Bull Environ Contam Toxicol (2007) 78:535–538
DOI 10.1007/s00128-007-9083-0

Accumulation of Cadmium, Copper, Lead and Zinc in the Pacific Oyster, *Crassostrea gigas*, Collected from the Pearl River Estuary, Southern China

W. Liu · P. Y. Deng

Received: 14 June 2006 / Accepted: 15 March 2007 / Published online: 10 May 2007

Oysters are marine bivalve mollusks, which feed on phytoplankton and detrital organic matter by filtering the seawater through their gills. During the filter-feeding activity, pollutants in the water may enter their body through the gills and associate with the detritus indirectly through the digestive tract. Oysters have been used as biomonitors of pollutants in coastal environments because they are very common in most coastal ecosystems, sessile, sufficiently long living, and can accumulate pollutants to high levels without affecting their survival (Presley et al., 1990; Rainbow, 1995).

With the rapid economic development and increase in population density in the Pearl River Delta region of Southern China over the past two decades, large amounts of pollutants have been discharged into the Pearl River. Oysters have been used in monitoring metal contamination in this region since the late 1970s (Phillips, 1982, 1989; Wong et al., 1982; Ashton et al., 1991; Cheung et al., 1992). However, most of these studies were conducted with oysters sampled in Hong Kong. There are only few reports of metal levels in oysters from other areas of the Pearl River Delta. Moreover, studies previously carried out in Hong Kong were focused on food safety, very often using samples purchased from the wet market. Results of these studies indicated that there was an increasing trend of heavy metal contamination in oyster tissues over the past two decades. There is a lack of information in other oyster populations along the Pearl River estuary to allow a comparison of heavy metal contamination levels among different oyster populations.

In this study we sampled the Pacific oyster *Crassostrea gigas* from the three main oyster culture areas along the Pearl River estuary and analyzed the levels of cadmium (Cd), copper (Cu), lead (Pb) and zinc (Zn) in their tissue.

Materials and Methods

The Pearl River estuary (Fig. 1) lies in southern China. The mean annual temperature is 22.8°C and the mean annual rainfall is 2,214 mm. Summer is the wet season, with 77% of the total annual rainfall falling between May and September. The annual discharge into the estuary is about 11,100 m^3 (Wong et al., 1995). The estuary is bell-shaped, with an area of approximately 8000 km^2. Bordering the estuary is Guangdong (which includes several big cities such as Guangzhou, Shenzhen, Zhuhai, and Dongguan), with a total population of over 10 million, and Hong Kong, with a population of 7 million.

There are extensive mudflats along the estuary, which provide the habitat for the Pacific oyster. Oyster cultivation has been practiced in this region for more than 700 years (Cheung et al., 1992). There are three traditional oyster farming centers (Fig. 1). Henqin (HQ) is situated on the western coast of the estuary, near Zhuhai. Baishizhou (BSZ) and Lau Fau Shan (LFS) are situated on the eastern coast, in Shenzhen and Hong Kong respectively. Oysters were sampled from these three sites in November 2003. At each site 30 oysters were collected directly from the mudflat. Only one oyster was collected randomly within the reach of an arm to ensure the samples were representative of the population.

W. Liu
Institute of Hydrobiology, Jinan University, Guangzhou 510632, P. R. China

P. Y. Deng (✉)
Institute of Environmental Science, South China Normal University, Guangzhou 510631, P. R. China
e-mail: dpy213@126.com

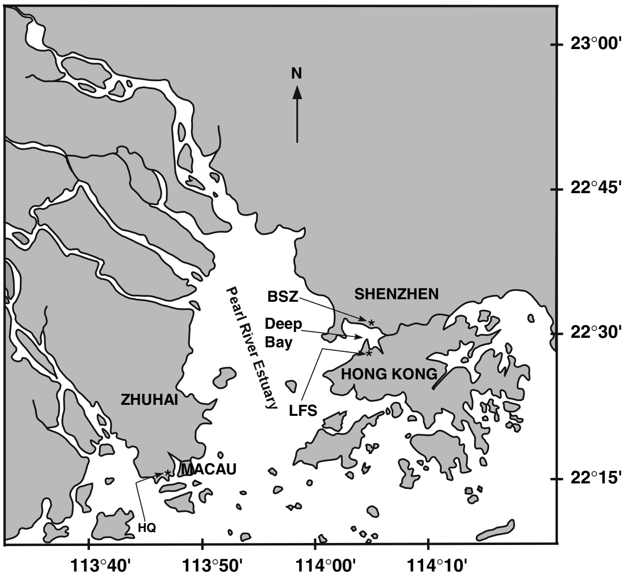

Fig. 1 Sites for collection of *Crassostrea gigas* in the Pearl River estuary, southern China

