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Biological risk assessment of heavy metals in sediments and health risk assessment in bivalve mollusks from Kaozhouyang Bay, South China

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ABSTRACT

The concentrations of heavy metals (Cd, Pb, Cr, Ni, Cu, Zn, Hg and As) in surface sediments and bivalve mollusks in Kaozhouyang Bay were investigated. A biological risk analysis of the sediments indicated that ten sites (about 76.92% of the total number of sites) had a 21% incidence probability of toxicity. A health risk analysis of the bivalve mollusks indicated that Cu and As posed low risks to consumer health. On the basis of the target hazard quotient (THQ), adverse effects may occur based on total THQ (TTHQ). The highest TTHQ was found in the species, *Ostrea rivularis*, which had the highest capacity for the bioaccumulation (factor > 38) of Cd.

Posing serious risks to biota and humans when threshold concentrations are exceed (Long, 2006; Yi et al., 2011; Gu et al., 2016a), heavy metals are deadly pollutants characterized by their persistence, toxicity, and non-degradability in the environment (Gu et al., 2012a; Lu et al., 2017). The significance of heavy metals in coastal ecosystems originates from their potential adverse effects and excessive anthropogenic sources, which can equal or exceed the natural input (Pan and Wang, 2012; Zuykov et al., 2013; El Nemr et al., 2016). In this regard, the relationships between the concentrations of pollutants in the sediments and bivalve mollusks within area can be used as effective tools to evaluate contamination levels and risks to human population (Fan et al., 2002; Kumar et al., 2015; El Nemr et al., 2016).

Bivalve mollusks have high capacities for bioaccumulating heavy metals from their aquatic environments (Maanan, 2008; Zuykov et al., 2013; Liu et al., 2017). Bivalves, especially mussels and oysters, are considered reliable bioindicators of the bioavailability of heavy metals (Amoozadeh et al., 2014; Schöne and Krause, 2016; Lu et al., 2017) and have been used as sentinel organisms in many metal biomonitoring studies (Dondero et al., 2011; Zuykov et al., 2013).

The rapid development of industry and agriculture in China has been coupled with increasing heavy metal pollution of aquatic ecosystems (Yi et al., 2011; Hu and Cheng, 2013; Niu et al., 2013; Pan and Wang, 2012). When heavy metals enter an aquatic environment, they ultimately accumulate within sediments, which are major sinks, or, when environmental conditions change, are released from the sediments, which then act as sources, back into the overlying water to threaten the aquatic biota (El Nemr et al., 2016; Fang et al., 2016; Gu et al., 2016b). The bioaccumulation of sediment-associated metals by bivalve species can contaminate food webs and cause harm to human consumers of the organisms.

Guangdong is the province with the most developed economy in China. Kaozhouyang Bay, situated in the southeastern part of Guangdong Province near the Pearl River Estuary (Fig. 1), covers an area of 29.7 km^2 and has a water depth range of 0.1–1.1 m (OGCGP, 1989; Cai et al., 2005). There are seven rivers/streams discharging into this bay (Fig. 1). The main portion of the Chinese leather industry is strongly established in the surrounding areas of this bay, which is a typical and intensively managed maricultural base in eastern Guangdong. More than half of the seawater area in the bay is occupied by oyster or caged fish farms. However, the rapid development of industry and agriculture in the surrounding areas of the bay has resulted in the rapid deterioration of the marine environment (AOFH, 2017). The sedimentation rate (sediment deposition flux) has significantly increased over the last few decades (Yang-Guang Gu unpublished data), leading to changes in the geochemical properties of the surface sediments on an annual scale. To the best of our knowledge, a few studies, which were conducted more than 18 years ago, focused on the total metal concentrations in the surface sediments of Kaozhouyang Bay (Gan et al., 2002; Cai et al., 2005), whereas investigations into heavy metal bioaccumulation with the accompanying biological and health risks are scarce. Therefore, the present study aims to (1) survey the concentrations and biological risks of heavy metals in the surface sediments in

