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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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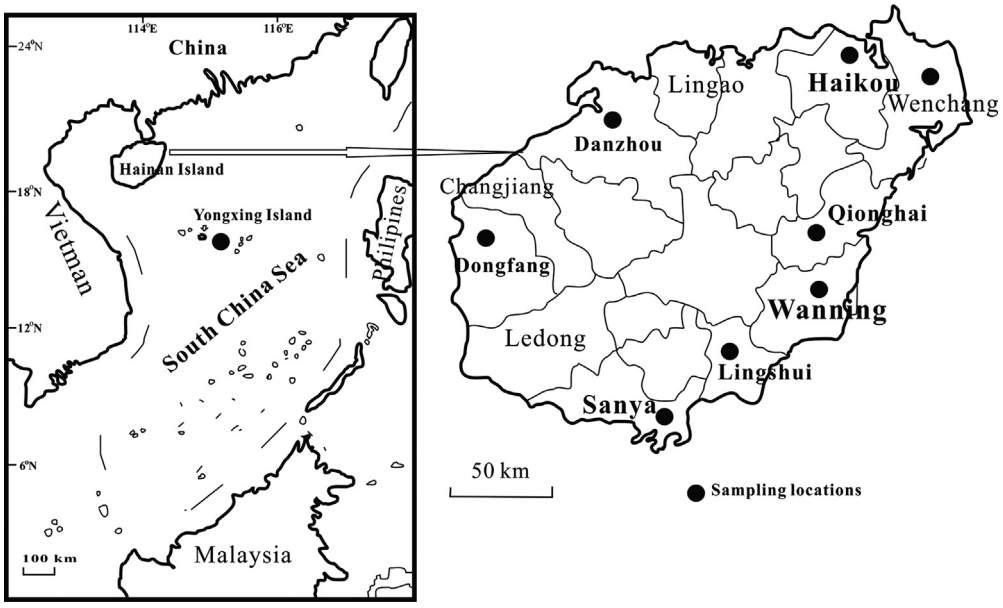

Fig. 1. Map of the study areas.

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)