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Bioaccumulation of heavy metals and health risk assessment in three benthic bivalves along the coast of Laizhou Bay, China

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ABSTRACT

This study investigated the tissue- and species-specific bioaccumulation of heavy metals (Cr, Cu, Hg, Zn, As, Cd, and Pb) in three benthic bivalves (the ark shell, *Scapharca subcrenata*; the surf clam, *Macra veneriformis*; and the Manila clam, *Ruditapes philippinarum*) collected from the coast of Laizhou Bay in the Bohai Sea. The results demonstrated that the visceral masses of the bivalves tended to accumulate heavy metals more efficiently than their muscles. The capacities of the bivalves to bioaccumulate metals followed a similar order: Cd > Hg > Zn = As > Cu > Cr = Pb. The conditions of metal contamination in the bivalves tended to be worse along the eastern coast than in other regions. Overall, the Manila clam was more severely contaminated by heavy metals than the surf clam and ark shell. Judging by the hazard quotients (HQ) of the metals in the muscles of the bivalves, the greatest hazard risk to human health comes primarily from As.

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1. Introduction

Bivalves are globally widespread benthic species that mostly inhabit coastal and estuarine regions. Their high capacity to bioaccumulate aquatic pollutants has made them well-established bio-indicators for monitoring the contamination and transference of pollutants between media in the aquatic environment. (Sarkar et al., 2008; de Souza et al., 2011; Shoults-Wilson et al., 2015; Won et al., 2016). Heavy metals are generally considered to be highly toxic and potentially hazardous contaminants because of their stable persistence in aquatic environments, easy bioaccumulation in the biotas and bio-magnification in food chains (Tam and Wong, 1997; Valdes et al., 2014).

Bivalves, and particularly mussels and oysters, have been extensively studied and used to monitor metal contamination in coastal systems and to reveal the bioavailability of contaminants. (Sericano, 2000; Apeti et al., 2010; Shoults-Wilson et al., 2015). However, mussels and oysters are sessile species that live on the surface of reefs and rocks and mainly feed on suspended substances and plankton in the water. Thus, they can be regarded as bio-indicators of the ambient contamination status of water rather than sediments. Benthic species (e.g., clams) generally inhabit sediments and primarily feed on organic detritus, benthic diatoms and zoobenthos. They tend to accumulate heavy metals more efficiently than sessile species because these contaminants are more likely to be

enriched in sediments (Zhao et al., 2012; Li and Gao, 2014; Won et al., 2016). While previous studies have usually focused on the distribution and bioaccumulation of heavy metals in sessile bivalves for monitoring marine pollution, concerns about the bioaccumulation of pollutants in benthic species have increased in recent years (Tarique et al., 2012; Zhao et al., 2012; Velez et al., 2015).

Laizhou Bay is a shallow embayment located in the southern Bohai Sea, the largest inland sea in China. The bay is occupied by vast estuarine areas because many rivers (particularly the Yellow River) flow into it (Liu et al., 2013). Due to the input of indispensable nutrients from the rivers, the bay was once an important spawning and nursery ground for many marine organisms. In the past decades, however, industrialization and urbanization have developed rapidly around the bay. A large part of its coastal area is now occupied by ocean facilities such as harbors, industrial hubs and sea farming facilities, leading to the discharge of large amounts of domestic sewage and industrial effluents with various pollutants into the bay. These pollutants produce considerable adverse effects on coastal biotas, especially on the bivalves living in the benthic environment. Pollution is thus believed to be one major cause for the decline in the living aquatic resources in this region. More concerning, pollution of these bivalves may constitute unpredictable hazardous risks to human health (Li and Gao, 2014; Jitar et al., 2015). Several studies conducted to date have documented and provided general information about the contamination of heavy metals in some of the bivalves and sediments in Laizhou Bay and its adjacent areas (Liang et al., 2004; Wang et al., 2005). However, comparative studies on the bioaccumulation of heavy metals in bivalves in terms of inter-

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specific patterns, geographical distribution, and the relationships between the bivalves and the sediment as well as assessments of the risk to human health have largely not been undertaken.

To address these problems, we investigated the bioaccumulation of metals in three commercial benthic bivalve species: the ark shell (*Scapharca subcrenata*), the surf clam (*Macra veneriformis*) and the Manila clam (*Ruditapes philippinarum*), collected from the coast of Laizhou Bay. These three species are the most cultured and consumed benthic bivalves in local coastal areas with a total annual yield of approximately 5.1×10^5 t, while other highly consumed bivalves are sessile species (e.g., scallops, oysters and mussels) (SBSP, 2010). The concentrations of seven heavy metals (Cr, Cu, Zn, As, Cd, Hg and Pb) in two tissues (muscles and visceral masses) of the bivalves and the associated sediments were studied to achieve the following objectives: 1) investigate the tissue- and species-specific bioaccumulation of heavy metals in the bivalves; 2) examine whether the levels of heavy metals in the bivalves are correlated with those in the associated sediments; 3) examine the geographic variation of heavy metals in the bivalves using the metal pollution index (MPI) and hierarchical cluster analysis (HCA); and 4) assess the risk of heavy metals in the bivalves to human health on the basis of national or international guidelines to determine the provisional tolerable weekly intake (PTWI) limit and hazard quotient (HQ).

