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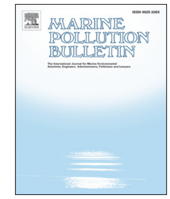

Concentration and potential health risk of heavy metals in seafoods collected from Sanmen Bay and its adjacent areas, China

Qiang Liu, Yibo Liao, Lu Shou*

Key Laboratory of Marine Ecosystem and Biogeochemistry, Second Institute of Oceanography, State Oceanic Administration, Hangzhou 310012, China

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ABSTRACT

Commercial marine fish, crustacean, and mollusc samples were collected from Sanmen Bay and its adjacent areas to investigate the accumulation of heavy metals in various species of marine organisms and evaluate the potential health risk for local consumers. The results indicated significant variations in metal contents among species. The highest concentrations of studied metals were found in molluscs, followed by crustaceans and fish. The first metal group of arsenic (As), cadmium, copper, and zinc were associated by their relatively high concentrations in the tissues of marine organisms, whereas chromium, mercury, and lead were identified the other group with low concentrations. Human health risk evaluation indicated that the exposure doses of most elements for coastal people were safe, except for As, which scored a high total target hazard quotient and target cancer risk value. Potential health risk of heavy metal exposure from seafood consumption should not be ignored.

1. Introduction

Seafoods contain all of the essential amino acids and thus are complete protein sources for human beings (Fuentes et al., 2009). People living in coastal areas rely on seafoods, notably fish, crustaceans, and molluscs, as their primary source of animal protein. In addition, seafoods are generally considered to be a reasonable source of vitamins and essential minerals with low contents of calories and fat (Cozzolino, 2001; Ozden et al., 2010). However, nutritional value and consumer health risk from seafoods must be taken into consideration, because marine organisms can accumulate chemical contaminants from the environment.

Heavy metals, such as cadmium (Cd), lead (Pb), and mercury (Hg), as well as metalloids like arsenic (As) from natural and anthropogenic sources continuously enter marine environment. In China, rapid urbanization and industrialization have resulted in increasing toxic metal pollution in coastal and estuarine environments as metals released with municipal and industrial wastes from land-based sources may enter the sea through rivers and land runoff (Pan and Wang, 2012). The total amount of metals fluxes, including copper (Cu), zinc (Zn), Cd, Pb, Hg, and As, only from the Yangtze River in the year of 2015 was estimated to be > 14,000 tons, which was much higher than the amounts from other rivers in China (SOA, 2016). Thus, the coastal and estuarine environments of East China Sea are under stress from heavy metal pollution.

Heavy metals are toxic and nonbiodegradable environmental contaminants that can accumulate in the tissues of organisms and may cause severe damage to the liver, kidney, central nervous system, mucus tissues, intestinal tract, and reproductive systems at high levels (Raknuzzaman et al., 2016). Moreover, metals may be transferred to human body through the food chain, posing a potential health risk to human beings (Parker and Hamr, 2001; Burger, 2008; Brahmia et al., 2013). The health risk of heavy metal contamination is regarded as a global crisis, and several investigations have focused on human exposure to heavy metals resulting from the consumption of contaminated seafood (Chien et al., 2002; Raknuzzaman et al., 2016). Therefore, heavy metal concentrations in commercial marine organisms in heavily polluted areas should be monitored for seafood safety, and their potential health risk for local consumers should be evaluated.

The Sanmen Bay, located in the middle coast of Zhejiang Province, is a semi-enclosed bay that opens to the East China Sea. Sanmen Bay is one of the most critical aquaculture bays in China, with a drainage basin of approximately 3160 km^2 (Liu et al., 2012). Heavy metal contamination and potential ecological risk in the marine environment of Sanmen Bay have been highlighted from 1990s (Li et al., 2011). However, few studies on heavy metal contamination in marine organisms have been conducted along coastline of Zhejiang Province especially the aquaculture bays, and the available information mainly focuses on bivalve shellfish (Huang et al., 2007). Data with regard to different marine species are not sufficient. Thus, the heavy metal contents in various commercial marine organisms collected in aquaculture bays along Zhejiang Province are essential to be investigated, and

* Corresponding author at: Second Institute of Oceanography, State Oceanic Administration, No. 36, Baochu North Road, Hangzhou 310012, China.
E-mail address: shoulu981@sina.com (L. Shou).

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potential health risk of heavy metal for coastal people through seafood consumption should be evaluated.

In this study, we chose a typical marine aquaculture bay, Sanmen Bay, and its adjacent areas as study areas. Commercial species of fish, crustaceans, and molluscs were collected, and the amount of heavy metals in their tissues was analyzed. The implications of heavy metal accumulation for seafood consumption safety were also assessed. The aims of the present study were (Bartolomé et al., 2010) to investigate levels of heavy metals in various species of marine organisms collected from the Sanmen Bay and its adjacent areas, (Bates et al., 1992) to explore associations among metals accumulated in the tissues of marine organisms, (Bernhoft, 2012) to calculate the daily intake and target hazard quotient (THQ) of heavy metals resulting from the consumption of seafood, and (Bille et al., 2015) to evaluate potential health risk for local consumers.