Table 1 Shell length and dry tissue weight of *Crassostrea gigas* from study sites (mean ± SD, *n* = 30 at each site). Different letters in the same column indicate significant difference at 5% level

Station	Shell length (cm)		Dry tissue weight (g)	
	Mean ± SD	Range	Mean ± SD	Range
LFS	12.27 ± 1.81a	9.1–16.5	1.04 ± 0.24b	0.69–1.72
BSZ	9.61 ± 1.41c	7.3–13.8	1.34 ± 0.31b	0.85–2.23
HQ	11.03 ± 1.3b	8.5–14	3.63 ± 1.06a	2.03–6.82

In the laboratory the oysters were scrubbed to remove mud, rinsed with water, and blotted dry. The length of each oyster was measured with a caliper, and the shells were opened with a stainless-steel screwdriver. Soft tissues in each oyster was removed with a plastic spoon, rinsed with deionized water, and then dried in an oven at 60°C until a constant weight was reached. The dried tissues were weighed and then ground. About 0.5 g of the dried tissue was digested with 5 ml of 70% nitric acid, until a clear solution was obtained. The digested solution was filtered, diluted to 25 ml with Milli-Q water, and then analyzed for Cd, Cu, Pb and Zn using an Inductively Coupled Plasma Atomic Emission Spectrophotometer (ICP-AES) (Perkin-Elmer, Plasma 40). A standard reference material (LUTS-1 from National Research Council, Canada) was also used concurrently during oyster tissue digestion and measurement. The recovery efficiency ranged from 83 to 96%. The data were not corrected for recovery. To check for contamination blanks were also analyzed using this procedure. Correlation analyses were performed for dry weight and metal concentration to determine the size effect. The data were logarithmically transformed before the correlation analyses. For a particular metal, if a significant size effect was detected for data from any of the sites, inter-site comparisons of that metal was conducted using analysis of covariance (ANCOVA), followed by multiple comparisons using the Least Significant Difference (LSD) test. In the absence of a size effect, analysis of variance (ANOVA), followed by the LSD test, was carried out to compare the metal levels in oysters from different sites.

Table 2 Correlation analysis results of oyster heavy metal concentrations and dry weight in samples collected from three sites ($P = 0.05$). * Indicates significant correlation at 5% level

	Cd	Cu	Zn	Pb
LFS	–0.384*	—	—	—
BSZ	—	—	—	–0.551*
HQ	–0.624*	—	—	–0.467*

Table 3 Mean metal concentrations (μg·g^{-1}, dry weight) in *Crassostrea gigas* tissues from the three study sites (mean ± SD, *n* = 30 at each site). Different letters in the same column indicate significant difference at 5% level

	Cd	Cu	Pb	Zn
LFS	12 ± 4b	1411 ± 365a	3 ± 1b	3627 ± 834a
BSZ	25 ± 7a	889 ± 257b	8 ± 2b	2643 ± 661b
HQ	11 ± 3b	389 ± 200c	37 ± 15a	1272 ± 466c

Results

The shell length differed significantly among the sites, ranging from 9.6 cm at BSZ to 12.3 cm at LFS (Table 1). The dry weight also differed significantly among the sites, ranging from 1.0 g at LFS to 3.6 g at HQ. Significant negative correlation between oyster weight and metal concentration was detected for Cd in samples from LFS and HQ, and Pb in samples from BSZ and HQ (Table 2). Therefore, an among-site comparison of Cd and Pb concentrations in the samples was conducted using ANCOVA, whereas the among-site comparison of Cu and Zn was conducted using ANOVA.

Among the three sites, concentrations of Cd, Cu, Pb and Zn ranged from 11 to 25 μg·g^{-1}, 389 to 1411 μg·g^{-1}, 3 to 37 μg·g^{-1}, and 1272 to 3627 μg·g^{-1}, respectively (Table 3). For each metal the concentration differed significantly among the sites, but there was no consistent trend for the concentration of the four metals among the sites. For example, in samples from HQ, Cd, Cu and Zn concentrations were the lowest, but the Pb concentration was the highest among the three sites. In samples from LFS, Cu

Table 4 Correlation coefficients between different metals in the oyster tissues. * and ** indicates significant correlation at < 0.05 level and < 0.01 level, respectively

	Cd-Cu	Cd-Zn	Cd-Pb	Cu-Zn	Cu-Pb	Zn-Pb
LFS	–0.425*	–0.2273	0.4*	0.863**	0.101	0.227
BSZ	0.191	0.468*	0.067	0.725**	0.088	0.412*
HQ	0.283	0.425*	0.285	0.938**	-0.241	–0.120

and Zn concentrations were the highest, but Cd and Pb concentrations were relatively low. For all three sites, there was significant positive correlation between Cu and Zn concentrations (Table 4). However, the correlation between other metals was not consistent for the three sites. Overall, it seems to be evident that the metal profiles in oysters from LFS and BSZ were quite similar, but they differed quite dramatically from that in the oysters from HQ.

