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Research article

# Heavy metal contamination along the China coastline: A comprehensive study using Artificial Mussels and native mussels

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## ABSTRACT

A comprehensive study was carried out to assess metal contamination in five cities spanning from temperate to tropical environment along the coastal line of China with different hydrographical conditions. At each of the five cities, Artificial Mussels (AM) were deployed together with a native species of mussel at a control site and a polluted site. High levels of Cr, Cu and Hg were found in Qingdao, high level of Cd, Hg and Pb was found in Shanghai, and high level of Zn was found in Dalian. Furthermore, level of Cu contamination in all the five cities was consistently much higher than those reported in similar studies in other countries (e.g., Australia, Portugal, Scotland, Iceland, Korea, South Africa and Bangladesh). Levels of individual metal species in the AM showed a highly significant correlation with that in the native mussels (except for Zn in *Mytilus edulis* and Cd in *Perna viridis*), while no significant difference can be found between the regression relationships of metal in the AM and each of the two native mussel species. The results demonstrated that AM can provide a reliable time-integrated estimate of metal concentration in contrasting environments over large biogeographic areas and different hydrographic conditions, and overcome the shortcomings of monitoring metals in water, sediment and the use of biomonitors.

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## 1. Introduction

Traditional monitoring of heavy metals in the aquatic environment involves determining metal concentrations in water or sediment, but both methods present their own problems and limitations. The concentration of metals in water is typically low with large temporal fluctuations. Frequent sampling is therefore required to provide a representative estimate, while chemical analysis of a large number of samples may not be cost effective (Philips and Rainbow, 1993; Rainbow, 1995). Metal concentrations in sediment provides a time-integrated estimate of metal levels, but are significantly affected by particle size, organic content and redox conditions, which cannot be standardized (Philips and Rainbow, 1993). Due to their remarkable ability to concentrate metals from the ambient environment, biomonitors (especially mussels and barnacles), have been commonly employed to monitor and compare temporal and spatial changes of heavy metals in

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aquatic environments since the 1970's (Kennish, 1997; Rodriguez and Thebault, 2007), and this is exemplified by the global "mussel watch" program (Goldberg, 1975; Goldberg et al., 1978; Kimbrough et al., 2008). However, metal accumulation in biomonitors is not only species specific, but also significantly affected by physical and biological factors as well as pollution levels (Leung et al., 2001, 2002; Philips and Rainbow, 1993; Wu et al., 2005). This presents great difficulties in comparing biomonitors under different hydrographic conditions and seasons. Moreover, the limited natural distributions of species often prevent making comparison over large geographic areas.

To overcome the above limitations in metal monitoring, a passive sampling device known as the Artificial Mussel (AM), was developed (Wu et al., 2007). Results of laboratory studies demonstrated that the AM can uptake and release Cd, Cr, Cu, Pb, Zn and Hg in a concentration-dependent manner, including the bioavailable fraction. Field studies further showed the AM is field robust, and can provide a time-integrated estimate of metal concentrations in the environment, exhibiting a metal profile similar to that of the green-lipped mussel (*Perna viridis*) at the control site and the polluted site (Wu et al., 2007). Leung et al. (2008) deployed the AM alongside the blue mussel (*Mytilus edulis*) in Scotland and Iceland, and demonstrated that AMs can provide good indication on the dissolved metal (Cd, Cr, Cu, Hg, Pb, Zn) fractions in the marine environment. Degger et al. (2011) studied the metal accumulation profile of AMs deployed alongside with the brown mussel (*Perna perna*) in three distinct geographical regimes in South Africa, identifying pollution hot spots and demonstrating the effectiveness and robustness of the AM in metal monitoring over large biogeographical area. Likewise, a field study in Portugal (Gonzalez-Rey et al., 2011) also demonstrated that the AM had similar metal profiles to the Mediterranean mussel (*Mytilus galloprovincialis*), and concluded that the AMs can be used successfully to monitor metals under different hydrographical regimes. Kibria et al. (2012) employed AMs for monitoring heavy metal in water catchment areas of Australia and identified input and hotspots of metals for risk assessment. Ra et al. (2014) showed significant correlations between accumulation of Cd, Cu and Pb in AMs and mussel (*Mytilus edulis*) at all study sites in Korea, and concluded that AM provides a better time integrated estimates for dissolved metal concentration. The above studies showed that the AMs can serve as a reliable and robust tool to monitor metals and assess risk in both the marine and freshwater environments.

