.5 137	62.5 24.8 10.5 24.4 17.5 140	

 $M = male$; F $=$ female; FWA $=$ fish from freshwater aquaculture; $MC = fish$ from mariculture; OF = fish from ocean fishery; $S\&C =$ shrimp and crab; $Mo =$ mollusks.

or mollusk (Mo); bw is body weight; C is MeHg concentration in the seafood (ng/g) ; and IR is the daily intake rate of the seafood (g/d).

The hazard quotient (HQ) is simply the ratio of the estimated exposure to an effect concentration. If the value of the HQ was less than 1, it was considered to indicate acceptable risk. In the present study, HQ was calculated by dividing the PDIs by the guideline level $(0.23 \mu g/kg/d)$ recommended by the JECFA [8].

Statistical method

All data were analyzed using SPSS 11.5 for Windows. The basic features of the data were described as mean, standard deviation (SD), and median and examined by descriptive statistics. Because the fish THg data were in log-normal distribution, the data were transformed to obtain equal variance for statistical analysis. Differences of THg concentrations in different types of seafood, different functional groups, and cultured styles of fish were performed by analysis of variance (ANOVA). The correlation coefficients between fish THg and length and body weight were studied using the Pearson correlation analysis. The results of statistical tests were considered statistically significant if $p < 0.05$.

RESULTS AND DISCUSSION

Hg levels in seafood

In the 518 seafood samples analyzed, the median and mean concentrations of THg were 24.6 ng/g and 37.2 ng/g, respectively (Table 3). The THg concentrations were highly variable among different seafood groups, ranging from 1.02 to 317 ng/g in fish, 0.11 to 55.4 ng/g in shrimp, 9.45 to 107 ng/g in crabs, and 1.84 to 53.3 ng/g in mollusks. The THg concentrations in fish $(p < 0.001)$ and crabs $(p < 0.001)$ were significantly higher than in mollusks, but no significant difference of THg concentrations was observed between shrimp and mollusks ($p > 0.05$). It is suggested that bioaccumulation of THg in seafood products is highly species-specific, probably due to their different ecological characteristics, such as feeding habits and habitats.

Table 3. Statistics summary of total mercury concentrations in different seafood types (ng/g)

Seafood	n	Mean	SD	Median	Min	Max
$Fish***$	327	48.6	45.4	42.1	1.02	317
Shrimp	46	16.4	12.9	13.6	0.11	55.4
$Crab***$	35	31.1	21.0	24.8	9.45	107
Mollusk	110	13.9	9.30	11.4	1.84	53.3
Total	518	37.2	40.0	24.6	0.11	317

 $p < 0.001$, compared with mollusk.

 $SD =$ standard deviation.

The comparison of THg concentrations in different functional groups of fish is presented in Figure 2. In the present study, fish species were categorized by food preferences into three ecological functional groups (carnivorous, omnivorous, and herbivorous fish). The averages of THg concentrations in each functional group of fish were 67.6, 7.82, and 23.7 ng/g for carnivorous, omnivorous, and herbivorous fish, respectively. The THg concentrations in carnivorous ($p < 0.001$) and herbivorous fish ($p < 0.01$) were significantly higher than that in omnivorous fish. The results demonstrate Hg bioaccumulation in species with higher trophic levels [25,26]. But the two species of omnivorous fish (Tilapia and Mugil cephalus) had very low THg concentrations, with the means of 7.81 and 7.97 ng/g, respectively.

According to the cultured styles, the fish species were divided into three groups (FWA, MC, and OF). The averages of THg concentrations were 34.7, 65.1, and 65.9 ng/g for FWA, MC, and OF fish, respectively. Two-way ANOVA (two factors: functional group and cultured style) was used to test the difference of THg concentrations in fish tissues. Interaction was not observed between the two factors. The THg concentrations in MC ($p < 0.01$) and OF fish ($p < 0.01$) were significantly higher than that from FWA fish. Compared with OF fish, significantly higher THg concentrations were also found in MC fish $(p < 0.05;$ Fig. 3). The results indicated that the cultured style had significant effects on THg concentrations in the fish. The degradation of food supplies and fish excretion can produce anoxic conditions at the sediment/water interface, which favor Hg methylation; but these organic matters can potentially be complex Hg and reduce Hg bioavailability to the methylating

Fig. 2. Comparison of total mercury (THg) concentrations in fish with different functional groups. Each box represents interquartile range (25th to 75th percentile). The horizontal band near the middle of the box is the 50th percentile (the median), and the whisker represents the 5th and 95th percentiles. $*** p < 0.001$, and $** p < 0.01$, compared with omnivorous fish.

