Biol Trace Elem Res
DOI 10.1007/s12011-012-9412-y

Heavy Metal Concentrations in Surface Sediments and Manila Clams (*Ruditapes philippinarum*) from the Dalian Coast, China after the Dalian Port Oil Spill

Liqiang Zhao · Feng Yang · Xiwu Yan ·
Zhongming Huo · Guofan Zhang

Received: 12 February 2012 / Accepted: 29 March 2012

Abstract We conducted an investigation of heavy metal concentrations in Manila clams (*Ruditapes philippinarum*) and surface sediments after the Dalian Port oil spill. Samples were collected from three mariculture zones (Jinshitan, Dalijia, and Pikou) along the Dalian coast. Heavy metal concentrations in *R. philippinarum* were consistent and ranked in decreasing order of Zn > Cu > As > Cr > Pb > Cd > Hg, while concentrations in surface sediments were ranked as Zn > Cr > Cu > Pb > As > Cd > Hg, respectively. Bioaccumulation of Zn, Cd, and Hg had obviously occurred in *R. philippinarum*. Statistically significant correlations ($p < 0.05$) between concentrations of Pb, Cd, and Hg in *R. philippinarum* and in surface sediments were observed. Except for Cr and As, heavy metal concentrations in *R. philippinarum* were well within the legal limits for human consumption.

Keywords Heavy metal · Bioaccumulation · Surface sediments · *Ruditapes philippinarum*

Introduction

On 16 July 2010, a large amount of crude oil spilled into the water after a pipeline exploded at Dalian Port, Liaoning Province, China. The explosion at an oil storage depot hit the oil pipeline when a tanker ship was unloading. The first explosion triggered a second blast from a smaller adjacent pipeline, which released more crude oil into the sea. A large oil slick formed, creating a dark brown belt of crude oil and other pollution that stretched at least 50 km^2 off the port. After the oil spill, combinations of biological, physical, and chemical methods were used to contain and clean up the oil slick. Strategies to monitor and evaluate environmental changes along the Dalian coast were subsequently adopted to assess environmental damage and long-term effects.

Huge oil spills, such as the Gulf War oil spill [1] and the *Exxon Valdez* oil spill [2], had devastating effects on the marine environment, as crude oil is full of toxic materials. Although the composition of crude oil is complex, heavy metals are an integral component of it [3, 4]. Heavy metals are persistently toxic, prone to bioaccumulation, and pose a risk to humans [5, 6]. They reflect the current quality of the marine environment and also can be indicators of long-term problems because they tend to accumulate in the food chain and ultimately are consumed by organisms, including humans [7, 8].

Due to their wide distribution from tropical to high latitudes, sedentary lifestyle, ability to accumulate heavy metals, and ease of collection [9], bivalves are widely used as indicators of heavy metal contamination [10–14]. As filter feeders, bivalves accumulate heavy metals from the water, sediments, and particulate matter in their soft tissues [15, 16], and a significant relationship has been found between heavy metal concentration in surface sediments and in soft tissues of bivalves [17–19]. Therefore, tracing heavy metal accumulation in bivalves should be part of the monitoring and evaluation programs conducted after the Dalian Port oil spill.

Manila clam (*Ruditapes philippinarum*; Adams et Reeve 1850), which belongs to Mollusca, Lamellibranchiata, Heterodonta, Veneroida, Veneridae, has been cultured for more

L. Zhao · F. Yang · X. Yan (✉) · Z. Huo
College of Fisheries and Life Science, Dalian Ocean University,
Dalian 116023, People's Republic of China
e-mail: zlq830418@163.com

G. Zhang
Institute of Oceanology, Chinese Academy of Science,
Qingdao 266071, People's Republic of China

than 50 years in China. The production of Manila clam has reached over 3.0 million tons per annum in China, which accounts for about 90 % of worldwide Manila clam production. Manila clam culture is particularly strong along the Dalian coast, where this species has become an important fishery [20].

The primary purpose of this study was to obtain quantitative information about the concentrations of heavy metal in surface sediments and *R. philippinarum* collected from the Dalian coast after the Dalian Port oil spill. The results reported here will provide the most recent and valuable information about whether the oil spill poses a threat to the culture of *R. philippinarum.* To that end, the safety of *R. philippinarum* for human consumption was also evaluated.