Discussion

Given that the accumulated concentrations are integrated measures of the amounts of bioavailability of each metal to the oyster over time, the bioavailability of the four metals varies geographically in the Pearl River estuary. Li et al. (2001) noted that Pb concentration in the western side of the estuary was higher than that in the eastern side of the estuary. Li et al. (2001) further found that a higher percentage of Pb in the western area of the estuary was associated with the Fe-Mn fraction, as compared to a higher percentage of the metal associating with the residual fraction in the eastern side. These authors proposed that the different chemical forms of Pb could influence the solubility and mobility of the metal from sediments in different parts of the estuary. Our finding of higher Pb concentration in the oyster tissues from the HQ site on the western side might reflect the high bioavailable Pb in the environment. Similarly, the higher Cd, Cu and Zn concentrations in the oysters collected from the BSZ and LFS sites might reflect the higher bioavailability of these three metals in the eastern waters of the estuary.

Caution should be taken when comparing the tissue metal burden reported in different studies. The oysters in Philips (1982) and Cheung et al. (1992) came from LFS wet markets, whose native culture sites could not be positively identified because LFS wet market not only serves as a local distribution center for oysters and other live seafood; some of the oysters might have come from the Shenzhen. Ashton (1991) put mudflat-collected oysters in cages and suspended them in Deep Bay for a certain period of time before collecting them for metal analyses. The oysters used in these studies (over 15 cm long) were generally larger than those used in this study (under 15 cm long). Small oysters have generally been reported to have higher metal concentrations than large ones (Shulkin et al., 2003; Phillips, 1982; Cheung and Wong, 1992). A valid comparison of the present data for metal concentrations in *Crassostrea gigas* tissues with those in previous reports should thus be limited to the oysters collected directly from the intertidal mudflats (i.e. Wong et al., 1982; Phillips et al., 1982, 1989). The comparison shows that, except Zn contents, the maximal metal concentrations (Cd, Cu and Pb) are high, indicating that the bioavailability of these metals to oysters may have increased in the past two decades. Cheung et al. (1992) proposed that the rapid industrialization and urbanization of Shen Zhen and Yuen Long might have resulted in an increase of heavy metal discharge into the water and subsequently led to high metal concentrations in oyster tissues in the Deep Bay area.

Table 5 Comparison of soft tissue trace metal concentrations (range or means ± S.D., $\mu g \cdot g^{-1}$, dry weight) in *Crassostrea gigas* from the Pearl River estuary

Location	Cd	Cu	Pb	Zn	Reference
LFS mudflat, Hong Kong	12 ± 4	1411 ± 365	3 ± 1	3627 ± 834	This study
BSZ mudflat, Shenzhen	25 ± 7	889 ± 257	8 ± 2	2643 ± 661	This study
HQ mudflat, Zhuhai	11 ± 3	11 ± 3	37 ± 15	1272 ± 466	This study
A few locations along the coast of Guangdong province	2.35 ~ 29.4	65.5 ~ 2275		350 ~ 4654	Lu (1996)
LFS Market, Hong Kong	0.6 ~ 5.4	110 ~ 309	0.1 ~ 0.7	269 ~ 662	Phillips (1982a)
LFS Market, Hong Kong	7.44 ~ 10.98	589 ~ 1071			Cheung & Wong (1992)
Deep Bay water, Hong Kong	~1.0	375 ~ 700	~0.6	1.6 ~ 2250	Ashton et al. (1991)
Deep Bay mudflat, Hong Kong	3.5 ~ 4.1	192 ~ 744	8.2 ~ 14.3	780 ~ 1046	Wong et al. (1982)
Deep Bay mudflat, Hong Kong	0.23 ~ 5.88	12 ~ 495	0.01 ~ 1.0		Phillips et al. (1989)
Deep Bay mudflat, Hong Kong	0.42 ~ 12	44 ~ 254	0.1 ~ 0.4	240 ~ 795	Phillips et al. (1982b)

