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Inter-species differences of total mercury and methylmercury in farmed fish in Southern China: Does feed matter?

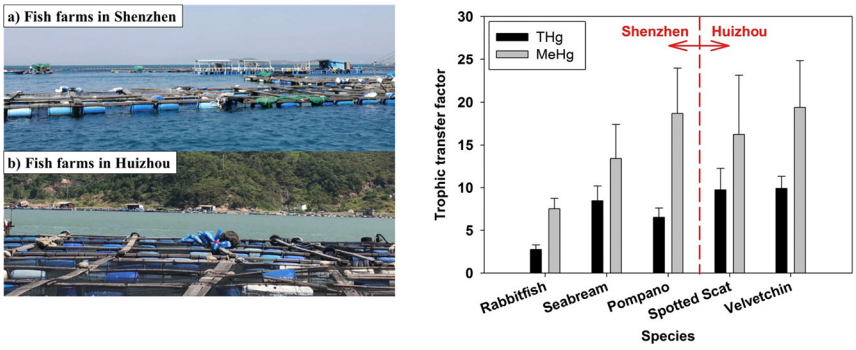
Wanze Li, Wen-Xiong Wang *

Marine Environmental Laboratory, HKUST Shenzhen Research Institute, Shenzhen 518057, China
Department of Ocean Science, Hong Kong University of Science and Technology, Clearwater Bay, Kowloon, Hong Kong

HIGHLIGHTS

- Mercury levels in artificial pellets were the main determinants of Hg accumulation in fish.
- Muscle was the primary organ for Hg and contained the highest ratio of MeHg/THg, and intestine was a critical organ for Hg biotransformation.
- Fish solely fed on the artificial pellets bioaccumulated Hg differently.
- $\delta^{15}N$ could not be used to determine the trophic levels in culturing systems where artificial practices were involved.
- Hg bioaccumulation in fish depended on internal Hg biotransformation and biokinetics.

GRAPHICAL ABSTRACT



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ABSTRACT

China is now the largest producer of marine farmed fish and there is a considerable concern of seafood safety due to potential mercury contamination. We analyzed both the total mercury (THg) and methylmercury (MeHg) concentrations in nine species of commercial fish from two marine-cage farms in Southern China. ^{13}C and ^{15}N stable isotopes were concurrently analyzed to identify the artificial feed sources and the trophic levels of farmed fish. Mercury concentrations of all species were much lower than the human health screening values and safety limits established by different countries. Mercury levels in artificial pellets were the main determinants of Hg accumulation in fish between two sites, while somatic growth dilution and size also played an important role. Among the different fish tissues, muscle was a major reservoir for Hg and contained the highest ratio of MeHg/THg, and liver was the second important organ for Hg accumulation in most fish species. Intestine was a critical organ for Hg biotransformation with its %MeHg differing greatly among different fish species. $\delta^{15}N$ analysis could not be used to determine the trophic levels in culturing systems where artificial practices were involved. Based on the $\delta^{13}C$ signatures, five species of fish were identified to solely feed on the artificial pellets, yet the Hg bioaccumulation differed significantly among these species. We therefore concluded that Hg bioaccumulation in different fish species may be dependent on their internal Hg biotransformation as well as their biokinetics.

1. Introduction

Mercury (Hg) is a very toxic metal produced both naturally and anthropogenically as a result of industrial activity and coal burning, and considered as a global pollutant due to its atmospheric presence and

* Corresponding author.
E-mail address: wwang@ust.hk (W.-X. Wang).

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transport (Ratcliffe et al., 1996). Hg is among the few metals that can be biomagnified through all levels of food chains in aquatic systems (Lindqvist et al., 1991; Wang, 2002, 2012), and animals at higher trophic levels tend to contain higher concentrations of Hg. Hg presents high risks to human health, especially to neural system and fetus, and is positively correlated with the cardiovascular disease (WHO, 2007). Since 1970s, Hg emission has increased sharply, and China is now the largest Hg emission country in the world (Feng, 2005; Wang and Luo, 2017). Contamination of Hg in sediments was found in many areas in China (Qiu et al., 2005; Yu et al., 2012), while high Hg concentrations were also documented in both estuary and coastal waters (Wang et al., 2009; Zhang and Wong, 2007).

Diet (i.e., trophic transfer) is considered as the main uptake pathway of Hg for most populations in the world (Wang, 2012). The consumption of Hg contaminated fish is the major source of MeHg exposure, especially in populations relying heavily on the consumption of predatory fish, and cooking cannot effectively eliminate mercury from fish (WHO, 2007; Bernhoft, 2011). Hg has both inorganic and organic forms, and most of the Hg found in fish are in the form of MeHg (Harris et al., 2003), which is the most toxic Hg species found in fish. Due to its high assimilation efficiency and low excretion rate, MeHg can be easily absorbed and stored in human bodies, which may cause serious toxic effects (WHO, 2007).

As the largest producer, consumer, and processor of fish in the world, China contributes about 50% of the global aquaculture value (FAO, 2016). Previous reports documented that the total production of both farmed and wild fish in China had tripled over the past two decades, and aquaculture is mostly responsible for all such increase (Cao et al., 2015). Bioaccumulation of Hg is traditionally regarded mainly through trophic transfer, and predator fish generally accumulate higher levels of Hg than the prey species (Jernelöv and Lann, 1971; Kidd et al., 1995; Renzoni et al., 1998; Newman, 2012). There is thus a considerable concern of Hg in marine farmed fish. In China, farmed fish are commonly fed with artificial diets without actually considering their trophic levels. One natural question is thus whether biomagnification, a very unique characteristic for Hg, is still applicable in the farming system which is drastically different from the natural systems.