Materials and Methods

Study Area and Sample Collection

Figure 1 shows the three sampling sites (Jinshitan, Dalijia, and Pikou), which were located in the mariculture zone along the Dalian coast in China. Samples were collected from three clam-farming areas at each sampling site in November 2010; during November, *R. philippinarum* has no reproductive activity [21]. When the tide ebbed, clams were collected from each sampling site using plastic buckets. The 0–10-cm surface layer of sediment samples was also gathered using a plastic scoop and stored at 4°C immediately. Meanwhile, five replicates were conducted at each sampling site. To reduce individual variations in metal concentrations [22], similar-sized clams were randomly chosen after measuring the shell lengths and weights, which were 24.98–27.24 mm and 4.12–5.04 g, respectively. There were no significant differences ($p>0.05$) among the shell lengths and weights at each sampling site.

Clams

After collection, similar-sized individuals were transported immediately to the laboratory and cultured in filtered seawater for 24 h to depurate them. Subsequently, 100 individuals from each site were chosen randomly as a pooled sample for analysis. The tissues of these clams were dissected with a plastic knife and then immediately stored at −20°C. After samples were thawed at room temperature and rinsed with distilled water, they were dried at 80°C to constant weight, homogenized, and stored in desiccators for future analysis. Prior to chemical analysis, 0.5 g of dry sample was added to 10 ml of purified nitric acid and 2 ml of purified perchloric acid, and then the solution was heated until dry. Next, 2 ml of purified nitric acid and 3 ml of distilled water were added to the cooling residue and then heated to dissolve the residue into the mixture. At this point, all digested samples were cooled at room temperature and eventually diluted to 10 ml in a volumetric flask with distilled water [23].

Surface Sediments

Sediment samples were air-dried [24], and then sieved through a 63-μm nylon mesh. The fractions <63 μm were

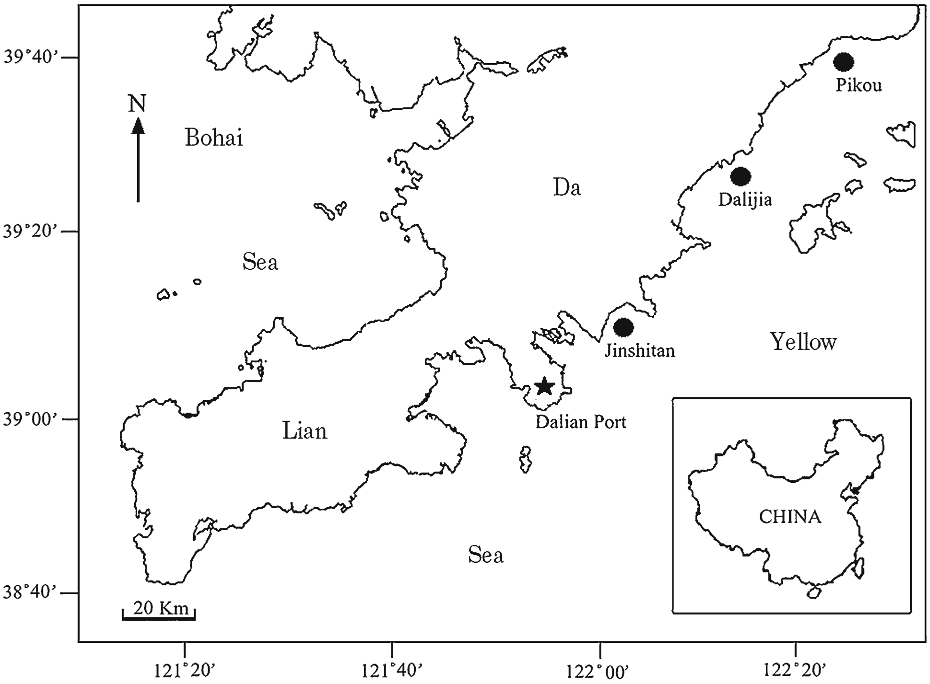

Fig. 1 Map showing the location of Dalian port (*black star*) and sampling sites (*black circles*) from the Dalian coast, China

stored in clean plastic beakers for analysis. The samples were prepared in triplicate and digested using a mixture of concentrated nitric acid, perchloric acid, and hydrofluoric acid. The digested mixture then was processed prior to chemical analysis, following the procedure described above for the clam tissues.

