

Levels of heavy metals in seawater, sediment and in the tissue of *Crassostrea belcheri* in the western estuary of Ganh Rai bay

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Abstract

Recent studies have shown that the seawater, sediment and commercial bivalve molluscs in the studied area have been contaminated by some heavy metals Zn, Cu, Pb and Cr. Highly toxic heavy metals like As, Cd, Pb, Hg have tended to accumulate in the tissue of clams. This paper presents the levels of some heavy metals in the seawater, sediment and soft part of oyster (*Crassostrea belcheri*) samples collected from the western estuaries of Ganh Rai bay in 2015 and 2017. The results also showed that Ha Thanh and Rach Lo sites recorded the highest contents of most studied metals in oyster samples. Levels of metals in oyster were in the order of Zn > Cu > As > Pb > Cr > Cd > Hg and the contents of metals in oyster did not reflect a correlation with those in surrounding environment. Concerning food safety criteria, Pb, Cd and Hg contents were lower than acceptable limit given by the compilation of FAO (1983), whereas As, Cu and Zn contents exceeded the legal limit, especially Zn contents. Cu and Cr contents in sediment samples of Nga Bay and Dong Hoa estuaries were between LEL-SEL values (\geq LEL and $<$ SEL), which may cause biological impacts at moderate level.

Keywords: Heavy metals, estuary, coastal area, oyster, Can Gio.

INTRODUCTION

Heavy metals have reached the marine environment by the different pathways. Some heavy metals enter the sea by the interaction of atmospheric and sea surfaces. Beside, rivers are considered as major sources of metals. The concentration of metals in river water depends on the characteristics of catchments. Metals tend to participate in suspended matters formed by flocculation and then, accumulate onto sediments through the sedimentation process. On the other hand, the coastal regions are habitats of a large portion of aquatic organisms so that most of seafood is harvested from coastal zones. For this reason, heavy metals in coastal pollution have significant economic and social influences.

Molluscs are the major bottom organisms in the marine ecosystem. Molluscs are filter feeders, they can take all the elements in their food, which are algae, zooplankton, excreta of aquatic organisms in water column. Metals can be accumulated in their tissues to cause the risk for the higher species in the food chain. Moreover, exposure of molluscs in contaminated environment may lead to decline of population, reduced fertility and survival of larvae, juveniles. Two major uptake of metals in feeding aquatic organisms are (1) ingestion of metal enriched sediments and suspended particles, and (2) uptake from water [1].

Heavy metals can be subdivided into two main groups: essential and non-essential metals. One group includes iron (Fe), manganese (Mn), magnesium (Mg), cobalt (Co), zinc (Zn), copper (Cu) which are essential elements for the growth and life cycle of organisms, but are toxic at high concentrations [2]. Heavy metals of second group consist of lead (Pb), cadmium (Cd), mercury (Hg), arsenic (As) which are

toxic even at low concentrations, have no biological functions in the metabolisms of aquatic organisms [3].

Can Gio, which is one of 24 districts of Ho Chi Minh city, is located 50 km southeast. Can Thanh coastal area of Can Gio district is the deltaic confluence of Sai Gon, Dong Nai and Vam Co rivers which spread out through Ho Chi Minh city and Dong Nai province that are the most populated areas and also the developed heavy industrial zones. Therefore, this area receives the harmful substances (from domestic and industrial wastewater, materials from erosion, oil spill,...) [4]. The previous studies showed that heavy metals exist in environments (water, sediment) and the commercial bivalve molluscs. For example, metals as Zn, Cu, Pb, Cd and Cr were found at high levels in water, suspended particulate matters (SPMs), sediment and clams (*Meretrix lyrata*) at the coastal area of Can Gio district [5]. They tend to accumulate into the soft tissues of clams (*Meretrix lyrata*), especially, As accumulated with the highest level in soft body of clams [6]. Therefore, the study on metal pollution in oyster and the environment of coastal areas of Can Gio is necessary. The objectives of this paper were (1) to quantify the spatiotemporal distribution of heavy metals (Zn, Cu, Cr, As, Cd, Pb and Hg) in sediments, seawaters, and oyster, and (2) to discuss the possible relationships between metal levels in oyster and those in the surrounding environment.

MATERIALS AND METHODS

Sampling

All samples were collected at low tide time in dry and rainy seasons in 2015 and 2017 in Ganh Rai bay. Sampling site details are given in table 1 and figure 1.

