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Mercury pollution in fish from South China Sea: Levels, species-specific accumulation, and possible sources

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ABSTRACT

Both total mercury (THg) and methylmercury (MeHg) levels in fish collected from South China Sea (SCS) were studied to understand Hg pollution in Chinese tropical marine ecosystems. The average THg concentrations in fish species ranged from 39.6 μg/kg for rabbitfish (*Siganus fuscessens*) to 417 μg/kg for thornfish (*Terapon jarbua*), while those of MeHg varied from 13 μg/kg (rabbitfish) to 176 μg/kg (thornfish). The median values of MeHg/THg ratios in different fish species ranged from 36 to 85%. Significant inter-species differences of THg and MeHg in fish were observed due to feeding habits and fish sizes. Overall, carnivorous fish had 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.

1. Introduction

As a global pollutant, mercury (Hg) has received considerable attention due to its toxic, persistent, and bioaccumulative natures and the potential for long-range atmospheric transport (Cheng and Hu, 2010). Mercury biogeochemical cycling in aquatic environment, i.e. methylation of inorganic and reactive Hg, leads to methylmercury (MeHg) bioaccumulation in aquatic organisms and biomagnification via food chain (Fitzgerald et al., 2007). Due to long-range atmospheric transport, MeHg levels in fish in remote areas far from major Hg contamination sources and in regions with low Hg levels in environmental matrices could be elevated (Evans et al., 2005). Therefore, the global biogeochemical cycling of Hg is related to human health because marine fish are important protein supplies for global population (Zhang and Wong, 2007).

China consumes about half of the global Hg supply and is believed to be responsible for a quarter of anthropogenic Hg emission in the world (Cheng and Hu, 2010, 2012a). Multiple environmental matrices, including air, soils, sediments, surface water, and organisms were contaminated by elevated levels of Hg in China (Lin et al., 2012). Besides heavily contaminated industrial and mining areas (Feng and Qiu, 2008; Lin et al., 2012; Zhang and Wong, 2007), megacities consuming coal as the main source of energy were found to have considerably elevated levels of Hg in their environmental matrices (Li et al., 2013). It has been estimated that approximately 77 t of Hg were released annually into the coastal areas via river inputs in China (National Bureau of Oceanography of China (NBO), 2012). Mercury levels ranging from not detectable up to 41.1 mg/kg dry weight were found in the sediments collected from coastal areas of China (Pan and Wang, 2012).

The South China Sea (SCS), the largest semienclosed sea in the western tropical Pacific Ocean surrounding fast-developing Asian countries, plays an important role in the global biogeochemical cycle of Hg. The Pearl River, an important terrestrial contaminant input source of the SCS, has been found to contain up to 318 ng/L of Hg in the brackish water near Guangzhou (Fu et al., 2010). Atmospheric Hg concentration in the SCS ranged from 2 to 7 ng/m^3, which was considerably higher than those in the Arctic Ocean (1.6 ng/m^3) (Fu et al., 2010; Tseng et al., 2010). Previous studies have demonstrated that bioaccumulation of Hg was more significant in fish species with higher trophic levels (Li et al., 2009, 2013). However, Hg bioaccumulation in marine fish in the SCS has not been systematically investigated even though this region is one of the most important fishery zones in China.

Except for a few heavily polluted sites, Hg concentrations in fish on the Chinese markets, which are dominated by freshwater fish, were within the ranges of 0.2–1.0 μg/g for total Hg and

