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Bioaccessibility and human health implications of heavy metals in different trophic level marine organisms: A case study of the South China Sea

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ABSTRACT

This study investigated the total concentrations and bioaccessibility of heavy metals in edible tissues and trophic levels of 12 marine organism species in the South China Sea. The results were used to estimate health risks to humans. Of the heavy metals detected, nickel (Ni) was present at the highest concentrations, followed in descending, order by iron (Fe), zinc (Zn), manganese (Mn), chromium (Cr), copper (Cu), cadmium (Cd) and lead (Pb). Cd had the highest percentage bioaccessibility (61.91%). There were no correlations between log-transformed total metal concentrations and trophic level values, nor between log-transformed bioaccessibility metal concentrations and trophic level values. This indicates there is no biomagnification among these trace metals. The carcinogenic risk probabilities for Pb and Cr to urban and rural residents were below the acceptable level ($< 1 \times 10^{-4}$). The target hazard quotient (THQ) value for each metal and the total THQ values for all metals studied indicated no significant risk of non-carcinogenic effects to urban and rural residents from consuming marine organisms from the South China Sea.

1. Introduction

Heavy metals are of increasing global concern due to their persistence in the environment, effects on biogeochemical recycling, and ecological risks (Gu et al., 2016a; Souza Machado et al., 2016; Jonsson et al., 2017). Heavy metals are classified as potentially toxic (*e.g.,* Cd, Pb, Ni) or nutritionally essential (*e.g.,* Cu, Zn, Fe, and Mn). Even at low levels, toxic metals can threaten human health when ingested over extended periods, and essential metals may have toxic effects at supraoptimal concentrations (USEPA, 2007a; Subotić et al., 2013; Gu et al., 2017).

Over the past decades, human consumption of aquatic animals has increased rapidly around the world, because they provide beneficial high quality proteins, have low levels of saturated fat, and also contain omega-3 fatty acids that, support wholesomeness (Bosch et al., 2016; Golden et al., 2016; Jennings et al., 2016; Gu et al., 2017). However, heavy metal pollution in aquatic animals is of great concern, because the pollutants threaten aquatic animal health and create hazards for humans who consume the animals (Jitar et al., 2015; Varol and Sünbül, 2017). Aquatic animals accumulate metals from the surrounding water, sediment, and their diet (Zhao et al., 2012; Jayaprakash et al., 2015; Jitar et al., 2015). As a result, aquatic animals are considered indicator of heavy metal pollution in aquatic environments (Holt and Miller, 2010; Amoozadeh et al., 2014). Metal accumulation in fish can produce long-term impacts on ecosphere biogeochemical cycling (Amoozadeh et al., 2014; Chakraborty et al., 2014; Gu et al., 2017). Metals accumulate as they move up the food chain, and can lead to health problems when consumed in amounts exceeding safe levels.

Total concentrations of heavy metals in aquatic animals can provide valuable information on overall pollution levels in food matrices. However, the ingested dose of heavy metals does not always reflect the actual level of heavy metals available to the consumer (Zhuang et al., 2016). Understanding the bioavailability of heavy metals in aquatic animals helps evaluate the amount of heavy metals that can be absorbed by the human body. This involves learning about the oral bioaccessibility of heavy metals in aquatic animals. The physiologically based extraction test (PBET) and the simple bioaccessibility extraction test (SBET) have been successfully established and verified to estimate the bioaccessibility of soil and food metals (Gu et al., 2016a; Zhuang et al., 2016). SBET is a simplified form of PBET and was developed as a

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fast, easy, and reproducible extraction test. A standard operational protocol of SBET has been established to investigate *in vitro* lead bioavailability (Wragg and Cave, 2003; USEPA, 2007b; Hu et al., 2011a, 2011b). The validity of these tests has recently been demonstrated in testing of soils and rice (Guney et al., 2010; Mingot et al., 2011; Gu et al., 2016a; Zhuang et al., 2016). However, there is a lack of data on the bioaccessibility of heavy metals in different trophic level marine organisms based on *in vitro* digestion. This highlights the importance of performing more bioaccessibility studies of heavy metals in different trophic level marine organisms to improve risk assessments.

The main objectives of this study were: (1) measure the total concentrations and the bioaccessibility of heavy metals in marine organisms, (2) determine relationships among total concentrations, heavy metal bioaccessibility and trophic levels of marine organisms, and (3) evaluate human health risks based on the bioaccessible heavy metals from marine organisms taken in through the ingestion pathway.

2. Materials and methods

2.1. Marine organism collection

From March to April 2015, 282 marine biota samples were collected at 19 sampling stations in South China Sea (Fig. 1). The samples included a total of 12 species, including seven fish species and five cephalopod species. Table S1 presents the ranges in physical characteristics of samples, based on our previous study (Ke et al., 2017) and this investigation. Once the organisms were on deck, they were transported to the laboratory, identified, and rinsed four times with double deionized water. The edible tissues were removed from each individual, mixed thoroughly other samples from the same species, freeze-dried to a constant, then homogenized, and placed in plastic zip-lock bags at − 20 °C until further analysis.