Chemical Analysis

Concentrations of Cu, Pb, Zn, Cd, and Cr in the sediment and clam samples were measured with a flame atomic absorption spectrophotometer using deuterium background correction. Hg and As were analyzed by the cold vapor technique using Portable Zeeman Lumex mercury analyzer. Standard reference materials (mussel tissue sample, GBW08571; coastal sediment sample, GBW07314; both from the National Research Center for Certified Reference Materials, Beijing) were used for quality assurance and quality control procedures. Satisfactory recoveries were obtained for Cu (92.3–102.6 %), Pb (97.2–108.4 %), Zn (87.6–95.1 %), Cd (96.6–101.4 %), Cr (94.2–106.3 %), Hg (88.2–97.5 %), and As (93.8–102.5 %). Metal concentrations in *R. philippinarum* and surface sediments are presented as milligrams per kilogram dry weight.

Statistical Analysis

To compare the total content of metals at different sampling sites, the metal pollution index (MPI) was used. It was obtained according to the equation [25].

$$\mathrm{MPI} = (\mathrm{Cf}_1 \times \mathrm{Cf}_2 \ldots \mathrm{Cf}_n)^{1/n}$$

where Cf_1=concentration of the first metal, Cf_2=concentration of the second metal, Cf_n=concentration of metal n, and n=the total number of studied metals in the sample.

To further evaluate the efficiency of metal bioaccumulation in *R. philippinarum*, the biosediment accumulation factor (BSAF) was calculated. The BSAF is defined as the ratio of the metal concentration in the organism to that in the sediment [22].

$$\mathrm{BSAF} = \mathrm{Cx}/\mathrm{Cs}$$

where Cx and Cs are the average concentrations of a given metal in the organism and the associated sediment, respectively.

Statistical analyses were executed by the programs Statview 5.0 for windows (SAS Institute Inc, Cary, NC, USA). A one-way analysis of variance followed by a significant difference (LSD) test was used to verify any statistically significant difference ($p<0.05$) among sampling sites. Pearson correlation and linear regression model were used to assess the correlation between heavy metal concentrations in *R. philippinarum* and in surface sediments.

Results and Discussion

Heavy Metal Concentrations in Surface Sediments

Table 1 shows the heavy metal concentrations in surface sediments collected from the Dalian coast. All trace metal concentrations in surface sediments collected from Jinshitan, Dalijia, and Pikou, in general, were ranked in decreasing order, as follows: Zn > Cr > Cu > Pb > As > Cd > Hg. Concentrations of Cu, Pb, Zn, Cd, and Cr in surface sediments collected at Pikou were significantly higher than these in Jinshitan ($p<0.05$), while there were no significant differences in Hg and As levels among these sampling sites ($p>0.05$). This is not surprising, considering that Pikou is an important port town, and its waters are subjected to contamination by persistent organic pollutants, municipal waste disposal, and metals from nautical, tourist, and agricultural industries. In contrast, the lowest concentrations of the metals mentioned above were found at Jinshitan. Jinshitan is a famous national tourist attraction without industries, and it is considered to be relatively free of anthropogenic contamination. Although the Dalian Port oil spill have occurred closer to Jinshitan than Pikou, the lowest concentrations of Cu, Pb, Zn, Cd, and Cr collected at Jinshitan should be considering the background levels before oil spill.

However, oil spill investigations usually lack pre-impact data, which precludes assessment of the effect of the oil spill on the marine environment [26]. This was the case for the Dalian Port oil spill. Because pre-impact data were unavailable for the Dalian coast, the data collected herein were compared with metal levels from other geographic areas to infer whether heavy metal concentrations were altered at the three sites due to the oil spill. Metal levels in surface sediments from the Dalian coast were similar to those from other locales reported by other researchers (Table 2). Levels of Cu, Pb, Zn, and Cr were in agreement with those reported for Jiaozhou Bay, which is another important clam-farming area in China [23]. Hg and As, which were widespread and in high concentrations in surface sediments from Venice Lagoon in Italy [27] and southern Atlantic in Spain [19], were found in lower concentrations in the present study.