China has one of the longest coast lines (14,500 km) in the world, stretching from Guangxi in the south (18°15′N) to Liaoning (53°30′) in the north. The rapid industrialisation and urbanisation of China in the last three decades saw an increased load of wastes from the industry and mining tailings to its coastal environments (Pan and Wang, 2012). A number of "hot spots" with high levels of contamination have been identified along the coast of China, including Liaodong Bay, Xiamen Bay, Pearl River Delta and the Yangtze River catchment (Li et al., 2012). In this study, five cities spanning from temperate to tropic environment along the China coast and with different hydrographic conditions, were selected, and at each of the five cities, AM were deployed together with a native species of mussel at a control site and a polluted site. The objectives were to: (a) provide a systematic account and comparison on the levels of metal contaminations along the China coast; and (b) test and compare the performance of AMs with native mussels under contrasting hydrographical conditions and different pollution levels.

## 2. Materials and methods

Five cities along the coastline of China, spanning from temperate to tropical (from north to south: Dalian, Qingdao, Shanghai, Shantou and Shenzhen), were selected for the present study (Fig. 1).

Dalian has a population of 6 million people and the coastal area has become increasingly threatened by pollution generated from industry, shipyards, fuel, aquaculture and domestic waste, and heavily contaminated by Zn, Pb and Hg (Mao et al., 2009; Zhao et al., 2012).

Qingdao is situated in Jiaozhou Bay, a semi-enclosed coastal water body on the east coast of China. Petroleum contamination, untreated industrial and municipal water discharges and run-off form the major anthropogenic sources of heavy metals entering the Bay (Wang et al., 2007), and increased levels of metals such as Cu, Pb and Cr have been found (Wang et al., 2006).

With 24 million people, Shanghai is the largest city in China and also one of the largest cities in the world. Numerous industrial enterprises and over 10,000 factories proliferated in the city in the last two decades, making coastal pollution an imminent problem (Cao et al., 2008; Wang et al., 2009), and high levels of Cd, Cr, Ni and Pb have been reported in marine sediments (Yuan et al., 2004; Zhang et al., 2009).

A recent study conducted by Qiao et al. (2013) in Shantou Bay revealed elevated concentrations of heavy metals in marine sediment, which have been largely attributed to agricultural practices along the Rongjiang River basin, the introduction of industrial and municipal discharges from the Meixi River from Shantou City.

Shenzhen is amongst the fastest developing city in the world in the last 20 years, where industrial and land reclamation occur in an enormous scale and contributed to high heavy metal levels (Pb, Cu and Zn) in Shenzhen Bay (Chen and Jiao, 2008; Huang et al., 2007) resulting in adverse effects on the marine environment (Huang et al., 2007).

### 2.1. Artificial Mussels and mussels deployment and collection

Within each of the above five cities, a reference site remote from pollution sources and urban activities, and a site receiving moderate to heavy pollution loading, were selected for this study (Table 1). At each of these ten sites, 60 AMs secured in a plastic cage were deployed at 0.5–1.0 m below chart datum. Concurrently, over 100 native mussels (i.e. the blue mussel *Mytilus edulis* for Dalian, Qingdao and Shanghai; and the green-lipped mussel *Perna viridis* for Shantou and Shenzhen) were collected from a clean site at the respective city. *M. edulis* (umbo length: 3.5–6 cm) and *P. viridis* (umbo length: 5.5–8 cm) were acclimated in clean seawater for a month and deployed in a nylon bag alongside the AMs in June 2011.

Six AMs and eight native mussels were randomly collected from each site 30 days after deployment for metal analysis. Another six AMs were further collected from each site 90 days after deployment for metal analysis, to provide information on temporal variations of metal concentration.

### 2.2. Metal analysis

All metal analyses were conducted following the protocol described in Wu et al. (2007). Briefly, the contents of each individual AM were emptied into a sintered glass filter followed by eluting two times with 12.5 mL 6 M $HNO_3$ (analytical grade). The elutriant was made up to a known volume with deionized double distilled water and the concentrations of Cd, Cr, Cu, Hg, Pb and Zn were determined, using a Perkin Elmer Optima 2100 DV ICP-AES (Plasma flow: 15 L/min; auxiliary flow: 0.3 L/min; nebulizer flow: 0.8 L/min; RF flow: 1300 W, pump rate: 1.0 mL/min). Metal standard solution (1000 μg/mL in 2% nitric acid) from"High Purity Standards" was used as calibration standards for construction of calibration curves.

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