Fig. 3. Comparison of total mercury (THg) concentrations in fish with different cultured styles. Each box represents interquartile range (25th to 75th percentile). The horizontal band near the middle of the box is the 50th percentile (the median), and the whisker represents 5th and 95th percentiles. $a = p < 0.01$, compared with freshwater aquaculture; $b = p < 0.05$, compared with ocean fishery.

bacteria [26]. In fieldwork conducted in Hong Kong, Liang et al. [21] found that mariculture increased Hg loading from uneaten fish feed and fish excretions, whereas organic matter inhibited Hg methylation in surface sediments.

Regarding the regional difference, the averages of THg concentrations in the fish collected from the western area of the province (Zhanjiang, Maoming, and Yangjiang) were higher than that from Pearl River Delta and the eastern area (Fig. 4). The THg concentrations in the fish collected from Zhanjiang, Maoming, and Yangjiang were significantly higher ($p < 0.05$) than that in fish collected from Dongguan. Most fish from the western areas were carnivorous fish, which may explain the high THg concentrations in the fish tissues.

The relationship between THg concentrations and fish length is well established, with larger fish tending to accumulate substantial Hg concentrations [27,28]. However, this phenomenon was not observed in the present study. There were no significant correlations between THg concentrations in fish and

250

200 150 $Hg(ng/g)$ 100 50 θ $\mathbf{M}\mathbf{M}$ YJ $_{\rm DG}$ $\mathbf{G}\mathbf{Z}$ ZJ FS JM ZH ZS

Fig. 4. Comparison of total mercury (THg) concentrations in the seafood samples from different cities. Each box represents interquartile range (25th to 75th percentile). The horizontal band near the middle of the box is the 50th percentile (the median), the square near the middle of the box is the mean, and the whisker represents the 5th and 95th percentiles. $ZJ =$ Zhanjiang; $MM =$ Maoming; $YJ =$ Yangjiang; $DG =$ Dongguan; $FS =$ Foshan; $GZ =$ Guangzhou; JM = Jiangmen; ZH = Zhuhai; ZS = Zhongshan; SW = Shanwei; $ST =$ Shantou.

Fig. 5. Individual total mercury (THg) concentrations in fish plotted against

the fish length (Fig. 5) and body weight (Fig. 6). Previous studies have confirmed that this relationship can be confounded by other variables, such as changes in growth rate, diet, and activity [29]. Most of the fish in the present study were cultured fish, and the relatively high growth rate might have a dilution effect on THg concentration in the fish muscle [30]. Moreover, the main Hg source for cultured fish is food ingestion, and fish do not have competitive trophic position in the same

The THg concentrations in the seafood samples in the present study were relatively low compared with those reported in United States and Canada (Table 4). No fish samples exceeded the recommended maximum THg limit for human consumption $(0.5 \mu g/g$ wet wt) set by the World Health Organization and the Standardization Administration of China. The results in the present study were comparable to most studies in China and were lower than these from Zhoushan Island [23] and Hg-contaminated Ya-Er Lake [38]. Although China is considered to be the largest source of anthropogenic Hg emission [10], and serious Hg pollution problems were found in local environments [43,44], the THg concentrations in the fish samples showed rather low levels. This may be due to the specific

aquaculture pond [22].

Fig. 6. Individual total mercury (THg) concentrations in fish plotted against body weight.

aquaculture characteristics, young age, high growth rate, and substantially short and simple aquatic food chains [45] of the samples collected for the present study.

Dietary intake and risk assessment

The average daily intake of seafood was 132 g/d for individuals in the present study (Table 2), which was about twice that for Guangdong urban residents (61.5 g/d) and about three times the national average for urban residents (42.3 g/d). The exceptionally large fish consumption may be related to the geographic location of the study area, because coastal residents generally eat a substantial amount of fish.