Heavy Metal Concentrations in *R. philippinarum*

Table 1 also shows the heavy metal concentrations found in *R. philippinarum*. Trace metal concentrations in *R. philippinarum* collected from Jinshitan, Dalijia, and Pikou were consistent and ranked, in decreasing order of concentration, as follows: Zn > Cu > As > Cr > Pb > Cd > Hg, and similar trends were reported previously for *R. philippinarum* from other geographical areas (Table 2). Compared to the other metals analyzed, Zn and Cu were found in much higher concentrations [31, 32]. In contrast, Hg was found in the lowest concentration. Usero et al. [25] reported a similar concentration of Hg in *R. philippinarum* from

Table 1 Heavy metal concentrations (in milligrams per kilogram dry weight) in surface sediments and *R. philippinarum* collected from the Dalian coast

Sampling sites	Trace metals						
	Cu	Pb	Zn	Cd	Cr	Hg	As
Surface sediment							
Jinshitan	11.07±2.62b	7.05±1.75c	37.2±6.09b	0.06±0.006b	15.58±5.34bc	0.007±0.002a	3.86±1.04a
Dalijia	15.67±2.31b	11.01±0.78b	51.6±5.69a	0.11±0.03a	33.64±3.34ab	0.009±0.002a	4.03±1.19a
Pikou	22.55±3.54a	17.95±2.42a	59.0±3.15a	0.12±0.03a	44.67±9.92a	0.008±0.002a	3.91±0.93a
R. philippinarum							
Jinshitan	6.07±2.71a	2.19±0.72a	146.0±13.6a	0.62±0.04a	2.35±1.48a	0.054±0.012a	2.60±1.86b
Dalijia	5.85±2.51a	0.85±0.17b	54.3±11.73b	0.74±0.21a	2.25±1.18a	0.045±0.008a	2.45±2.17b
Pikou	6.33±2.02a	0.90±0.39b	115.3±36.7c	0.59±0.10a	1.57±0.46b	0.051±0.010a	4.10±2.51a

Data are mean±SE ($n=5$). For each metal, different letters indicate significant difference among the three sampling sites ($p<0.05$)

southern Atlantic coast in Spain. Except for Cu and Hg, the concentrations of other metals in *R. philippinarum* varied notably depending on the location of the sampling sites.

The concentrations of Pb and Zn in *R. philippinarum* collected at Jinshitan were significantly higher than these in Dalijia and Pikou ($p<0.05$), whereas As was the prominent metal in Pikou. There were no significant differences in Cu, Cd, and Hg levels among these sites ($p>0.05$). Many studies have reported that, for benthic bivalves, there was a significant correlation between metal concentrations in their soft tissues and surface sediments where they live [17–19]. Considering the lowest metal concentrations in surface sediments at Jinshitan, however, the higher metal concentrations in *R. philippinarum* collected from Jinshitan may be polluted from the oil spill. Meanwhile, the MPI was used to compare the total content of metals at those sampling sites. As shown in Table 3, the MPI value at Jinshitan was somewhat higher than other sites. This tendency was especially marked in the case of Pb. However, these MPI values are within the ranges reported in other studies [19, 25, 31].