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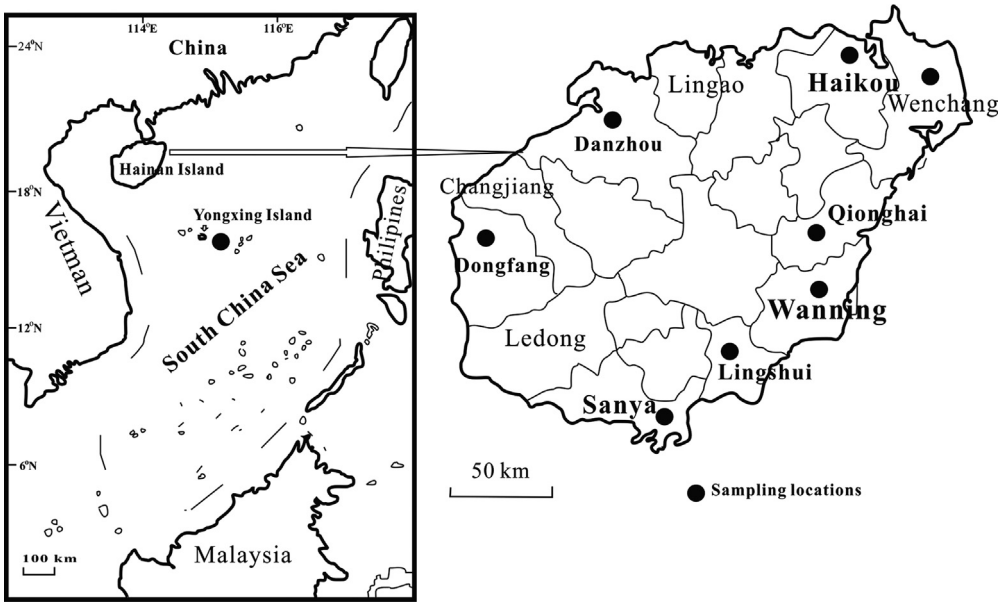

Fig. 1. Map of the study areas.

0.3–1.0 μg/g for MeHg on wet weight basis and were generally below safe limits of (Standardization Administration of China (SAC), 2012; U.S. Environmental Protection Agency (USEPA), 2005 and World Health Organization (WHO), 1990; Cheng and Hu, 2012b; Lin et al., 2012). However, this result seems not to coincide with the elevated Hg levels in Chinese environmental matrices. It is unclear whether the same trend would occur in the Chinese marine ecosystems? This study was carried out to investigate Hg bioaccumulation in various marine fish species from the SCS to answer the question. Total mercury and MeHg contents of marine wild fish species collected from coastal areas of Hainan Island and Yongxing Island in the SCS were measured and compared. Possible sources of Hg in wild fish from the SCS were also discussed.

2. Materials and methods

2.1. Study area and fish collection

Collection of the marine wild fish samples took place at Hainan Island and Yongxing Island (Fig. 1). Hainan Island is the biggest island (33,210 km^2 of land area) in the SCS. It is a main marine aquaculture base with annual production of nearly 990,000 metric tons, accounting for 6.8% of the total marine production in China in 2010 (Food and Agriculture Organization (FAO), 2013). Aquaculture has been expanding fast in Hainan Island to meet the seafood demands from European Union and China over the past decades. Yongxing Island, located approximately 350 km south of Hainan Island, is the biggest island (2.13 km^2 land area) in the middle part of the SCS. Marine fish are the major protein sources for local residents of both islands.

A total of 166 fish samples, consisting of 7 species of wild-caught fish, were collected from local fish markets in 8 towns/cities along the Hainan Island coastline and Yongxing Island between August 2012 and April 2013 (Fig. 1). The fish samples were purchased directly from local fishermen who caught fish in the nearby marine waters. The major characteristics of each fish species, including habitat, feeding habit, size, and water content are summarized in Table 1. These species are oviparous and the period from a larva to adult for these fish is about 6–24 months. All the collected samples were adult fish. These fish were categorized by food preference into three groups, carnivorous, omnivorous, and herbivorous, to study the influence of feeding habit on their mercury bioaccumulation. The collected fish samples were frozen and transferred to the laboratory. After measurement of weight and length of each fish, dorsal muscle was dissected, weighted and freeze-dried. The dried samples were ground and sieved to homogenization and stored in clean polyethylene plastic bags at −20 °C prior to analysis.

2.2. Chemical analysis and QA/QC

The total Hg contents of all fish samples were measured by cold vapor atomic fluorescence spectrometry (CVAFS) after total digestion. Briefly, about 0.1 to 0.2 g of ground fish muscle samples were weighted in 25 mL Teflon vessels and digested with 10 mL of a fresh mixture of nitric acid and sulfuric acid (HNO_3: H_2SO_4=4:1, v/v) in a water bath at 95 °C for 3 h. After cooling, the digested solution was oxidized by BrCl for 24 h, followed by determination of Hg concentration by $SnCl_2$ reduction, purge, gold trap, and cold vapor atomic fluorescence spectrometry (CVAFS) (Liang et al., 1996).