2. Materials and methods

2.1. Sample collection and preparation

Samples of the three bivalves (ark shell, AS; surf clam, SC; Manila clam, MC) were collected from nine sites (S1–S9) in the coastal areas of Laizhou Bay from June to August 2011 (Fig. 1). These sites represented the main estuarine areas showing regional characteristics in terms of the coastal terrain, industrial development and living aquatic resources. To investigate the geographical variation in the bioaccumulation of the

heavy metals, the sampling sites were grouped into eastern (S1–S3), southern (S4–S5) and western (S6–S9) coastal zones in this study.

After collection, the attached mud, barnacles and other debris on the surface of the shells were thoroughly removed using a stainless steel brush. The bivalves were then placed in polyethylene buckets for *in vivo* purging of the mud, sand and other waste in the guts. The shell lengths and weights of the bivalves were then measured and recorded. Two hundred individuals of similar size (ark shell, 34–38 mm and 11.2–13.5 g; surf clam, 35–40 mm and 8.3–11.6 g; Manila clam, 37–42 mm and 8.9–12.3 g) were sampled and then frozen at -20°C . Additionally, sediment samples were collected *in situ* from the upper 0–10 cm at each sampling site and stored in polyethylene bags at 4°C . Both the bivalves and sediment samples were stored until chemical analysis.

In the laboratory, the sediment samples were dried at 40°C in an oven, then ground to powder and sifted through a 100-mesh sieve to remove the shell fragments for chemical analysis. The thawed bivalves were dissected, and two tissue samples (muscle and visceral mass) were collected from each individual. The visceral masses of 15 individuals of each species were grouped and homogenized as a single mixed sample to obtain a sufficient tissue mass for chemical detection. The muscle tissues (including the mantles and feet) from 3 individuals of each species were grouped and homogenized as a single mixed sample. Five mixed samples were chemically analysed for each sampling site. The tissue samples were then lyophilized and ground into powder for chemical analysis. Both the wet and the frozen-dried tissue samples were weighed to calculate the ratio of dry mass to wet mass for use in risk assessment.

2.2. Chemical analysis

Chemical analyses of the tissues and sediment samples were conducted as described previously (Liu et al., 2013). Briefly, the tissues were digested using HNO_3 (Superpure, Merck, Germany) in Teflon bombs. The concentrations of six heavy metals (Cr, Cu, Zn, As, Cd, and

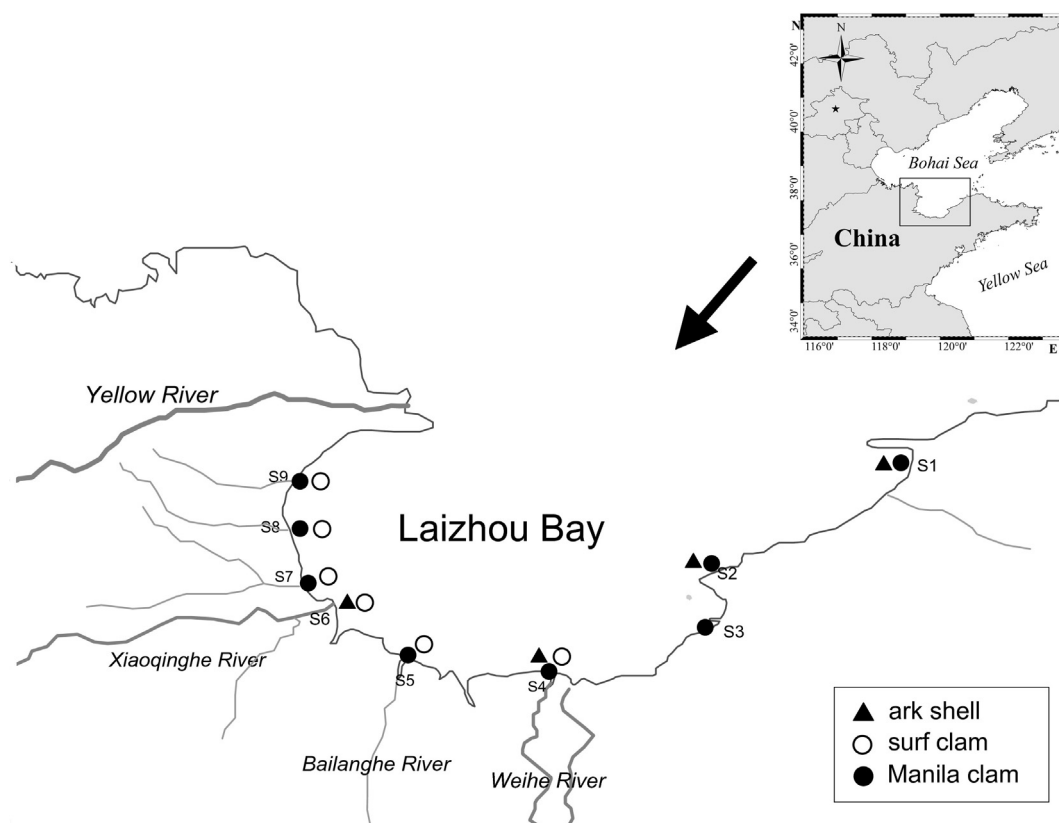


Fig. 1. Study area and sampling sites of the three bivalves from Laizhou Bay. S1, Longkou; S2, Sanshandao; S3, Zhuwang; S4, Xiaying; S5, Yangzi; S6, Yangkou; S7, Guanglihe; S8, Yongfenghe; S9, Xiaodaohe.

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