2. Materials and methods

2.1. Sample collection and preparation

Bottom trawling was used to collect 47 marine organism samples, including 14 crustacean and 33 fish samples, from 29 sites in Sanmen Bay and its adjacent areas in October and November of 2015 (Fig. 1). Three crustacean species, namely coastal mud shrimp (*Solenocera crassicornis*), shiba shrimp (*Metapenaeus joyneri*), and swimming crab (*Portunus trituberculatus*), were collected with 3, 4, and 7 replicates for each species, respectively. Five fish species, namely bombay duck (*Harpadon nehereus*), silvery pomfret (*Pampus argenteus*), Japanese mackerel (*Pneumatophorus japonicus*), spottedtail goby (*Acanthogobius ommaturus*), and croaker (*Collichthys lucidus*), were collected in 25 sites with 14, 3, 3, 4, and 9 replicates for each species, respectively. Six mollusc species, namely blood clam (*Tegillarca granosa*), razor clam (*Sinonovacula constricta*), oblique ark shell (*Barbatia obliquata*), toothed top shell (*Monodonta labio*), murex snail (*Thais bronni*), and blue mussel (*Mytilus edulis*), were purchased from local fishermen who caught samples from the coastal sites of Sanmen Bay in November 2015, and 6, 3, 6, 3, 3, and 6 replicates were collected for each species, respectively.

After collection, living organism samples were immediately washed with seawater, placed in clean polyethylene bags, and transported to laboratory in an icebox. After transportation, marine organism samples were frozen and rinsed in deionized water to remove surface adherents. The edible portions of samples were dissected using a steam-cleaned stainless steel knife. The soft tissues of each organism species collected from the same sampling site were homogenized in a blender and then immediately stored at −20 °C until further analysis.

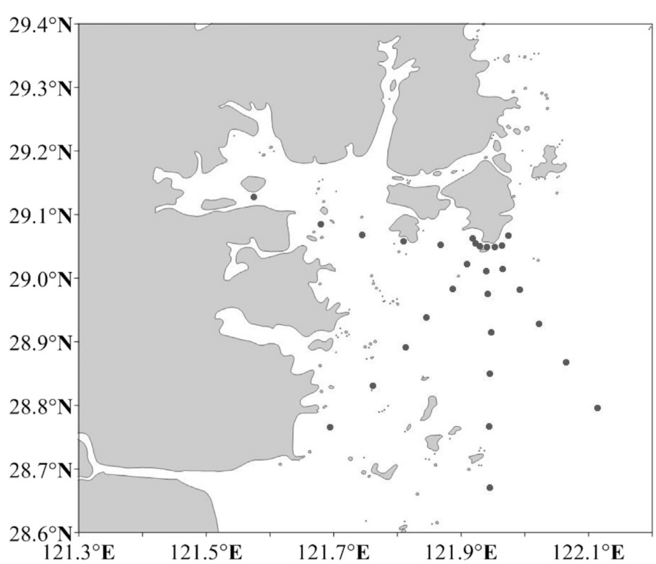

Fig. 1. The sampling sites (black dots on the map) of marine organisms in Sanmen Bay and its adjacent areas.

2.2. Heavy metal analysis

Approximately 1.0 ± 0.010 g of the soft tissue samples of marine organisms were weighed directly into acid-washed Teflon digestion vessels and digested with 10 mL of ultrapure nitric acid by using a microwave system (CEM Mars 6, USA) under the conditions described by Liu et al. (2015). After digestion, samples were diluted to 50 mL with ultrapure water (18.2 MΩ cm^{-2} Milli-Q water, Millipore) for analysis.

The concentrations of As, Cd, Cr, Cu, Pb, and Zn were analyzed using an inductively coupled plasma mass spectrometer (ICP-MS, PerkinElmer, Elan DRC-e, USA) according to an internal standard of the rhodium (Rh) and rhenium (Re) method. Hg was measured through atomic fluorescence spectrometry (AFS, Skyray Instrument, AFS200, China). Heavy metal concentrations are expressed as μg/g wet weight (ww) in this study.

The detection limits of the methodology (MDL) calculated using the triple standard deviation values of blanks and the average wet weight of samples were as follows: As (0.002 μg/g), Cd (< 0.001 μg/g), Cr (0.007 μg/g), Cu (0.010 μg/g), Pb (0.002 μg/g), Zn (0.003 μg/g), and Hg (0.001 μg/g). Standard reference materials of CRM shrimp GBW10050 (GSB-28) was obtained from the Center of National Standard Reference Material of China. The reference materials were prepared and analyzed for heavy metals by using the same procedures that were applied to the samples. The recoveries for standard reference materials were within 10% of the certified values. All analyses of blank, standard reference materials, and samples were performed in triplicate. Relative standard deviation in replicates and reference material analyses were below 10%. For undetected levels, we assigned the one-half value of the limit of detection.

2.3. Statistical analysis

Data normality was assessed using the Kolmogorov–Smirnov (K–S) test, and all the data showed normal distribution in this study. Species variation of mollusc, crustacean, and fish were assessed using one-way analysis of variance. Statistical significance was determined at $p < 0.05$. Pearson correlation analysis, principal component analysis (PCA), and hierarchical cluster analysis (HCA) were used to explore associations among heavy metals in the tissues of marine organisms. All statistical analyses were performed using the statistical package SPSS 20.0 (IBM SPSS Statistics, IBM Corp., USA).

2.4. Health risk assessment

Estimated daily intakes (EDIs) of heavy metals for coastal people through seafood consumption were calculated using the following formula and is expressed as μg per kg of body weight per day (μg/kg bw/day):

$$EDIs = \frac{EF \times ED \times SIR \times C}{WAB \times TA}$$

where EF is the exposure frequency (365 days/year); ED is the exposure duration (70 years, equivalent to the average lifespan); SIR is the seafood ingestion rate of molluscs, crustaceans, and fish (g/person/day); C is the heavy metal concentration in seafood (μg/g ww); WAB is the average body weight (55.9 kg for adults) as used in previous studies (Hang et al., 2009); and TA is average time (365 days/year multiplied by the number of exposure years, for which 70 years was assumed in this study). The average daily consumption of molluscs, crustaceans, and fish of coastal people was assumed to be 17.1, 8.3, and 105 g/person/day, respectively, as used in previous studies (Jiang et al., 2005; Guo et al., 2007).

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