Methylmercury concentration in selected fish samples was measured with the following procedure: aliquot of 0.1–0.2 g of dried fish samples were 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 (Liang et al., 1996).

Method blanks, certified reference material, and blind duplicates were also measured for QA/QC purposes. The detection limits of THg and MeHg in fish muscles were 0.013 and 0.002 μg/kg, respectively. The levels of THg and MeHg in the certified reference material of fish sample (TORT-2, NRCC, Canada) were determined as 0.27 ± 0.02 μg/g and 0.149 ± 0.014 μg/g, respectively, which were in good agreement with the certified values (THg: 0.27 ± 0.06 μg/g; MeHg: 0.152 ± 0.013 μg/g). The relative standard deviations of THg and MeHg in the duplicate samples of fish were lower than 10%. Recoveries for matrix spikes were 95% to 98% and 90% to 110% for THg and MeHg, respectively.

2.3. Statistical analysis

All statistical analyses were performed using SPSS 16.0 for windows (SPSS® Inc., Chicago, IL, USA). Based on Kolmogorov–Smirnov normality test, the THg and MeHg data were log-normally distributed. A one-way analysis of variance (ANOVA) was used to test the difference of Hg concentrations among different fish species. Correlation was considered statistically significant at $p < 0.05$.

In the box plots presented in this study, the line, hinges and whiskers inside the box indicate the median, 25% and 75% quartiles, and 1 and 99 percentiles, respectively. Unless otherwise stated, mercury concentrations in fish samples were expressed in μg/kg wet weight.

3. Results and discussion

3.1. Levels of THg and MeHg in marine wild fish from SCS

Taken together all the fish species, the median, mean, and geometric mean of THg concentrations in marine wild fish were 94.7, 152 and 94.7 μg/kg, respectively (Table 2). There was a large variation of THg concentrations (11.9 to 1772 μg/kg) in marine wild fish from the SCS. The results of ANOVA analysis indicated significant inter-species variation of THg concentrations in fish ($p < 0.05$). As shown in Table 2, thornfish (*Terapon jarbua*), which are carnivorous and feed on crustaceans, small fish and other demersal fish, had the highest THg concentrations (mean 417 μg/kg; median 246 μg/kg)

Table 1
Characteristics of the fish species.

English name	Species	*n*	Feeding habits	Food items	Length (cm)	Weight (g)	Water content (%)
Hainan Island							
Golden threadfin bream	*Nemipterus virgatus*	56	Carnivore	Crustaceans, cephalopod and other small fish	13.4–40	28.0–545	59–78
Thornfish	*Terapon jarbua*	24	Carnivore	Crustaceans, small fish and other demersal fish	12–32.5	27.9–510	68–76
Rabbitfish	*Siganus fuscessens*	32	Herbivore	Algae and seagrass	17.5–28	58.8–271	67–85
Goldsilk seabream	*Acanthopagrus berda*	25	Omnivore	Algae, crustaceans, mollusks and small demersal fish	19–24.5	128–258	66–77
Yongxing Island							
Blue spotted grouper	*Cephalopholis argus*	9	Carnivore	Small fish	20–29	168–410	74–78
Banded reef cod	*Epinephelus fasciatus*	10	Carnivore	Crabs, shrimp, small fish	23–39	185–775	74–78
Yellowband sweetlip	*Plectorhynchus lineatus*	10	Carnivore	Crustaceans,and small fish	29–42	508–889	75–80

Table 2
Summary of descriptive statistics on total mercury (THg) and methylmercury (MeHg) concentrations (μg/kg) in fish muscle.