Table 1. Sampling sites description

Stations	Coordinate	Samples	Remarks: locations
2	10.473035°N, 106.940753°E	Water, sediment	Long Tau estuary, near the cultured oyster farms
3	10.439129°N, 106.922493°E	Water, sediment	Nga Bay estuary, near the cultured oyster farms
4	10.403630°N, 106.966017°E	Water, sediment	The mud flats, the cultured clam area of Can Thanh
Cau Den	10.420583°N, 106.958833°E	Water, sediment	Dong Hoa estuary, coastal area
Site 1	10.519418°N, 106.954974°E	Oyster	The cultured oyster places, LongTau estuary
Site 2	10.426104°N, 106.918164°E	Oyster	The cultured oyster places, Ha Thanh river
Site 3	10.419857°N, 106.950132°E	Oyster	The cultured oyster places, Huu Tri dam
Site 4	10.410768°N, 106.927147°E	Oyster	The cultured oyster places, Rach Lo river

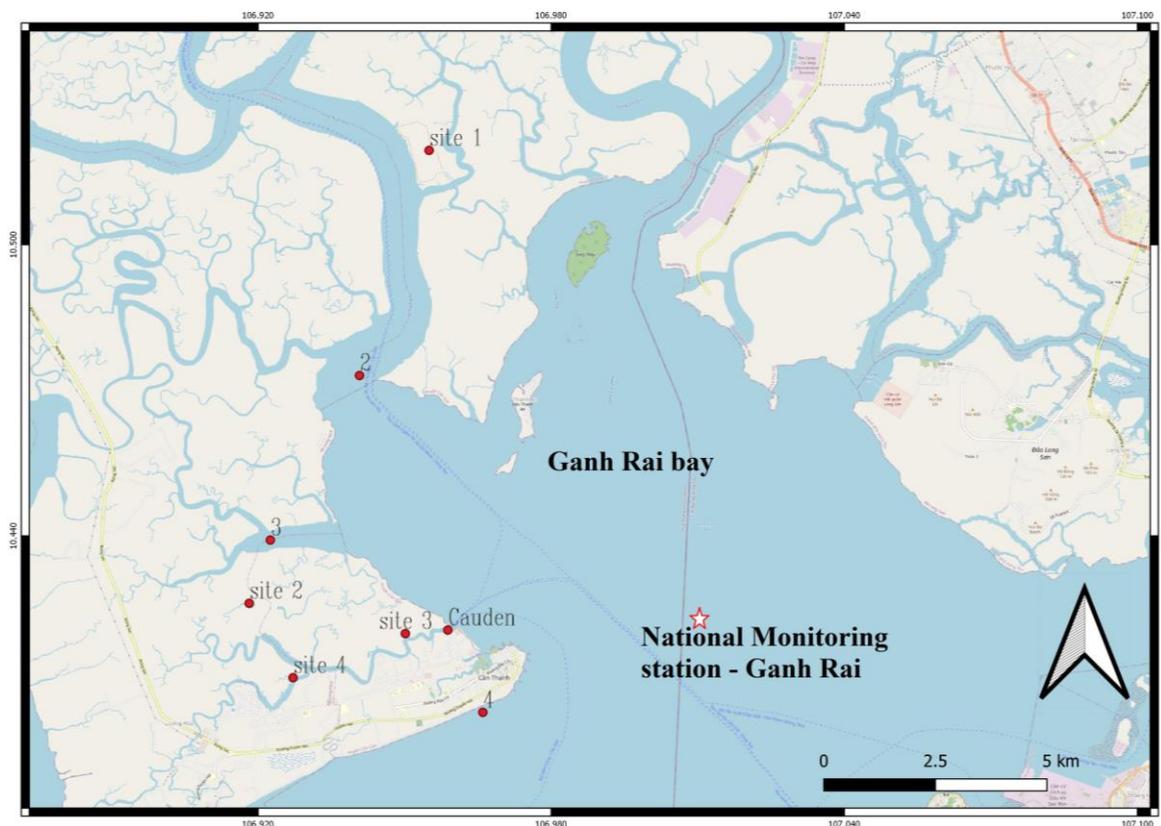


Figure 1. The study area

Oyster samples

10 to 20 samples of oyster with similar size of shell were selected at each of 4 farms (table 1). The shells were then washed and removed by stainless steel knife. Obtained soft tissues were then transferred to polyethylene bags and kept in cooler.

Seawater samples

About 500 ml of seawater was taken at stations 2, 3, 4 and Cau Den by Van Dorn sampler and stored in polyethylene bottles at low temperature.