Because of the high variability of THg levels among individual seafood samples, the median concentrations in each group (fish, shrimp, crabs, and mollusks) were used for health risk assessment. The PDI_{50} of MeHg via seafood consumption ranged from 42.6 to 71.4 ng/kg/d for different age groups (Table 5). The daily intake of seafood was highest among male children and teenagers (6–17 years old). This may be due to the relative low body weight and large seafood consumption. Therefore, based on the results of the present study, the sensitive

Table 5. The median probable daily intake (PDI_{50}) of MeHg via seafood consumption for different age groups (ng/kg/d)

Age and gender	FWA	МC	OF	S&C	Mo	Total
$6-17$ M	13.2	28.3	13.6	12.9	3.4	71.4
$6-17$ F	13.6	20.9	10.1	8.3	2.3	55.1
$18-44$ M	12.8	18.3	8.8	5.3	1.6	46.8
$18 - 44$ F	9.9	15.2	10.2	5.6	1.7	42.6
$45 - 59$ M	12.8	18.3	8.8	4.1	1.4	45.5
$45 - 59$ F	24.9	21.7	7.3	7.7	1.9	63.5
>60 M	14.1	18.5	8.8	4.5	2.0	48.0
>60 F	24.9	21.5	6.1	4.4	1.7	58.7

 $M = male$; F $=$ female; FWA $=$ fish from freshwater aquaculture; MC $=$ fish from mariculture; OF = fish from ocean fishery; $S\&C =$ shrimp and crab; $Mo =$ mollusks.

subgroup should avoid eating carnivorous fish with high THg concentrations.

The PDIs of MeHg from seafood consumption $(0.054 \,\mu\text{g})$ kg/d) in the present study were higher than PDIs in previous reports on MeHg exposure from fish consumption in 12 provinces in China $(0.006 \mu g/kg/d$ [20]), on U.S. adult women $(0.02 \mu g/kg/d$ [46]), and on the Korean population $(0.03 \mu g/m)$ kg/d [47]), but were comparable with those in reports on Hong Kong residents (0.053 µg/kg/d [21]), on Southeast Asia populations $(0.027-0.097 \mu g/kg/d$ [48]), and on a Norwegian group $(0.058 \mu g/kg/d,$ with a range of $0.028-0.18 \mu g/kg/d;$ G. Mangerud, 2005, Master's thesis, Nordic School of Public Health, Goteborg, Sweden]). The PDIs were lower than those reported in Japanese populations with frequent fish consumption $(0.14 \,\mu g/kg/d [49])$.

Regarding the relative contribution of each seafood group to the PDIs of MeHg (Fig. 7), fish contributed the majority (with an average of 84%), followed by shrimp and crab (with an average of 12%). The high contribution from fish mainly resulted from its relatively high consumption rate and high THg concentrations compared with other seafood groups.

For different age groups, the HQ was used to evaluate the exposure risks to MeHg through seafood consumption (Table 6). The HQ in all age groups $(PDI₅₀/PTWI$ and PDI95/PTWI) were less than 1, which indicates that MeHg exposure through seafood consumption may not lead to adverse health effects. However, taking the PTWI of 230 ng/kg/d as the boundary, approximately 2.9% of the population (about 1.58 million) in these 11 coastal cities in Guangdong Province, South

Fig. 7. The relative contribution of each seafood group to median probable daily intakes (PDI₅₀) of MeHg in different age groups. Mo = mollusk; $S&C =$ shrimp and crab; OF = ocean fishery; MC = mariculture; FWA = freshwater aquaculture; $M =$ male; $F =$ female. [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com.]

Table 6. Hazard quotients of MeHg exposure through seafood consumption for different age groups

Age	$6 - 18$		18–44		$45 - 59$		>60	
Gender	M	\mathbf{F}	M	\mathbf{F}	M	- F	М	F
$PDI50/PTWI$ 0.31 0.24 0.20 0.19 0.20 $PDI_{95}/PTWI$ 0.92 0.74 0.63 0.57					0.61	0.28 0.91	0.21 0.65	0.26 0.85

 $PDI₅₀$ median probable daily intake; PTWI = provisional tolerable week intake: $M =$ male; $F =$ female.

China, had a PDI that exceeded the JECFA guideline. Thus, potential health risks are associated with seafood consumption for the general population in the coastal area of Guangdong, South China. The more sensitive populations, such as young children and pregnant women, should avoid eating carnivorous fish with high THg concentrations.

CONCLUSIONS

The THg levels varied significantly among the different seafood types examined in the present study, but the THg concentrations were relatively low and no samples exceeded the national limit. The ecological functional groups and cultured styles have significant effects on Hg accumulation in the fish tissue. Among the different seafood types, consumption of fish provides the main the main pathway of human exposure to MeHg. Thus, potential health risks are associated with seafood consumption for the population in the coastal area of Guangdong, South China.

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