Fish species	THg					MeHg					MeHg/THg (median)[a]
	Mean	Median	SD	Geometric mean	Range	Mean	Median	SD	Geometric Mean	Range	
Golden threadfin bream	138	109	90.2	116	27.9–455	92.7	88.8	5.07	79.7	23.5–253	30–98 (60)
Thornfish	417	246	471	257	55.3–1772	176	166	91.0	151	36.8–358	16–99 (71)
Rabbitfish	39.6	30.1	29.5	32.6	11.9–148	13.0	12.8	4.31	12.3	6.22–18.8	11–75 (36)
Goldsilk seabream	94.5	59.8	84.7	70.0	23.4–388	72.5	39.9	48.5	58.4	21.3–163	16–99 (41)
Blue spotted grouper	164	149	79.0	148	77.6–303	149	126	73.2	135	73.1–273	80–94 (85)
Banded reef cod	106	89.9	40.6	100	68.9–202	85.7	83.2	26.9	82.2	56.7–135	67–90 (80)
Yellowband sweetlip	122	105	56.0	110	41.5–209	100	102	58.2	81.5	22.1–165	39–85 (75)
Total	152	94.7	221	94.7	11.9–1772	105	91.7	77.6	75.0	6.22–358	11–99 (69)

[a] % In unit.

among all marine wild fish species. In contrast, rabbitfish (*Siganus fuscessens*), which are herbivorous and feed on algae and seagrass, had the lowest concentrations of THg (mean 39.6 μg/kg, median 30.1 μg/kg).

The median, mean, and geometric mean of MeHg concentrations in marine wild fish were 91.7, 105, and 75.0 μg/kg, respectively (Table 2). Significant inter-species variation was found in the fish MeHg levels ($p < 0.05$). The average MeHg concentrations in fish species ranged from 13.0 μg/kg for rabbitfish (*S. fuscessens*) to 176 μg/kg for thornfish (*T. jarbua*). Among the fish samples collected, the highest concentration of MeHg, 358 μg/kg, was measured in a thornfish (*Parupeneus chrysopleuron*). Levels of MeHg in all fish samples from the SCS did not exceed the maximum concentration (0.5 μg/g) recommended by the Standardization Administration of China (SAC) (2012). The species-specific differences of THg and MeHg concentrations were significant ($p < 0.001$), which were probably related to their sizes and feeding habits (Table 1).

It is well accepted that MeHg is the predominant form of mercury in fish muscles (Cheng et al., 2009; Liang et al., 2011; Driscoll et al., 2013). In previous studies, the ratio of MeHg to THg was generally found to be about 80% (Cheng et al., 2009; Liang et al., 2011). However, in the present study, the ratios of MeHg/THg in the fish samples ranged from 11 to 99%, with a median value of 69% (Table 2). Significant differences of MeHg/THg ratios were also found among different fish species ($p < 0.05$), which might be caused by the differences in Hg and MeHg absorption and metabolism among fish species. Moreover, median MeHg/THg ratios in carnivorous fish (60–85%) were higher than those of omnivorous fish (41%) and herbivorous fish (36%) (Table 2). Positive correlation between the MeHg/THg ratio and trophic level of fish has been found in a previous study (Van der Velden et al., 2013). Thus, different trophic levels of fish species may be another factor responsible for the differences in MeHg/THg ratios among fish species.

3.2. Comparison of THg and MeHg concentrations among fish species of different trophic levels

The marine wild fish species were categorized by food preferences into three groups: carnivorous, omnivorous, and herbivorous (Table 1). Among these three groups (Fig. 2), the carnivorous, representing the top trophic level, had the highest average THg (197 μg/kg) and MeHg concentrations (124 μg/kg). The average concentrations of THg and MeHg in the omnivorous fish were 94.6 and 72.5 μg/kg, respectively. As expected, the lowest average Hg concentrations were found in the herbivorous fish (39.6 μg/kg for THg and 13.0 μg/kg for MeHg). The increased Hg bioaccumulation in fish species of higher trophic levels is consistent with the findings of previous studies (Li et al., 2009, 2013). The difference in Hg concentrations among different trophic groups might be due to fish size.

