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Trace elements in major marketed marine bivalves from six northern coastal cities of China: Concentrations and risk assessment for human health

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ABSTRACT

One hundred and fifty nine samples of nine edible bivalve species (*Argopecten irradians*, *Chlamys farreri*, *Crassostrea virginica*, *Lasaea nipponica*, *Meretrix meretrix*, *Mytilus edulis*, *Ruditapes philippinarum*, *Scapharca subcrenata* and *Sinonovacula constricta*) were randomly collected from eight local seafood markets in six big cities (Dalian, Qingdao, Rizhao, Weifang, Weihai and Yantai) in the northern coastal areas of China for the investigation of trace element contamination. As, Cd, Cr, Cu, Hg, Pb and Zn were quantified. The risk of these trace elements to humans through bivalve consumption was then assessed. Results indicated that the concentrations of most of the studied trace element varied significantly with species: the average concentration of Cu in *C. virginica* was an order of magnitude higher than that in the remaining species; the average concentration of Zn was also highest in *C. virginica*; the average concentration of As, Cd and Pb was highest in *R. philippinarum*, *C. farreri* and *A. irradians*, respectively. Spatial differences in the concentrations of elements were generally less than those of interspecies, yet some elements such as Cr and Hg in the samples from different cities showed a significant difference in concentrations for some bivalve species. Trace element concentrations in edible tissues followed the order of Zn > Cu > As > Cd > Cr > Pb > Hg generally. Statistical analysis (one-way ANOVA) indicated that different species examined showed different bioaccumulation of trace elements. There were significant correlations between the concentrations of some elements. The calculated hazard quotients indicated in general that there was no obvious health risk from the intake of trace elements through bivalve consumption. But care must be taken considering the increasing amount of seafood consumption.

1. Introduction

The Earth system is currently operating in a no-analogue state, and human activities are significantly altering the environment on a global scale (Steffen et al., 2004). Massive amounts of inorganic/organic substances are discharged into the environment each year, transforming into pollutants and usually having negative effects. Trace elements are natural and fundamental components of the geosphere and biosphere. Many trace elements are essential to maintain the metabolism of organisms. However, they can be poisonous if their concentrations exceed certain limits. Once entering the environment, these elements cannot be degraded or destroyed but tend to bioaccumulate and increase in the concentrations over time, pass along the food chain and directly/indirectly influence human health. Many environmental quality guidelines, within which trace elements such as arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), lead (Pb) and zinc (Zn) are usually used as criteria, have been developed to deal with environmental concerns as well as to respond to regulatory programs (e.g. Rawson and Burton, 2002).

Fishery products are a very valuable source of protein and can provide essential micronutrients for human beings. The Food and Agriculture Organization of the United Nations (FAO) reported that fisheries and aquaculture supplied the world with about 128 million tons of food for people in 2010 (FAO (Food and Agricultural Organization of the United Nations), 2012). The share of aquaculture in the world's total fishery yield has been increasing significantly in recent years. China has been the world's leading aquaculture producer for decades and the aquaculture production accounted for 82 percent of the world's marine farmed

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fishery yield in 2010 (FAO (Food and Agricultural Organization of the United Nations), 2012). Marine aquaculture production accounted for 39 percent of the total aquaculture production of China in 2010, about 70 percent of this was comprised of bivalves such as clams, oysters, scallops, mussels and cockles. As the country's first- and fourth-biggest marine aquaculture producers, respectively, Shandong and Liaoning Provinces together contributed 42.4 percent of the marine aquaculture production in China in 2011 according to the report from the BFMAC (Bureau of Fisheries of the Ministry of Agriculture of China) (2012).

The coastal zone is the main area for marine bivalve farming; meanwhile it traps a majority of pollutants in the process of transporting pollutants from land to sea. Most marine bivalves bury themselves in sediment on the seabed, some lie on the sea floor or attach themselves to rocks or other hard surfaces and a few can swim a short distance. Their living habits make them good indicators of environmental quality on a local/regional scale.

The entire coast of the Bohai Sea and the coast of the Yellow Sea in Shandong and Liaoning Provinces are located in the Bohai Sea Economic Rim (Fig. 1), one of the three most densely populated and industrialized zones in China (Gao et al., 2014). Extraordinarily rapid industrial and agricultural development in the northern coastal areas of China has resulted in a significant anthropogenic increase in the ambient levels of pollution and environmental damage in the marine ecosystem (Australian Agency for International Development, 1996; Jacinto, 1997; Xu et al., 2010; Zhang et al., 2010; Gao et al., 2014). Previous reports indicated that the edible marine bivalve species sampled from the Bohai Sea had been contaminated with trace elements to different degrees (Liang et al., 2004; Wang et al., 2005; Du et al., 2009). However, the purpose of these studies was merely monitoring the environmental quality, and there was a lack of risk assessment concerning human health from seafood consumption.

In this study, the edible tissues of nine most marketed marine bivalve species collected from six northern big coastal cities of China, namely Dalian, Qingdao, Rizhao, Weifang, Weihai and Yantai, were analyzed for the determination of As, Cd, Cr, Cu, Hg, Pb and Zn. The sampled cities were all located in the Bohai Sea Economic Rim of China. The main objectives of this study were to examine whether the widely consumed bivalve species were contaminated with trace elements, and to test whether there were any spatial or interspecific variations in the concentrations of trace elements. Health risk assessment was then conducted to evaluate whether these bivalves posed any potential risk to human beings as a result of consumption. From a public health perspective, another aim of this study was to provide consumers with better knowledge of contamination problems associated with seafood consumption. We also tried to compare the trace element results with those obtained from other studied coastal areas.