Metal levels in *R. philippinarum* in this study were compared with data from previous studies conducted in China and

Table 2 Comparison heavy metal concentrations (in milligrams per kilogram dry weight) in surface sediments and *R. philippinarum* obtained in the present study with literature data from China and other regions all over the world

Region	Trace metals						
	Cu	Pb	Zn	Cd	Cr	Hg	As
Surface sediment							
This study	11.07–22.55	7.05–17.95	37.2–59.0	0.06–0.12	15.58–44.67	0.007–0.009	3.86–4.03
Jiaozhou Bay [23]	5.31–45.07	4.34–35.01	32.1–151.7	0.76–2.37	15.88–54.0	n.a.	n.a.
Southern Atlantic [19]	6–92	2–46	18–460	0.26–0.72	10–33	0.11–0.41	3.5–102
Venice Lagoon [27]	2.73–42.94	4.99–47.28	4.10–319	0.10–1.43	32.2–86.19	0.13–1.22	4.0–17.1
Gulf of Aden [9]	3.1–270	5.3–23.0	13.0–51.0	0.3–2.6	5.7–25.3	n.a.	n.a.
Southern Black Sea [28]	15–119	12–69	21–141	n.a.	13–238	n.a.	n.a.
Naples Harbour [29]	12–5,743	19–3,083	17–7,234	0.01–3.00	7–1,798	n.a.	n.a.
Mediterranean Sea [30]	14.1–44.6	n.a.	60.7–305.5	1.5–8.1	n.a.	n.a.	n.a.
R. philippinarum							
This study	5.85–6.33	0.85–2.19	54.3–146.0	0.59–0.74	1.57–2.35	0.045–0.054	2.45–4.10
Jiaozhou Bay [23]	5.07–26.03	0.89–14.77	52–110	0.65–3.31	9.64–35.47	n.a.	n.a.
Bohai Sea [31]	7.53–25.71	0.47–2.00	59–190	0.82–5.71	0.94–19.06	n.a.	n.a.
Southern Atlantic [25]	8.24–16.47	0.41–1.94	76–124	0.47–2.71	0.65–3.410	0.02–0.467	2.17–5.54
Korea estuary [32]	5.47–14.30	0.34–1.72	64.7–162.0	0.53–2.20	0.61–2.38	n.a.	na
Venice Lagoon [27]	8.18–29	0.49–2.54	60.0–122	0.26–2.19	1.89–5.70	0.19–1.78	18.9–45.8
Gironde estuary [33]	9.1±1.0	n.a.	97.2±8.0	0.52±0.11	n.a.	n.a.	n.a.

n.a. no available data

Table 3 Mean BSAF and the metal pollution index (MPI) values in *R. philippinarum* collected from the Dalian coast

Sampling sites	Trace metals							MPI
	Cu	Pb	Zn	Cd	Cr	Hg	As	
Jinshitan	0.55	0.31	3.93	10.33	0.15	7.71	0.67	2.33
Dalijia	0.37	0.08	1.05	6.73	0.07	5.00	0.61	1.62
Pikou	0.28	0.05	1.96	4.92	0.04	6.38	1.05	1.96

elsewhere in the world. Except for Zn, which had the maximum concentration of 216 mg kg^{-1} dry weight, concentrations of Cu, Pb, Cd, Cr, Hg, and As were not obviously higher than the normal range, which might be considered to be background levels (Table 2). All of these results suggest that clams inhabiting sediments along the Dalian coast are relatively unpolluted with heavy metals following the Dalian Port oil spill.

Bioaccumulation of Heavy Metals

To evaluate the efficiency of metal bioaccumulation in *R. philippinarum*, the BSAF values were calculated. Bioaccumulation is expected to occur in organisms if the BSAF is >1 [34]. Table 3 shows that the metals with BSAF values <1 were Cu, Pb, Cr, and As, indicating almost no bioaccumulation of these metals in *R. philippinarum*. In contrast, Zn, Cd, and Hg were the metals with the BSAF values >1, especially Cd showed the BSAF value as high as 10.33, which suggested that bioaccumulation had obviously occurred in *R. philippinarum* from the Dalian coast. Finally, if we compare these sampling sites, we can find that *R. philippinarum* collected from Jinshitan had the highest BSAF values of Cu, Pb, Zn, Cd, Cr, and Hg, while *R. philippinarum* collected from Pikou had the highest BSAF value of As.