Pearson correlation analysis showed significant positive correlations between THg and MeHg concentrations in fish and fish size, except of blue spotted grouper (*Cephalopholis argus*), banded reef cod (*Epinephelus fasciatus*), and yellowband sweetlip fish (*Plectorhynchus lineatus*) (Table 3). Such relationship has also been found in various fish species previously (Dang and Wang, 2012; Peterson and Sickle, 2007). It revealed that the Hg bioaccumulation was related to fish size (Trudel and Rasmussen, 2006), as well as different biokinetic characteristics (Dang and Wang, 2012). Mercury, primarily in the form of fat-soluble MeHg, is efficiently absorbed but only slowly excreted by fish, resulting in bioaccumulation of Hg in fish as they grow (Cheng and Hu, 2012b).

3.3. Comparison of Hg concentrations in fish from SCS and other regions

Compared with those reported in other studies, the mean THg concentration (152 μg/kg) in marine wild fish samples of the SCS was in the middle level (Table 4). Overall, the THg concentrations

in fish from the SCS were lower than those in fish from the East Sea (Cheng et al., 2009), gulf of California (Ruelas-Inzunza et al., 2012), mercury-polluted lake in Central China (Jin et al., 2006), and the US fish with highest levels of Hg (U.S. Food and Drug Administration (USFDA), 2010), but higher than those in marine wild fish from the US (U.S. Food and Drug Administration (USFDA), 2010), Korea coastal area (Kim et al., 2012), Bohai Sea (Liu et al., 2013), and the Northern SCS (Li et al., 2013). The high levels of THg in marine wild fish from the SCS were probably related to the Hg discharge by human activities in coastal areas and atmospheric Hg deposition (Li et al., 2009; Pacyna et al., 2010).

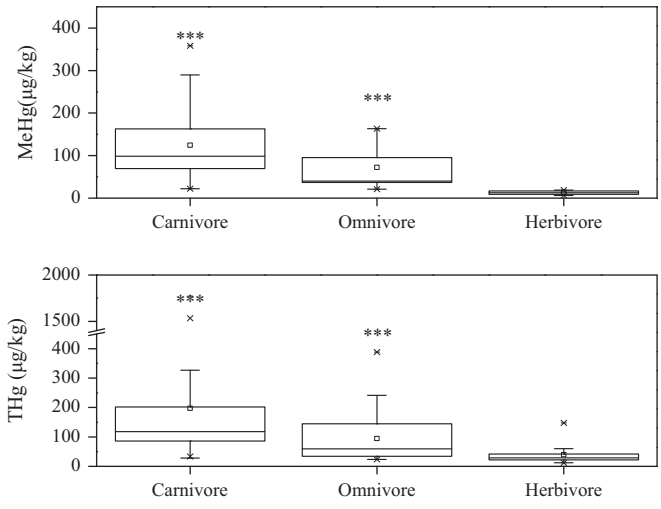

Fig. 2. Comparison of total mercury (THg) and methylmercury (MeHg) concentrations in fish by different functional groups. ***$p < 0.01$, compared with herbivorous fish.

Table 3
Pearson correlation coefficients between mercury concentrations in fish and fish size.

Fish species	Hg	*n*	Length	Weight
Golden threadfin bream	THg	56	0.409**	0.390**
	MeHg	26	0.671**	0.721**
Thornfish	THg	24	0.771**	0.732**
	MeHg	22	0.429*	0.319
Rabbitfish	THg	32	0.475**	0.437*
	MeHg	11	−0.043	−0.088
Goldsilk seabream	THg	25	0.474*	0.458*
	MeHg	13	0.3	0.177
Blue spotted grouper	THg	9	−0.287	0.002
	MeHg	8	−0.434	−0.124
Banded reef cod	THg	10	0.366	0.541
	MeHg	9	0.298	0.107
Yellowband sweetlip	THg	10	0.596	0.872**
	MeHg	7	0.619	0.73

* $p < 0.05$.
** $p < 0.01$.