In the present study, both total mercury (THg) and methylmercury (MeHg) concentrations in selected species of commercial marine fish collected from two culturing farms in Southern China were quantified. The Southern China (Guangdong and Fujian Provinces) now hosts one of the largest scale of fish farming, and therefore the concerns for Hg contamination in these farmed fish are enormous. We also measured the levels of Hg in the artificial feeds collected from both sites, since they served as the main diets for many species of marine-caged fish. Although the use of plants (as protein and lipid sources) has increased in feed (Hardy, 2010), most of the artificial feeds are manufactured from other smaller fishes or the viscera which provide abundant nutrients as proteins, fatty acids, vitamins, minerals and several growth factors for fish (Lovell, 1989; Abowei and Ekubo, 2011).

Earlier studies demonstrated that Hg levels in fish tissues were correlated with the contamination in feeds, and Hg in the fish feeds could be used to track the source of Hg pollution (Choi and Cech, 1998). Due to the very unique feeding practices in the fish farms, we specifically examined whether fish at different trophic levels but fed with the same feeds displayed different Hg concentrations. We further conducted stable isotope analysis to identify the possible food sources and trophic levels of the farmed fish. Coupling analysis of stable isotope signatures and Hg levels allowed the evaluation of Hg bioaccumulation through dietary exposure pathway.

2. Materials and methods

2.1. Fish sampling

Nine species of marine-caged fish were sampled from two major fish farming sites in the eastern coastal areas of Guangdong Province of Southern China (Table 1) in November 2016. The two farming sites had different species of fish farming as well as different natural environments. The two sites had different levels of Hg levels in the fish feeds. Specifically, the Shenzhen site was situated along the coastline of Mirs Bay, whereas the Huizhou site was a semi-enclosed bay (Fig. 1). Furthermore, the fish diets were different between the two sites (Table 1). Six species were collected from Nan'ao, Shenzhen (22° 31.578′ N, 114° 29.023′ E), including rabbitfish (*Siganus fuscescens*), gilthead seabream (*Sparus aurata*), golden pompano (*Trachinotus blochii*), red drum (*Sciaenops ocellatus*), brown marbled grouper (*Epinephelus fuscoguttatus*), and mangrove red snapper (*Lutjanus argentimaculatus*). The other three species were sampled in Huidong, Huizhou (22° 43.178′ N, 114° 56.144′ E), including spotted scat (*Seatophagus argus*), short barbeled velvetchin (*Hapalogenys nigripinnis*), and javelin grunter (*Pomadasys kaakan*). At each site, individuals of each species were randomly selected from the stock, while no gender difference was considered. All species were the widely farmed commercial fishes and their culturing methods have been well established. Fish of similar size per species were sampled to minimize the influence of fish size on mercury determination. Meanwhile, four different fish feeds used in each farm were also collected (Table 2). Feed-1 (Nanyu Co, China, 8 mm diameter, dry) and Feed-2 (Yuanpai Co, China, 2 mm diameter, dry) were sampled in Nan'ao, Shenzhen, while Feed-3 (Shangyi Co, China, 4 mm diameter, dry) and Feed-4 (Nanyu Co, China, 1 mm diameter, dry) were sampled in Huidong, Huizhou. All the feeds were the complex artificial pellets composing of nutrients such as protein, carbohydrate, lipid and vitamin, and were fed to various fish according to their sizes. Any mercury presented in these fish feeds was considered to be originated from their background diet compositions. However, the detailed compositions of these artificial diets were not shown by the manufacturers.

2.2. Sample preparation

The fish samples were placed in ziplock plastic bags and transported back to the laboratory of the Hong Kong University of Science and Technology, Hong Kong, within 2–3 h. Fish species were sorted out and their total length and body weight were measured (Table 1). All the fish were dissected with stainless steel knife and scissors washed by deionized water, and their liver, stomach, intestine and muscle were separated by dissection (except for rabbitfish which did not have any distinguishable stomach as herbivores, and only liver, intestine and muscle tissues were therefore collected). Samples were grinded by a homogenizer and freeze dried at −80 °C (Freeze-dryer, Edwards Super Modulyo) for 72 h before they were stored at −20 °C. All chemical analyses were completed within 3 months after sampling to avoid any potential Hg loss. The concentrations of THg and MeHg were all based on tissue dry weight (dw).

2.3. Chemical analyze

The THg and MeHg concentrations were determined in the Hong Kong University of Science and Technology, while the stable isotopes were analyzed by the Stable Isotope Facility (SIF) of University of California at Davis of USA.

THg concentrations in fish and feeds were determined using the method of EPA 7474 (USEPA, 2007) with some modifications. Fifty miligrams of the freeze dried samples or feed pellets powder were digested with 2 mL aqua regia at 80 °C overnight in a heating block after pre-digestion for 2 h at room temperature (25 °C). After digestion, the sample volume was adjusted to 5 mL with Milli-Q water. Two mL of diluted sample was then removed for analysis by adding 2 mL of bromide/bromate solution (11.89 g potassium bromide and 2.78 g potassium bromate in 1 L Milli-Q water) and 2.5 mL of concentrated hydrochloride. The mixture was brought up to 50 mL for cold digestion for 15 min, and then 100 μL of hydroxylammonium chloride solution