Note: * wet weight

Table 6 Median and 85th percentile of metal concentrations in different Oyster-watch programmes (μg·g^{-1}, dry weight). WMW - World Mussel Watch, NS&T - National Status and Trends, United States, RNO - Reseau National d'Observation de la Qualite du Milieu Marin, France. These data were obtained from Cantilo (1998)

	WMW		NS&T		RNO		This study
	Median	85th perc.	Median	85th perc.	Median	85th perc	Range
Cd	2.6	12.0	2.6	5.5	1.4	3.7	11–25
Cu	160	680	120	280	130	320	389–1411
Pb	2.5	8.6	0.5	0.9	1.4	2.4	3–37
Zn	1600	4500	2100	4300	2100	3500	1272–3627

To determine if heavy metal concentrations in local oysters were unusually high, data were compared to the long term data (means and 85th percentiles) for oysters from the World Mussel Watch (WMW), the National Status and Trends, United States (NS&T), and the Reseau National d'Observation de la Qualite du Milieu Marin (RNO) (Table 6). The 85th percentiles can be considered to represent high concentrations in these Oyster-watch programmes (Cantilo, 1998). Mean Cd concentration in oysters from the three sites in the present study was similar to or even higher than the 85th percentiles from WMW. This was also true for Cu in oysters from LFS and HQ, and for Pb for oysters from BSZ and HQ. Mean Zn concentrations in the present study are relatively low, but some individuals contained Zn concentrations reaching the median Zn level in WMW. Pearl River estuary thus shows high bioavailable metals, when considered on a global scale.

The Pacific Oyster (*Crassostrea gigas*) is an important seafood and thus has great economic value at the Pearl River Delta area. However, oysters from the three natural distribution centers all contained heavy metals at alarming levels. Our present study indicates that the metal burden profiles differ between oyster populations inhabiting the western and eastern sides of the estuary, and such metal profiles might be related to the metal bioavailability in local areas. Furthermore, for the Hong Kong oyster population, there seems to be an increasing trend of metal contamination in the past two decades, probably reflecting the increasing industrial activities in the Shenzhen and the surrounding areas. Therefore, legislation enforcement is needed to regulate the discharge of metal containing industrial effluents into the local environment, and further monitoring work is needed to check whether such increasing trend of metal contamination will persist in the future.

Acknowledgments The authors would like to thank Dr. Qiu and Dr. Gu Jidong for helpful comments on the manuscript. This study was supported by the Guangdong Province Natural Science Fund (No. 5300407).

References

Ashton A (1991) ''Oyster-watch'' for monitoring coal ash lagoons in an environmentally sensitive area of Hong Kong. Mar Pollut Bull 22(7):334–339

Cantillo AY (1998) Comparison of results of Mussel Watch Programs of the United States and France with Worldwide Mussel Watch Studies. Mar Pollut Bull 36(9):712–717

Cheung YH, Wong MH (1992) Trace metal contents of the Pacific oyster (*Crassostrea gigas*) purchased from markets in Hong Kong. Environ Manage 16:753–761

Lu CH (1996) Bioavailable heavy metals (cadmium, copper and zinc) in the coastal waters of Guangdong Province. Mar Environ Science 2:58–63 (in Chinese)

Li X, Shen Z, Wai OWH, Li YS (2001) Chemical forms of Pb, Zn and Cu in the sediment profiles of the Pearl River estuary. Mar Pollut Bull 42: 215–223

Phillips DJH (1982a) Trace elements of toxicological significance to man in Hong Kong seafood. Environ Pollut 3:27–45

Phillips DJH, Ho CT, Ng LH (1982b) Trace elements in the Pacific oyster in Hong Kong. Arch Environ Con Tox 11: 533–537

Phillips DJH (1989) Trace elements in the Pacific oyster in Hong Kong. Mar Pollut Bull 20:319–327

Presley BJ, Taylor RJ, Boothe PN (1990) Trace metals in Gulf of Mexico oysters. Sci Total Environ 97:551–593

Rainbow PS (1995) Biomonitoring of heavy metal availability in the marine environment. Mar Pollut Bull 6(12):180–187

Shulkin VM, Presley BJ, Kavum VI (2003) Metal concentrations in mussel *Crenomytilus grayanus* and oyster *Crassostrea gigas* in relation to contamination of ambient sediments. Environ Int 29: 493–502

Wong CK, Chu KH, Chen QC, Ma XL (1995) Environmental Research in Pearl River and coastal areas. Gongdong Higher Education Press, Guangzhou, China

Wong MH, Choy CK, Lau WM, Cheung YH (1982) Heavy-metal contamination of the Pacific Oyster (*Crassostrea gigas*) cultured in Deep Bay, Hong Kong. Environ Res 25: 302–309