3.4. Possible sources of Hg in marine wild fish from SCS

The possible sources of Hg in marine wild fish from Hainan Island of SCS could be inferred from the mercury emission inventory and transport routes of mercury in the SCS. On one hand, there are two big municipal solid waste (MSW) incineration plants in Hainan province. Although there was no data on the Hg emission from the MSW plants of Hainan province, the MSW plant in China was estimated to contribute 10 t Hg in 2003 and was considered as an important Hg emission source (Cheng and Hu, 2010; Chen et al., 2013). Mercury released from the MSW incineration plants could be deposited into marine environment and eventually uptaken by aquatic organisms, including fish. On the other hand, the eastern coastal areas of Hainan face the PRD on the southwest, which is a heavily industrialized region in China with serious Hg pollution (Liu et al., 2011; Shi et al., 2010). It has been reported that a total of 17,244 kg of Hg were emitted from anthropogenic sources in the PRD in 2008 (Zheng et al., 2011). Thus, the Hg released from the PRD might be another important source of Hg input to the eastern coastal areas of Hainan Island.

The high levels of THg (106–164 μg/kg) and MeHg (85.7–149 μg/kg) in marine wild fish collected from Yongxing Island indicated Hg pollution was probably widespread in marine environment of the SCS. Yongxing Island is located in the middle of the SCS and there are no industrial activities on the island. It is well recognized that atmospheric deposition is the main source of Hg in open seas (Fu et al., 2010; Mason and Sheu, 2002; Sunderland and Mason, 2007). Riverine input and lateral and vertical seawater flow also contribute to Hg contamination in marine environment (Sunderland and Mason, 2007). A previous study indicated that South China and the Indochina peninsula were the main atmospheric Hg source regions for the SCS (Fu et al., 2010). The concentration of dissolved Hg in the seawater surrounding Yongxing Island (0.018 μg/L) was found to be much higher than that of the Pacific Ocean (0.0003 μg/L), but comparable to those of offshore areas in Sanya (0.02 μg/L) and Qionghai (0.03 μg/L) (Zhou et al., 2007). These findings, together with the results in the present study, indicate that the marine environment of Yongxing Island was influenced by mercury emitted from anthropogenic sources in the coastal areas of the mainland surrounding the SCS. In addition, coastal reclamation for land expansion with the development of Yongxing Island, as well as domestic sewage and landfill leachate due to the lack of wastewater or waste treatment facilities on the island, could also contribute to anthropogenic Hg

Table 4
Comparisons of total Hg (THg) concentrations in fish from different areas worldwide (μg/kg).

Study area	Fish	*n*	Mean	Range	References
SCS (Hainan Island, Yongxing Island)	Wild	166	152	11.9 – 1772	This study
Northern SCS (Guangdong Province coastal cities)	Wild and farmed	327	48.6	1.02 – 317	Li et al. (2013)
Bohai Sea (Laizhou Bay)	Wild	67	63.8	2.05 – 344	Liu et al. (2013)
East Sea (Zhoushan Island)	Wild	148	260	20 – 660	Cheng et al. (2009)
Gulf of California	Wild	417	248	26.5 – 639	Ruelas-Inzunza et al. (2012)
Korea (Yellow Sea, East Sea)	Wild	686		6.31 – 96.6	Kim et al. (2012)
US fish with highest levels of Hg	Mackerel king (wild)		730	230 – 1670	U.S. Food and Drug Administration (USFDA) (2010)
	Shark (wild)		979	ND-4540	
	Swordfish (wild)		995	ND-3220	
	Tilefish (wild)		1450	650 – 3730	
US fish with lower levels of Hg	Anchovies (wild)		17	ND-49	U.S. Food and Drug Administration (USFDA) (2010)
	Butterfish (wild)		58	ND-36	
	Catfish (wild)		25	ND-314	

contamination, although their contribution is expected to be rather small due to the island's small size and small population.

4. Conclusions

Although the MeHg levels in all fish samples from the SCS did not exceed the national advisory limit of China, the average concentration of THg in wild fish from the SCS was in the middle level compared with other regions globally. Significant inter-species differences of THg and MeHg in fish were observed, which could be attributed to different feeding habits and sizes of the marine wild fish. Carnivorous fish had higher levels of THg, MeHg, and MeHg/THg ratio than omnivorous and herbivorous fish. Anthropogenic activities as well as long-range atmospheric transport from mainland of China were believed to be important sources of Hg pollution in the SCS.

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