Environmental Pollution 224 (2017) 658–669

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Oyster-based national mapping of trace metals pollution in the Chinese coastal waters☆

Guang-Yuan Lu [a, b], Cai-Huan Ke [b], Aijia Zhu [c], Wen-Xiong Wang [a, d, *]

[a] Marine Environmental Laboratory, Shenzhen Research Institute, The Hong Kong University of Science and Technology, Shenzhen 518000, China
[b] State Key Laboratory for Marine Environmental Science, Xiamen University, Xiamen 361005, China
[c] South China Sea Environmental Monitoring Center, State Oceanic Administration, Guangzhou 510300, China
[d] Division of Life Science, The Hong Kong University of Science and Technology (HKUST), Clearwater Bay, Kowloon, Hong Kong

ARTICLE INFO

Article history:
Received 13 December 2016
Received in revised form
21 February 2017
Accepted 21 February 2017
Available online 3 March 2017

Keywords:
Metal
Oyster
China
National mapping
Biomonitoring
Na/P normalization

ABSTRACT

To investigate the distribution and variability of trace metal pollution in the Chinese coastal waters, over 1000 adult oyster individuals were collected from 31 sites along the entire coastline, spanning from temperate to tropical regions (Bohai Sea, Yellow Sea, East China Sea and South China Sea), between August and September 2015. Concentrations of macroelements [sodium (Na), potassium (K), calcium (Ca), magnesium (Mg) and phosphorus (P)] and trace elements [cadmium (Cd), copper (Cu), zinc (Zn), nickel (Ni), lead (Pb), chromium (Cr), silver (Ag), and titanium (Ti)] in these oysters were concurrently measured and analyzed. The results showed high Ti, Zn and Cu bioaccumulation in oysters from Guangdong (South China Sea) and Zhejiang (East China Sea). Oysters at Nanji Island (Wenzhou) and Daya Bay (Huizhou) accumulated significantly high concentrations of Ni and Cr. The elements in these oysters were several times higher than the national food safety limits of China. On the other hand, the present study found that normalization of metals by salinity (Na) and nutrient (P) could reflect more details of metal pollution in the oysters. Biomonitoring of metal pollution could benefit from incorporating the macroelement calibration instead of focusing only on the total metal concentrations. Overall, simultaneous measurement of macroelements and trace metals coupled with non-linear analysis provide a new perspective for revealing the underlying mechanism of trace metal bioavailability and bioaccumulation in marine organisms.

1. Introduction

Metal contamination is one of the major concerns for aquatic ecosystems due to their vast source, persistence, non-degradability, bioaccumulation and inherent toxicity on living organisms, potential ecological effects, and public food safety (Le et al., 2016; Wang et al., 2013). Bivalve mollusks (including mussels, oysters, and clams) could accumulate metals presented at undetectable levels in water to very high tissue concentrations, and offer advantages as pollution amplifiers or sentinels of certain chemicals (Hu et al., 2016). At the beginning of 20th century, V.I. Vernadsky proposed to use bivalve mollusks as biomonitors based on the biogeochemistry (Zuykov et al., 2013). Since 1970s, a worldwide monitoring scheme of contaminants using mussels and oysters had been implemented (Davies and Pirie, 1980; Goldberg, 1975; Phillips, 1977; Watling and Watling, 1976) for assessing the ocean health. The National Oceanic and Atmospheric Administration (NOAA) of USA established a National Status and Trends (NS&T) Program that has been running for over 30 years for continuous contaminant monitoring in bivalves (mussels and oysters) and sediments. Resident and transplanted mussels have also been used to assess the patterns in contaminant bioaccumulation (Melwani et al., 2014). The bivalve monitoring now covers numerous geographical areas of worldwide coastal waters as a primary component of monitoring program (Aguilar et al., 2012; Cantillo, 1997; González-Fernández et al., 2015; Guitart et al., 2012; Hédouin et al., 2010; Lee et al., 2015; Monirith et al., 2003; Tanabe et al., 2000).

In China, many studies have measured the metal concentrations in different species of marine bivalves and reported the seriously metals polluted regions in the coastal waters and sediments (Cai et al., 2008; Chen et al., 2014; Pan and Wang, 2012; Wang et al.,

☆ This paper has been recommended for acceptance by David Carpenter.