Sediment samples were collected using the stainless steel grab sampler, stored in the plastic bags and kept in coolers with the temperature maintained at 4°C.

Samples analyses

Heavy metals in seawater: Collected seawater samples were filtered using 0.45 µm micropore membrane filter and then the concentrations of heavy metals in seawater

were measured by using Inductively Coupled Plasma Mass Spectrometry (ICP-MS Agilent 7700). The precision and accuracy of analysis were checked by measurement of a standard sample for ICP-MS for each of target metals. The standard curve with four different concentrations was set up by measurement of a standard mixture solution for seven metals. The Limit of Detections (LOD) of instrument for target metals was 0.3 µg/l for Zn, 0.1 µg/l for Cu, 0.1 µg/l for Pb, 0.01 µg/l for Cd, 0.1 µg/l for Cr, 0.03 µg/l for As and 0.01 µg/l for Hg.

Heavy metals in sediment: Collected sediment samples were dried at room temperature. The procedure for heavy metal extraction in the sediment followed the method given in the workbook of ASEAN Canada Cooperative Programme in 1998: about 0.20 g of sediment was weighted accurately, 20 ml ultrapure water, 100 ml of mixture solution (1:1) of highly purified HCl (Merck) and HNO₃ (Merck) were added, and then heated at 200°C

in 24 h. The sample was filtered by a quantitative filter (0.80 µm), and diluted to final volume of 250 ml with ultrapure water. Concentrations of heavy metals in the extraction were measured using Inductively Coupled Plasma Mass Spectrometry (ICP-MS Agilent 7700).

Metals in oyster tissue: The soft tissues were washed with distilled water and dried using filter papers. All of oyster specimens were homogenised. For heavy metal extraction, a portion of samples were treated with 10 ml of nitric acid 65% (Merck) and 10 ml of sulfuric acid 95% (Merck) and heated at 90°C with constant stirring following CNEXO (1983). The samples were filtered by Advantec filter paper and added to final volume of 100 ml with ultrapure water in the glass flask. Concentrations of heavy metals in the extraction were measured using Inductively Coupled Plasma Mass Spectrometry (ICP-MS Agilent 7700). Heavy metals contents in oyster samples are reported in µg/g wet weight.

Data analysis

Student’s two-tailed *t*-test was used for comparisons between the concentrations of individual metals in two seasons. A probability level of 0.05 was used for test, for *p* < 0.05, the difference of concentration means is significant with the test.

Pearson correlation analysis was used to test the relations between the metal

concentrations in oyster, water and sediment, significance was set at *p* < 0.05.

RESULTS AND DISCUSSIONS

Contents of heavy metals in sediment

Heavy metal concentrations in sediments were observed at 4 stations along Can Thanh coast in dry season in 2015 and 2017. Those values (µg/g) were reported in table 2. Heavy metal contents were not significantly different between two years (*p* > 0.05). Maximum average contents of Zn, Cr, As, Cd were recorded at Cau Den station, estuary of Dong Hoa river. These values of Cu, Hg and Pb were detected at station 3 and station 2 (Nga Bay and Long Tau estuaries). Station 4 located at the mud flats, which are the cultured clam farm of Can Thanh, showed the lowest values. The grain size is the main factor which affect the accumulation of metals on surface sediment. The sediments composed of the smaller grain sizes contain more metals and organic matters than those of coarser ones [10]. From results of monitoring program at Can Thanh coastal area in 2015, grain sizes (< 62 µm) at stations 2, 3, Cau Den and 4 (83.25%, 95.55%, 97.64% and 12.21%, respectively) explained that high levels of metals were detected at stations 3 and Cau Den. In addition, Zn, Cu, Cr, As levels in sediment of the estuary areas were recorded high in comparison with those in sediment of Ganh Rai bay (National monitoring station).

Table 2. Heavy metal contents in sediments

Stations	Year	Zn (µg/g)	Cu (µg/g)	Cr (µg/g)	As (µg/g)	Cd (µg/g)	Hg (µg/g)	Pb (µg/g)
2	2015	53.6	22.4	22.2	1.7	0.14	0.16	23.9
	2017	62.1	13.2	30.1	3.2	0.10	0.21	27.7
3	2015	62.9	21.2	58.7	4.1	0.18	0.34	22.9
	2017	77.1	29.7	50.3	3.3	0.13	0.18	21.1
4	2015	20.2	3.9	24.1	1.4	0.13	0.06	8.5
	2017	24.8	4.0	21.0	2.2	0.04	0.14	9.2
Cau Den	2015	71.5	11.8	60.8	3.4	0.21	0.26	18.5
	2017	74.7	26.2	64.4	6.4	0.21	0.16	21.9
National monitoring station (2015–2017)	Mean	61.56	16.19	29.81	4.20	0.66	0.14	20.84
Legal limits*		271	108	160	41.6	4.2	0.7	112
Lowest Effect Level (LEL)**		120	16	26	6.0	0.6	-	31
Severe Effect Level (SEL)**		270	110	110	33.0	9.0	-	110

Notes: *: National Technical Regulation on Sediment Quality, QCVN 43:2017/BTNMT; **: New York Sediment Screening Criteria.