Relationship Between Heavy Metals in *R. philippinarum* and Surface Sediments

The relationships between heavy in *R. philippinarum* and surface sediments are presented in Table 4. Pearson correlation analysis clearly indicated a very strong relationship ($p<0.01$) between Hg concentration in *R. philippinarum* and surface sediments, as well as a strong correlation ($p<0.05$) between Pb and Cd in *R. philippinarum* and surface sediments. The relationship between Cu, Zn, Cr, and As in *R. philippinarum* and surface sediments was not significant ($p>0.05$), indicating that the concentrations of these metals

Table 4 Linear regression equations between heavy metal concentrations in *R. philippinarum* and in surface sediments collected from the Dalian coast

	Slop	Intercept	R^2	*p* value
Cu	0.163	3.397	0.130	>0.05
Pb	0.082	2.189	0.052	<0.05
Zn	0.280	168.262	1.521	>0.05
Cd	0.054	0.441	1.119	<0.05
Cr	0.003	1.951	0.321	>0.05
Hg	2.390	0.054	1.574	<0.01
As	0.799	−4.026	0.498	>0.05

Table 5 Comparison heavy metal concentrations (in milligrams per kilogram wet weight) in *R. philippinarum* obtained in the present study with the maximum permissible concentrations for bivalves in legislation from China and other geographic areas all over the world

	Cu	Pb	Zn	Cd	Cr	Hg	As
This study	4.7	0.9	93.7	0.5	1.6	0.04	2.6
	10[a]	0.1	20	0.2	n.a.	0.05	1.0
GB 18421-2001[b]	25[c]	2.0	50	2.0	n.a.	0.1	5.0
	50[d]	6.0	100	5.0	n.a.	0.3	8.0
GB 2762-2005 [e]	n.a.	n.a.	n.a.	n.a.	2.0	0.5	0.5
NY 5073-2006[f]	50	1.0	n.a.	1.0	n.a.	n.a.	n.a.
Hong Kong[g]	n.a.	6.0	n.a.	2.0	1.0	0.5	10.0
Australia[h]	70	2.0	1,000	2.0	n.a.	0.5	1.0
Europe[i]	n.a.	1.5	n.a.	1.0	n.a.	0.5	n.a.

Data based on dry weight in *R. philippinarum* are converted to wet weight basis for comparison using a wet weight/dry weight ratio of 5

n.a. no available data

[a] First limits

[b] "Marine Biological Quality," promulgated by the State General Administration of the People's Republic of China for Quality Supervision and Inspection and Quarantine: first limits, second limits, and third limits

[c] Second limits

[d] Third limits

[e] "Maximum levels of Contaminants in Foods," promulgated by the Ministry of Health, China

[f] "Residue Limit of Toxic Substances in Nuisanceless Foods and Aquatic Products," promulgated by the Ministry of Agriculture, China

[g] "Public Health and Municipal Services Ordinance," promulgated by the Government of the Hong Kong Special Administrative Region of the People's Republic of China

[h] "Food Standards Code," promulgated by the Australia New Zealand Food Authority

[i] "Commission regulation," No 466/2001of 8 March 2001, setting maximum levels for certain contaminants in foodstuffs, from the Official Journal of the European Communities L77

in surface sediments were not directly reflected in *R. philippinarum*. When the concentrations of metals in surface sediments are lower than the threshold below which the organisms are able to accumulate the metals in their bodies, bioaccumulation is not significant [19, 30]. Moreover, bioaccumulation is probably influenced by many physicochemical and biological factors [5].

Linear regressions for the concentrations of Pb, Cd, and Hg in *R. philippinarum* (y values) relative to their concentrations in surface sediments (x values) were calculated. Based on the value of their slopes, the abundance of Pb, Cd, and Hg can be ordered as follows: Hg (2.390)>Pb (0.082)>Cd (0.054). The slope for Hg was significantly >1, which suggests that the bioavailability of Hg is disproportionately increased with the degree of its enrichment in surface sediments.

Human Consumption

In the present study, metal levels were converted to values per wet weight and compared with legal limits to evaluate the safety of *R. philippinarum* for human consumption (Table 5). According to the Marine Biological Quality in China, Cu and Hg were well below the first quality standard limits. Although Pb, Zn, Cd, and As exceeded the first quality standard limits, they were well within the second quality standard limits. However, As significantly exceeded the Maximum Levels of Contaminants in Foods in China. Having compared metal levels with legal limits in Hong Kong, Australia, and Europe, we found that Cu, Pb, Zn, Cd, and Hg were well within the maximum permissible limits. However, Cr and As exceeded the maximum permissible levels in Hong Kong and Australia, respectively. Therefore, we can conclude that all of these metals in *R. philippinarum* measured in this study, with the exception of Cr and As, were within the range suitable for human consumption.