* Corresponding author. Room 313-1, No. 9 Yuexing 1st Rd, HKUST SZ IER Building, South Area, Hi-Tech Pack, Nanshan, Shenzhen, 518057, China.
E-mail address: wwang@ust.hk (W.-X. Wang).

http://dx.doi.org/10.1016/j.envpol.2017.02.049
0269-7491/

2013). Such studies provided valuable information on metal contamination in coastal and estuarine waters of China. However, none has been conducted to mapping the bioavailable metal pollution in entire Chinese coastal waters, largely due to the difficulty of identifying suitable sentinel species across a large geographic area of coastal waters. Furthermore, given the tremendous complexity of coastal waters in different regions due to anthropogenic and naturally hydrographic variability, it was extremely challenging to compare the metal contamination levels from different coastal ecosystems. Some Chinese coastal waters are classified as eutrophic waters due to vast nutrient inputs (Chai et al., 2006; Qiu et al., 2010), and macronutrients are known to significantly affect the metal accumulation in phytoplankton and zooplankton which serve as the food sources for bivalves (Qiu et al., 2010; Wang and Dei, 2001; Yu and Wang, 2004). Liu and Wang (2015) demonstrated that macronutrient variation in biomonitors should be taken into account when interpreting trace element accumulations in worldwide mussel tissues. Like mussels, oysters are the second group of bivalve mollusks and widely employed as metal biomonitors for their strong net accumulators of many metals (Cu, Zn, Cd, and Ni) from ambient environment (George et al., 1978; Luoma, 1985; Okazaki and Panietz, 1981; Pan and Wang, 2012; Rainbow, 1995; Scanes, 1996). China has a thousand-year history of oyster culture as early as in the Song Dynasty. Today the production of oysters in China is up to >3 million tons every year, accounting for 80% of the total global aquaculture production (from Food and Agriculture Organization of the United Nations (FAO), 2014). However, the concentrations of metals in the oysters are complex and dependent on the oyster size and species, polluted degree, nutrient and geographical environment, as well as on metal speciation. How to scientifically measure and assess the metal concentrations in these oysters is an important question under variable and complicated environments, especially in estuarine regions.

This study investigated five macroelements and eight trace elements of ~1000 oysters in 31 representative sites of Chinese coastal waters. The objective of the study was to conduct a national mapping of elements in the oysters of China in order to (a) understand the baseline of spatial distribution and variation of element bioaccumulation in the oysters under different coastal and estuarine waters; and (b) to distinguish the bioaccumulation mechanisms of metals in the oysters under variable geographic environments. The study presented a new perspective for biomonitoring and risk assessment of metals in aquatic ecosystem.

2. Materials and methods

2.1. Oysters sampling

The Chinese coastal seas are divided into four major basins: Bohai Sea (BS), Yellow Sea (YS), East China Sea (ECS), and South China Sea (SCS). Over one-thousand of wild oysters from 31 sampling sites were collected at intertidal and subtidal zones along the coastline of China, spanning from temperate to tropical regions during August to September 2015 (18.2 ^{o}N ~ 40.5 ^{o}N; BS, Liaodong Bay, Xinkai Mouth, Bohai Bay, and Chang Island; YS: Guanglu Island, Zhangzi Island, Sanggou Bay, Rushan, Aoshan Bay, Jiaozhou Bay, Gold Beach, and Haizhou Bay; ECS: Xidian, Yueqing Bay, Nanji Island, Shacheng Harbour, Xiapu, Liushui Town, Pinghai Bay, Dadeng Island, Haimen Island, Dacheng Bay, and Nan'ao Island; SCS: Daya Bay, Bao'an Airport, Chengcun Town, Guandu, Longmen, Dafeng Estury, Waisha, and Yulin Harbour) (Fig. 1, Table 1). These sites were exposed to different anthropogenic impacts and hydrographic and geographical environments; detailed descriptions of the sampling sites are given in Table 1. The oysters were collected towards the