Pearson correlation analysis indicated that Zn is positively correlated significantly with Cu, Pb, Cr. The correlation coefficients between Zn and Cu, Pb and Cr were 0.813, 0.792 and 0.782 ($p < 0.05$), respectively. Correlation between Cr with Cd and As showed the same trend ($r = 0.812, 0.834, p < 0.05$), especially, correlation coefficient between As and Cr was highest. The results of correlation analysis suggested that those metals may enter the environment from the same source. The strong positive correlation of Cr and other metals as Zn, Pb, Cu, Cd, As proved that sources of these metals had the same origin of Cr pollution, which comes from industrial, agricultural activities and household sewage into the river water. The poor associations of Cd may be explained by the biological effects, the study area is adjacent to the Can Gio mangrove forest, therefore Cd can be absorbed by the mangrove roots to decrease Cd levels in environment [10].

The obtained data show that most of metals had contents below LEL values, except for Cu and Cr. Contents of these metals at stations 3 and Cau Den had values in between LEL-SEL values, which may cause the moderate biological effects.

Concentrations of heavy metals in seawater

The analysis results (expressed as $\mu\text{g/l}$) were shown in table 3. The highest mean values of Zn, Cu and Cr were found at station 2 in rainy season, while the highest values of Pb, As, Hg and Cd were recorded at stations 2 and 4 in the same season. Zn concentrations were dominant in both seasons compared to the other metals, maximum Zn levels was detected at Cau Den (17.9 $\mu\text{g/l}$ and 19.8 $\mu\text{g/l}$ in dry and rainy seasons), concentrations ranged from 5.6 $\mu\text{g/l}$ to 19.88 $\mu\text{g/l}$. The variation trend of Zn was similar in both seasons, Zn levels tended to increase at station Cau Den, and decreased slightly at station 4. Maximum concentrations of Cu, Cr, Pb, As, Hg occurred in rainy season at stations 2 (Pb, As, Hg) and Cau Den (Cu, Cr), the highest Cd concentration was detected at Cau Den in dry season (0.9 $\mu\text{g/l}$). Cd concentration ranged from 0.09 $\mu\text{g/l}$ to 0.9 $\mu\text{g/l}$, Cd tended to increase at Cau Den and decreased sharply at station 4 in dry season, while in rainy season it increased slightly from

station 2 to station 4, low values of Cd contents may be explained by the absorption of roots of mangroves. Cu, Pb, As, Hg and Cr presented their major concentrations in rainy season, ranges of metal concentrations were from 1.8 $\mu\text{g/l}$ to 7.2 $\mu\text{g/l}$ with Cu, Pb: 1.1 $\mu\text{g/l}$ to 6.7 $\mu\text{g/l}$, As: 1.1 $\mu\text{g/l}$ to 9.2 $\mu\text{g/l}$, Cr: 1.5 $\mu\text{g/l}$ to 10.2 $\mu\text{g/l}$ and Hg: 0.1 $\mu\text{g/l}$ to 0.89 $\mu\text{g/l}$. The variation patterns of Cu, As and Cr were similar, metal levels increased at Cau Den (Cr, Cu) and decreased at station 4 (Cu, Cr, As). Levels of these 3 metals were clearly different at Cau Den, whereas slight variations were observed at the other stations. The variation trends of Hg were similar in both seasons, Hg concentrations decreased from station 2 to Cau Den, then raised at station 4. Mean values of metals in seawater were not statistically different between two seasons, except for Pb, average value of Pb concentrations in rainy season was significantly higher ($p < 0.05$) than these values in dry season. On the other hand, most of concentrations of metals at the estuary areas (stations 2, 3, Cau Den) were higher than those values of seawater in Ganh Rai bay (National monitoring station) in rainy season. It showed that the heavy metal contamination at the estuarine areas may be increased by the runoff from the residential areas in the wet seasons.