2. Materials and methods

2.1. Sampling

One hundred and fifty nine samples of nine marine bivalve species of similar sizes were randomly collected from eight large seafood markets in six big cities (Dalian, Qingdao, Rizhao, Weifang, Weihai and Yantai) in the northern coastal areas of China during 6th–27th November 2011 (Fig. 1). To ensure randomness and sufficient representativeness of sampling, 3–4 samples for each species from each city and 10–50 individuals with similar body lengths for each sample were collected. The samples were purchased with their usual conditions in markets and transferred to the laboratory in a cooler box with ice packs. The sampled bivalve species are identified as follows: *Argopecten irradians*, *Chlamys farreri*, *Crassostrea virginica*, *Lasaea nipponica*, *Meretrix meretrix*, *Mytilus edulis*, *Ruditapes philippinarum*, *Scapharca subcrenata* and *Sinonovacula constricta*.

2.2. Sample analysis

In the laboratory, the edible tissues of the collected bivalves were then dissected, rinsed with deionized water (18.2 MΩ-cm) three times to remove extraneous impurities, and any excess rinsing water was drained off and vaporized. The wet weights of all edible bivalve tissues were first recorded, and the tissues were then freeze dried until constant weights, after which their dry weights were recorded. The percentages of water were calculated and used to convert the trace element concentrations of the samples from a dry weight basis to a wet weight basis. The dried samples were homogenized and ground with a ceramic mortar and stored in small polyethylene zipper bags at −20 °C until further analysis.

For each sample, ~0.3 g of dried and ground soft tissues was weighed and added to a polytetrafluoroethylene (PTFE) digestion container with known weight. Each sample was added with 10 ml of concentrated nitric acid and left to predigest overnight at 80 °C. After cooling, the container was covered and placed in a high-pressure stainless steel bomb and then put in an oven. Thereafter the oven temperature was increased to 160 °C and kept for 8 h till clarification. After cooling to the room temperature, the solution was diluted with deionized water to the final weight of 50 g and then transferred into the polyethylene terephthalate (PET) bottle. As, Cd, Cr, Cu, Pb, and Zn were determined with the PerkinElmer ELAN DRC II

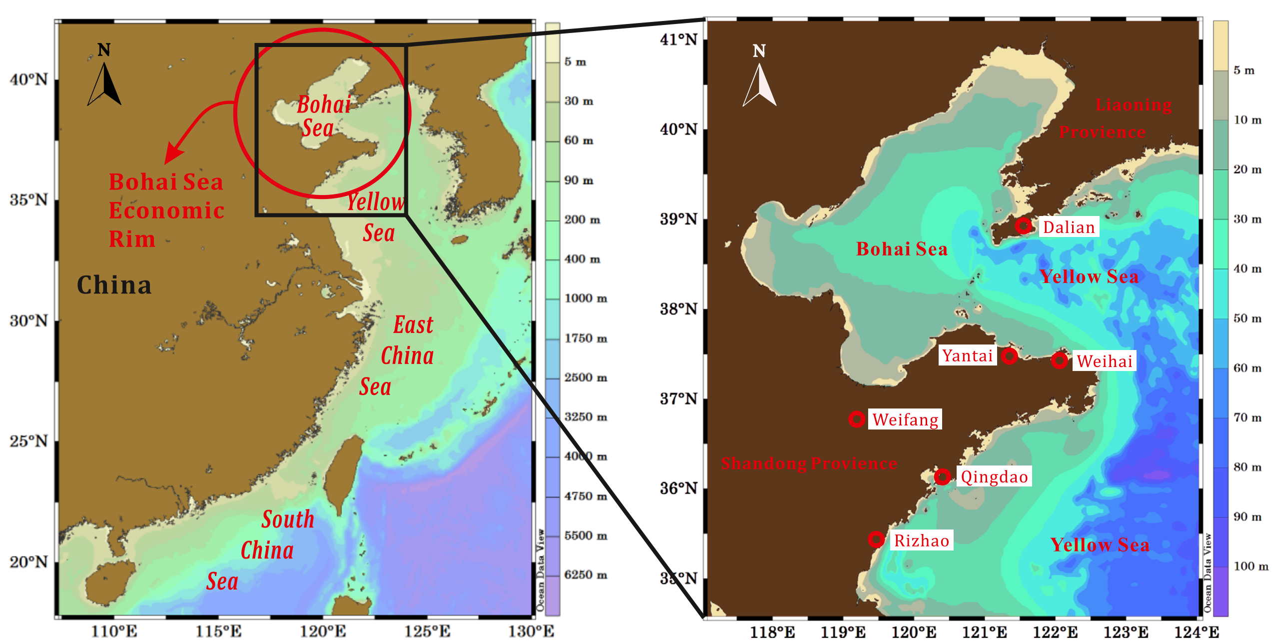

Fig. 1. Sampled cities along the northern coast of China.