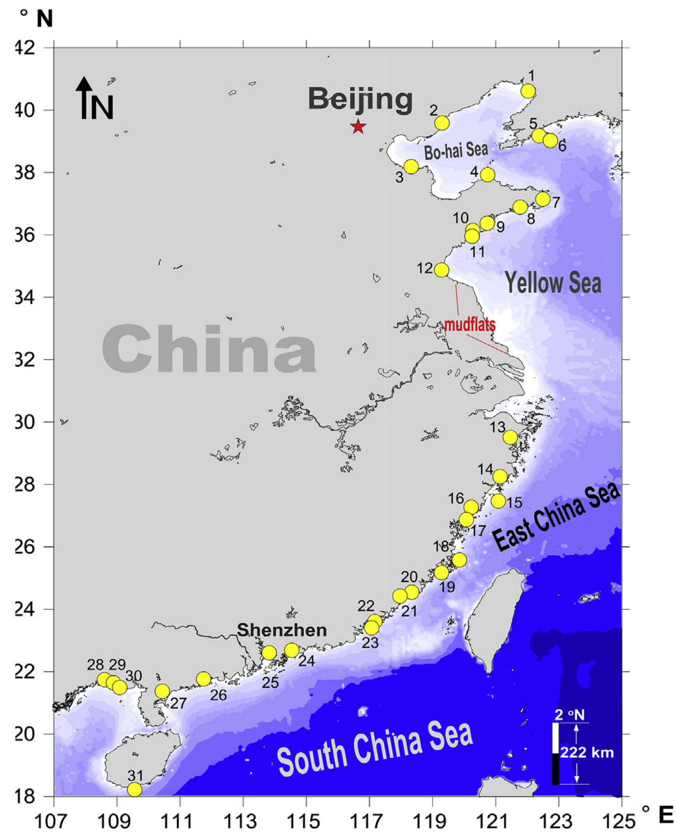

Fig. 1. Sample sites of oysters in China.
All sites in Chinese coastal waters: Bohai Sea (1. Liaodong Bay, Yingkou, 2. Xinkai Mouth, Qinghuangdao, 3. Bohai Bay, Dongying, 4. Chang Island, Yantai); Yellow Sea (5. Guanglu Island, Dalian, 6. Zhangzi Island, Dalian, 7. Sanggou Bay, Rongcheng, 8. Rushan, Weihai, 9. Aoshan Bay, Qingdao, 10. Jiaozhou Bay, Qingdao, 11. Gold Beach, Qingdao, 12. Haizhou Bay, Lianyuangang); East China Sea (13. Xidian, Ninghai, 14. Yueqing Bay, Wenzhou, 15. Nanji Island, Wenzhou, 16. Shacheng Harbour, Fuding, 17. Xiapu, Ningde, 18. Liushui Town, Pingtan, 19. Pinghai Bay, Putian, 20. Dadeng Island, Xiamen, 21. Haimen Island, Zhangzhou, 22. Dacheng Bay, Chaozhou, 23. Nan'ao Island, Shantou); South China Sea (24. Daya Bay, Huizhou, 25. Bao'an Airoport, Shezhen, 26. Chengcun Twon, Yangjiang, 27. Guandu, Zhanjiang, 28. Longmen, Qinzhou, 29. Dafeng Estuary, Beihai, 30. Waisha, Beihai, 31. Yulin Harbour, Shanya. 1°N in map is about 110.94 km. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

end of their reproductive seasons to avoid the influences of reproduction on metal concentrations. At each site, 50–60 adult individual oysters with similar shell length (about >4 cm) and reproductive stage were collected (total of 1500–1800 individuals) and ice-fresh transported under 10 °C to Shenzhen Marine Environmental Laboratory within 24 h. Then, the oysters were depurated at 25 °C for 2 days before the elemental analysis. We collected ordinary species *Crassostrea gigas* (Thunberg, 1793) in BS and YS coasts, *Crassostrea sikamea* (Amemiya, 1928) and *Crassostrea angulata* (Lamarck, 1819) in ECS coasts, *Crassostrea hongkongensis* (Lam and Morton, 2003) and *Crassostrea ariakensis* (Fujita, 1913) in SCS coasts. There were also other rare oyster species such as *Crassostrea* spp. (or *Ostrea*, uncertain), *Crassostrea nippona* (Seki, 1934), *Planostrea pestigris* (Hanley, 1846), and *Saccostrea cucullata* (Born, 1778) sampled at Jiaozhou Bay, Nanji Island, Daya Bay, and Yulin Harbour, respectively.

2.2. Elemental analysis

After depuration, the soft tissues of oysters were dissected and rinsed with Milli-Q water, and then homogenized (1–3 oysters) for