Acknowledgments We thank the anonymous reviewers for their helpful comments on this work. This project was supported financially by the China Agriculture Research System (CARS–48).

References

1. Jones DA, Plaza J, Watt I, Sanei M (1998) Long-term (1991–1995) monitoring of the intertidal biota of Saudi Arabia after the 1991 Gulf War oil spill. Mar Pollut Bull 36:472–489
2. Peterson CH, Rice SD, Short JW, Esler D, Bodkin JL, Ballachey BE (2003) Long-term ecosystem response to the Exxon Valdez oil spill. Science 302:2082–2086
3. Tariq WK, Kureishy TW (1993) Concentration of heavy metals in marine organisms around Qatar before and after the Gulf War oil spill. Mar Pollut Bull 27:183–186
4. Hassan M, Al S (1996) The determination of lead, nickel and vanadium in Saudi Arabian crude oil by sequential injection analysis/inductively-coupled plasma mass spectrometry. Talanta 43:1313–1319
5. Rainbow PS (1997) Trace metal accumulation in marine invertebrates: marine biology or marine chemistry? J Mar Biol Assoc UK 77:195–210
6. Jureša D, Blanuša M (2003) Mercury, arsenic, lead and cadmium in fish and shellfish from the Adriatic Sea. Food Addit Contam 20:241–256
7. Abbott P, Baines J, Fox P, Graf L, Kelly L, Stanley G, Tomaska L (2003) Review of the regulations for contaminants and natural toxicants. Food Control 14:383–389
8. Franca S, Vinagre C, Cacador I, Cabral HN (2005) Heavy metal concentrations in sediment, benthic invertebrates and fish in three salt marsh areas subjected to different pollution loads in the tagus estuary (Portugal). Mar Pollut Bull 50:998–1003
9. Mostafa AR, Al-Alimi AK, Barakat AO (2009) Metals in surface sediments and marine bivalves of the Hadhramout coastal area, Gulf of Aden, Yemen. Mar Pollut Bull 58:308–311
10. Usero J, Gonzalez-Regalado E, Gracia I (1996) Trace metals in the bivalve mollusc *Chamelea gallina* from the Atlantic coast of southern Spain. Mar Pollut Bull 32:305–310
11. Shulkin VM, Presley BJ, Kavun VI (2003) Metal concentrations in mussel *Crenomytilus grayanus* and oyster *Crassostrea gigas* in relation to contamination of ambient sediments. Environ Int 29:493–502
12. Harmed MA, Emara AM (2006) Marine molluscs as biomonitors for heavy metal levels in the Gulf of Suez, Red Sea. J Mar Sys 60:220–234
13. Li Y, Yu ZM, Song XX, Mu QL (2006) Trace metal concentrations in suspended particles, sediments and clams (*Ruditapes philippinarum*) from Jiaozhou Bay of China. Environ Monit Assess 121:491–501
14. Saha M, Sarkar SK, Bhattacharya B (2006) Interspecific variation in heavy metal body concentrations in biota of Sunderban mangrove wetland, northeast India. Environ Int 32:203–207
15. Baraj B, Niencheski LF, Corradi C (2003) Trace metal content trend of mussel *Perna perna* (Linnaeus, 1758) from the Atlantic Coast of Southern Brazil. Water Air Soil Pollut 145:205–214
16. Burger J, Gochfeld M (2006) Locational differences in heavy metals and metalloids in Pacific Blue Mussels *Mytilus* [*edulis*] *trossulus* from Adak Islan, in the Aleutian Chain, Alaska. Sci Total Environ 368:937–950
17. Perez UJ, Jimenez B, Delgado W, Rodriguez-Sierra CJ (2001) Heavy metals in the false mussel, *Mytilopsis domingensis*, from two tropical estuarine lagoons. Bull Environ Contam Toxicol 66:206–213
18. Yap CK, Ismail A, Tan SG, Omar H (2002) Correlations between speciation of Cd, Cu, Pb and Zn in sediment and their concentrations in total soft tissue of green-lipped mussel *Perna viridis* from the west coast of Peninsular Malaysia. Environ Int 28:117–126
19. Usero J, Morillo J, Gracia I (2005) Heavy metal concentrations in molluscs from the Atlantic coast of southern Spain. Chemosphere 59:1175–1181
20. Zhang G, Yan X, Yang F (2006) A new three-phase culture method for Manila clam, *Ruditapes philippinarum*, farming in northern China. Aquaculture 258:452–461
21. Yan XW, Zhang GF, Yang F (2006) Effects of diet, stocking density, and environmental factors on growth, survival, and metamorphosis of Manila clam *Ruditapes philippinarum* larvae. Aquaculture 253:350–358
22. Szefer P, Geldon J, Ali AA, Bawazir A, Sad M (1997) Distribution and association of trace metals in soft tissue and byssus of mollusc *Perna perna* from the Gulf of Aden, Yemen. Environ Int 23:53–61