Zooplankton samples were collected simultaneously with marine organism samples at the same sampling areas. These samples were pretreated using methods described in a previous study (Ning et al., 2016) and frozen at − 20 °C until stable isotope analysis. The nitrogen and carbon isotopes of the zooplankton samples were measured and used as baselines to calculate the trophic levels of the marine organism species.

2.2. Chemical analysis

The water content in edible tissues was measured gravimetrically by comparing the difference in weight before and after freeze-drying to a constant weight. Total metal digestion was conducted and measured based on procedures in a previous study (Gu et al., 2015a) with slight modifications. Approximately 0.2–0.3 g of each freeze-dried and homogenized biota sample was predigested using 8 mL concentrated HNO_3 (65%) in a high-pressure Teflon bomb at room temperature. The sample was covered with a watch glass overnight to prevent disturbance.

Predigested samples were treated with 2 mL H_2O_2 (30%) and then digested on the Ethos Plus microwave laboratory station (Milestone, Sorisole, Italy) at the following temperature stages: 30–200 °C for

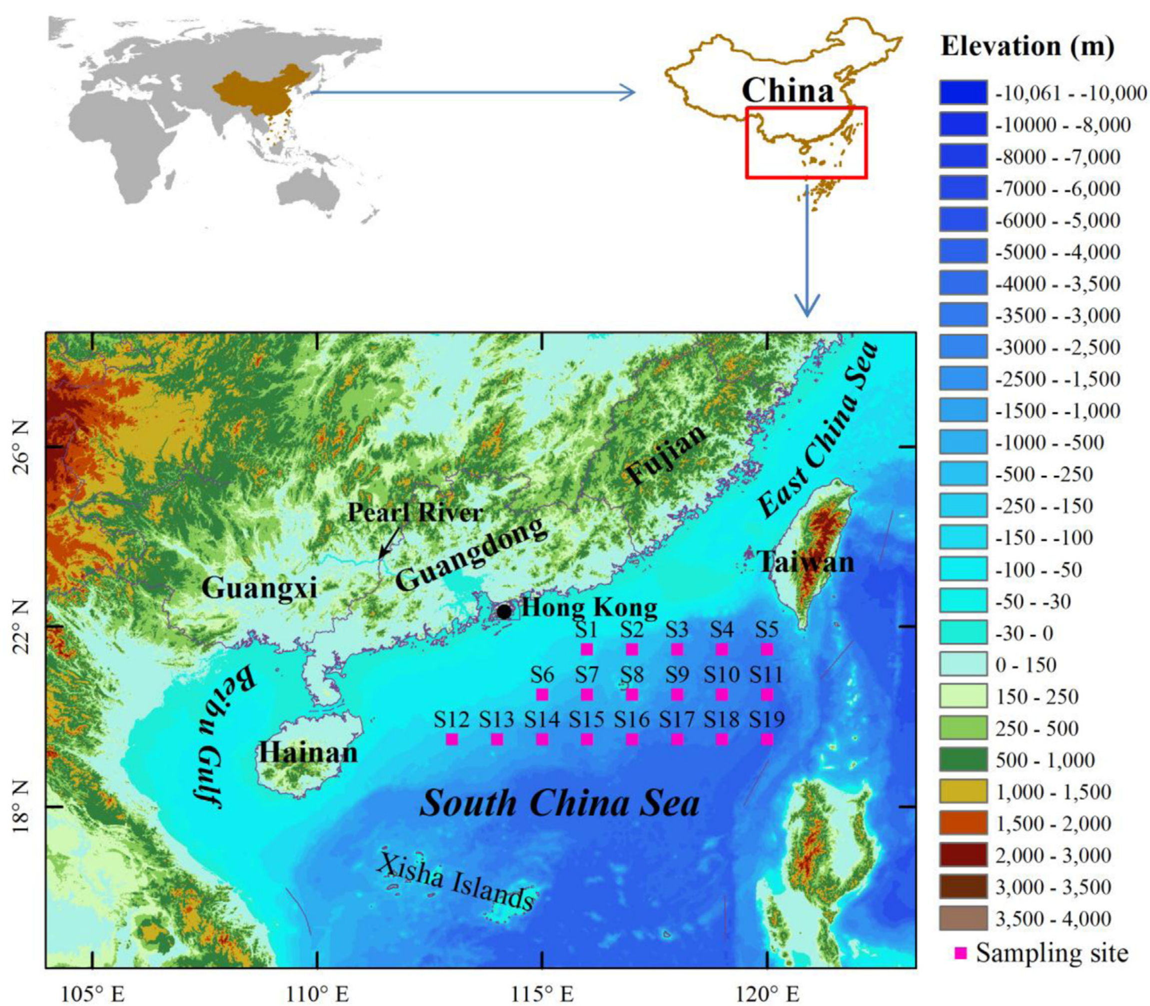

Fig. 1. Study area and sampling stations in South China Sea. Gridded elevation data are from http://www.bodc.ac.uk/data/online_delivery/gebco/.