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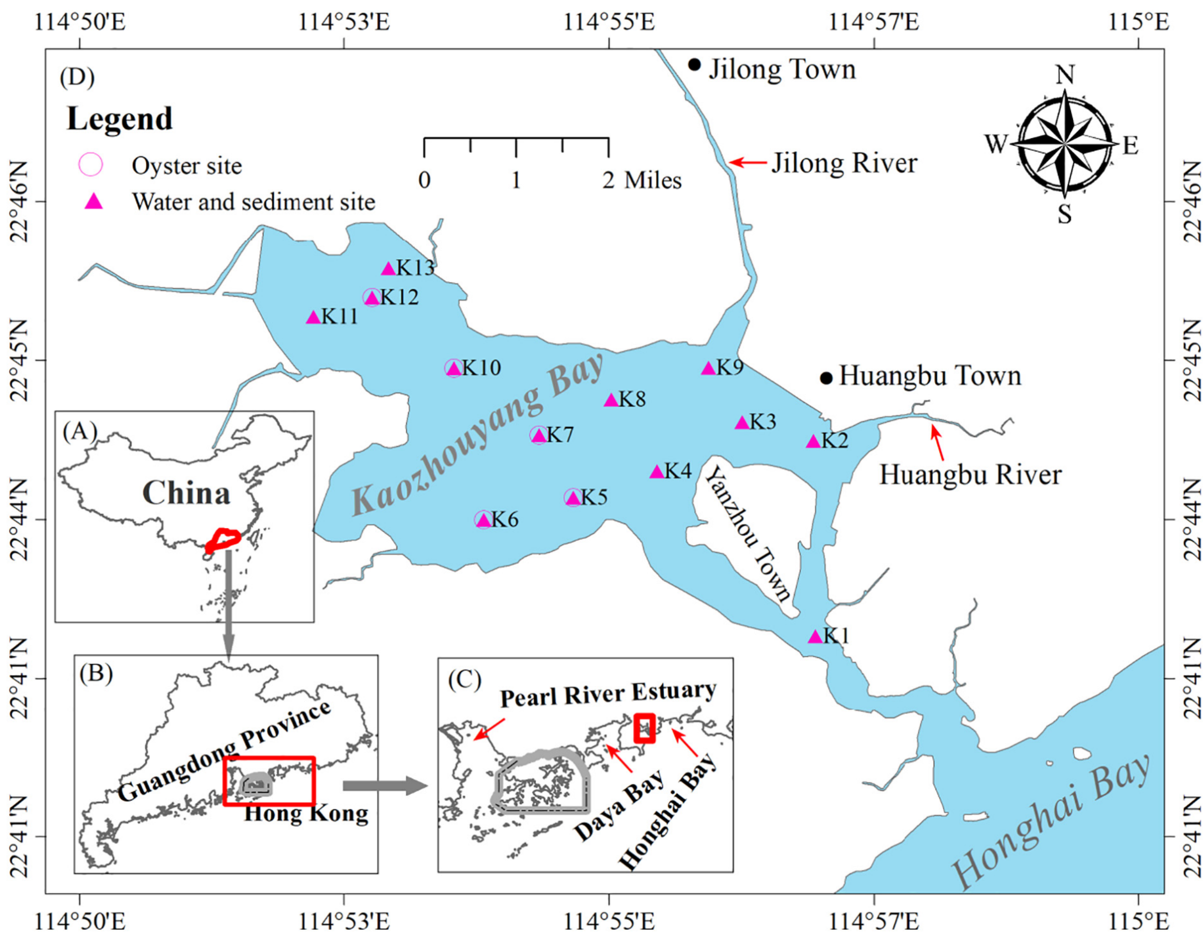

Fig. 1. Map of study area indicating location of Kaozhouyang Bay in China (A–C), and sampled sites in Kaozhouyang Bay (D).

Kaozhouyang Bay, (2) investigate the bioaccumulation by bivalve mollusks of the heavy metals released from the sediments, and (3) assess the health risks posed by the consumption of the bivalve mollusks.

Surface sediment samples were collected in September 2017 from thirteen sampling sites distributed throughout Kaozhouyang Bay (Fig. 1). Samples from each site were obtained using a 0.025 m^2 Van Veen grab sampler and placed into self-sealing polyethylene bags to be transported to a laboratory, where they were freeze-dried to a constant weight, ground with an agate mortar and pestle, sieved through a 63-μm mesh, and stored at −20 °C in brown glass bottles. Heavy metal microwave digestion was conducted in accordance with the EPA Method 3050B using an Ethos Plus Microwave Labstation (Milestone Inc., Italy).

As the sediment samples were gathered, specimens of three marine bivalve species (Fig. 1 and Table S1) were collected into plastic bags and placed within an icebox for immediate transportation to a laboratory, where the specimens were washed with both seawater and double distilled water. The edible tissues of each bivalve species were dissected, freeze-dried, homogenized, sieved through a 150-μm nylon mesh, and stored in brown glass bottles at −20 °C. The procedures for the metal extraction used in this study are described elsewhere (Gu et al., 2015a).

To verify sediment digestion recovery, the Chinese National Standard material (Offshore Marine Sediment, GBW 07314) was analogously evaluated. The recoveries of the metals ranged between 93%–109%.

Reagent blanks were used to correct the results of the analyses of the bivalves. Analyses of eight metals were performed on the Chinese National Standard scallop material (GBW10024), as well as on samples for quality control. Recoveries ranged from 89% to 105%. The concentrations of Cd, Pb, Cr, Ni, Cu, and Zn were measured by an atomic absorption spectrophotometer (AAS, Z2000, Hitachi, Japan) while the concentrations of Hg and As were determined by an atomic fluorescence spectrophotometer (AFS, 9230, Beijing Titan Instruments, China).

Prior to principal component analysis (PCA), a Q–Q plot was used to evaluate the normality of the dataset of each variable. The Q-Q plot data exhibited normal distributions of the concentrations of Ni, the metals (Cd, Cr, Cu, Hg, and As) that were transformed using the Box-Cox Equation, and those (Pb and Zn) that were log-transformed. The non-transformed and transformed variables were standardized for PCA while statistical analyses were performed using SPSS 19.0 for Microsoft Windows.

The heavy metal concentrations (mg/kg) displayed wide variations: Cd, 0.02–0.30; Pb, 10.89–67.70; Cr, 12.63–52.87; Ni, 5.46–20.68; Cu, 1.15–29.17; Zn, 29.29–137.52; Hg, 0.01–0.23; As, 2.92–12.38. The mean metals decreased in the order of Zn > Pb > Cr > Ni > As > Cu > Cd > Hg. The distribution patterns of eight heavy metals in surface sediments from Kaozhouyang Bay are illustrated in Fig. 2. Cd, Ni, Zn, Hg, and As were found to have similar distribution and their highest concentrations were found at K2, implying that these five metals may have been derived from the same source. The highest concentrations of Pb, Cr, and Cu were found at K9, K13, and K10, respectively. These concentrations may be ascribed to the downstream movements and depositions of suspended sediments containing heavy metals combined with local pollution. Compared to the average concentrations of most heavy metals in other bays in China, those in the surface sediments of Kaozhouyang Bay are lower than those in Qinzhou Bay, Daya Bay, Xiamen Bay, Hangzhou Bay, and Jiaozhou Bay (Fig. 3).