23. Wang XY, Zhou Y, Yang HS, Wang Q, Liu SL (2010) Investigation of heavy metals in sediments and Manila clams *Ruditapes philippinarum* from Jiaozhou Bay, China. Environ Monit Assess 170:631–643
24. Thomas RP, Ure AM, Davidson CM, Littlejohn D, Rauret G, Rubio R (1994) Three-stage sequential extraction procedure for the determination of metals in river sediments. Anal Chim Acta 286:423–429
25. Usero J, Gonzalez-Regalado E, Gracia I (1997) Trace metals in the bivalve molluscs *Ruditapes decussatus* and *Ruditapes philippinarum* from the Atlantic Coast of Southern Spain. Environ Int 23:291–298
26. Jackson JBC, Cubit JD, Keller BD, Batista V, Burns K, Caffey HM (1989) Ecological effects of a major oil spill on Panamanian coastal marine communities. Science 243:37–44
27. Sfriso A, Argese E, Bettiol C, Facca C (2008) *Tapes philippinarum* seed exposure to metals in polluted areas of the Venice lagoon. Estuar Coast Shelf Sci 79:581–590
28. Yücesoy F, Ergin M (1992) Heavy metal geochemistry of surface sediments from the southern Black Sea shelf and upper slope. Chem Geol 99:265–287
29. Sprovieri M, Feo ML, Prevedello L, Manta DS, Sammartino S, Tamburrino S (2007) Heavy metals, polycyclic aromatic hydrocarbons and polychlorinated biphenyls in surface sediments of the Naples harbour (southern Italy). Chemosphere 67:998–1009
30. Maha AM, Abdallah A, Abdallah AA (2008) Biomonitoring study of heavy metals in biota and sediments in the South Eastern coast of Mediterranean sea, Egypt. Environ Monit Assess 146:139–145
31. Wang YW, Liang LN, Shi JB, Jiang GB (2005) Study on the contamination of heavy metals and their correlations in mollusks collected from coastal sites along the Chinese Bohai Sea. Environ Int 31:1103–1113
32. Ji JY, Choi HJ, Ahn IY (2006) Evaluation of Manila clam *Ruditapes philippinarum* as a sentinel species for metal pollution monitoring in estuarine tidal flats of Korea: effects of size, sex, and spawning on baseline accumulation. Mar Pollut Bul 52:447–453
33. Baudrimont M, Schäfer J, Marie V, Maury-Brachet R, Bossy C, Boudou A, Blanc G (2005) Geochemical survey and metal bioaccumulation of three bivalve species (*Crassostrea gigas*, *Cerastoderma edule* and *Ruditapes philippinarum*) in the Nord Médoc salt marshes (Gironde estuary, France). Sci Total Environ 337:265–280
34. Liu HB, Yang J, Gan JL (2010) Trace element accumulation in bivalve mussels *Anodonta woodiana* from Taihu Lake, China. Arch Environ Contam Toxicol 59:593–601