Pearson correlation analysis indicated that positive correlation existed between Zn-Cu, Zn-Cr, Cr-Cu ($r = 0.497, 0.570, 0.535, p < 0.05$) and As-Cu, As-Pb, Cu-Pb, As-Hg ($r = 0.642, 0.513, 0.581, 0.646, p < 0.05$) in seawater. The positive correlation between Zn, Cr, Cu, As, Pb suggested that these metals were grouped together. These metals may come from the same origins. The positive correlations were reported between As-Pb and As-Hg with high coefficients ($r = 0.632$ and 0.646). Cadmium and the other metals did not significantly correlate, thus Cd pollution sources may differ from those of other metals. It was shown that positive correlation existed between metal pairs Zn-Cu, Zn-Cr both in sediment and seawater ($r = 0.813, 0.782$ and $0.497, 0.570, p < 0.05$), which suggested the same contamination origin with Cr pollution from industrial, agricultural, household sewage discharge and river transportation.

Table 3. Heavy metal concentrations in seawater

Stations	Seasons	Values	Zn ($\mu\text{g/l}$)	Cu ($\mu\text{g/l}$)	Cr ($\mu\text{g/l}$)	As ($\mu\text{g/l}$)	Cd ($\mu\text{g/l}$)	Hg ($\mu\text{g/l}$)	Pb ($\mu\text{g/l}$)
2	Dry	mean	9.85	3.35	4.7	4.2	0.16	0.46	2.1
		range	8.8–10.9	2.8–3.9	2.6–6.7	3.2–5.2	0.1–0.2	0.1–0.8	1.5–2.7
		n	2	2	2	2	2	2	2
	Rainy	mean	9.45	4.55	3.5	6.2	0.21	0.50	4.9
		range	9.2–9.7	3.2–5.9	2.1–4.9	3.2–9.2	0.1–0.3	0.1–0.9	3.1–6.7
		n	2	2	2	2	2	2	2
3	Dry	mean	8.40	3.05	3.6	3.8	0.205	0.44	2.5
		range	7.4–9.4	2.5–3.6	2.0–5.2	2.9–4.7	0.1–0.3	0.1–0.8	1.5–3.5
		n	2	2	2	2	2	2	2
	Rainy	mean	9.60	2.70	3.9	4.8	0.20	0.38	4.5
		range	8.6–10.6	2.5–2.9	1.6–6.2	3.8–5.7	0.1–0.3	0.1–0.7	3.7–5.2
		n	2	2	2	2	2	2	2
4	Dry	mean	9.50	2.70	3.7	2	0.175	0.25	1.6
		range	8.2–10.8	2.5–2.9	1.8–5.6	1.5–2.5	0.15–0.2	0.2–0.3	1.1–2.1
		n	2	2	2	2	2	2	2
	Rainy	mean	7.10	3.35	4.5	2.0	0.31	0.18	4.2
		range	5.6–8.6	3.1–3.6	3.8–5.1	1.1–2.9	0.1–0.5	0.1–0.2	2.6–5.7
		n	2	2	2	2	2	2	2
Cau Den	Dry	mean	12.55	2.63	2.9	2.2	0.6	0.12	1.8
		range	7.2–17.9	1.8–3.5	1.5–4.2	1.6–2.8	0.3–0.9	0.11–0.13	1.2–2.4
		n	2	2	2	2	2	2	2
	Rainy	mean	12.80	5.88	6.0	5.1	0.23	0.16	4.5
		range	5.8–19.8	4.6–7.2	1.7–10.2	3.3–6.8	0.16–0.3	0.13–0.2	4.45–4.5
		n	2	2	2	2	2	2	2
National monitoring station (2015–2017)	Dry	mean	9.80	4.10	4.15	3.20	0.16	0.12	2.18
	Rainy	mean	7.95	2.93	4.68	3.80	0.13	0.14	2.68
Legal limits*			500	200	100	20	5	1	50

Note: *: National Technical Regulation on Marine Water Quality, QCVN 10-MT: 2015/BTNMT.

Contents of heavy metals in oyster

The metal contents in oyster tissues ($\mu\text{g/g}$) were reported in wet weight (ww) and illustrated in figure 2, table 4. Length and weight of oysters were shown in table 5. Zinc concentrations were higher than those of dry season at four sites. The maximum value of Zn was found at site 3 - Can Thanh in rainy season, Zn levels ranged from 68.9 $\mu\text{g/g}$ to 159.4 $\mu\text{g/g}$. On a seasonal scale, the increase of Zn contents in soft tissues of oyster in rainy season was statistically significant ($p < 0.05$), whereas levels of other metals were not different between two seasons. The highest Cu level exhibited in dry season at site 2 - Ha Thanh, Cu levels varied from 8.43 to 26.16 $\mu\text{g/g}$ ww. The contents of Pb, Cd were reported with considerable values at site 4 - Rach Lo in dry season. Pb levels ranged from 0.3 to 0.98 $\mu\text{g/g}$ ww. In dry season, Pb contents increased

gradually from site 1 to site 4, and were not different between sampling sites in rainy season. Cd levels in dry season were higher than in rainy season, except for site 4, range of Cd contents was from 0.12 to 0.77 $\mu\text{g/g}$ ww. The seasonal variation of Cr levels was slight at sites 1 and 3, whereas at sites 2 and 4, higher values of Cr levels were recorded in dry season, variation of Cr contents was from 0.08 to 0.69 $\mu\text{g/g}$ ww. Hg contents in soft tissues were detected at the lowest levels, almost equal to detection limit, the seasonal variations of Hg were not clear at sampling sites, except for site 2 - Ha Thanh, concentration values varied from 0.03 to 0.26 $\mu\text{g/g}$ ww.

The most abundant metals in *Crassostrea belcheri* were Zn and Cu. These two metal levels in *Crassostrea belcheri* can be considered low in comparison with those values in other *Crassostrea* sp. studies [7, 11,

12]. However, Zn and Cu contents in this species were higher than those in the other bivalve molluscs from Khanh Hoa coast, Central Vietnam (Tran Thi Mai Phuong, Doctoral thesis). It may be explained by the different capacities for accumulating metals of different species. Zn levels were usually

higher than those of Cu in both seasons. It is common to find higher concentrations of Zn in the bivalve studies [11–14]. Hg levels presented the the lowest values in both seasons. Pham et al., (2007) reported similar concentrations of this metal in bivalve species from Can Thanh [6].

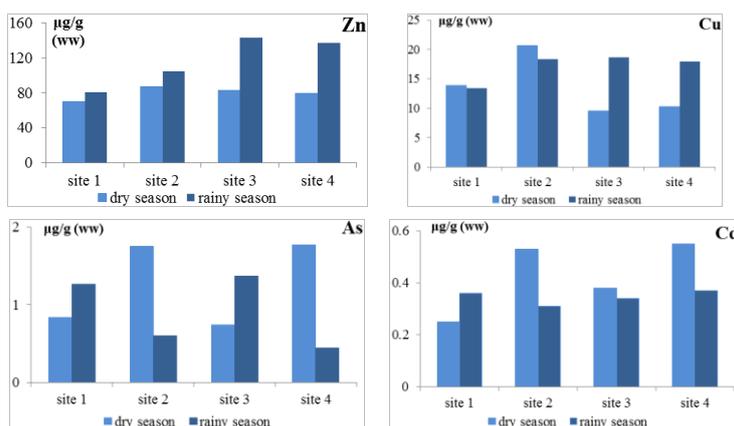


Figure 2. Heavy metal contents in oysters

Table 4. Heavy metal contents in oyster

Stations	Seasons	Values	Zn (µg/l)	Cu (µg/l)	Cr (µg/l)	As (µg/l)	Cd (µg/l)	Hg (µg/l)	Pb (µg/l)
Site 1	Dry	mean	70.4	13.9	0.4	0.8	0.3	0.054	0.37
		range	68.9–71.8	8.4–19.4	0.10–0.69	0.44–1.24	0.12–0.38	0.05–0.06	0.34–0.39
		n	2	2	2	2	2	2	2
	Rainy	mean	80.5	13.4	0.4	1.3	0.4	0.1	0.5
		range	76.8–84.2	10.5–16.3	0.15–0.68	0.73–1.78	0.22–0.50	0.04–0.19	0.42–0.66
		n	2	2	2	2	2	2	2
Site 2	Dry	mean	86.9	20.6	0.63	1.8	0.5	0.2	0.5
		range	74.0–99.8	15.1–26.2	0.63–0.63	0.74–2.76	0.36–0.70	0.12–0.26	0.42–0.55
		n	2	2	2	2	2	2	2
	Rainy	mean	104.8	18.3	0.4	0.6	0.3	0.1	0.6
		range	104.2–105.3	17.4–19.3	0.18–0.54	0.44–0.76	0.22–0.40	0.04–0.16	0.48–0.63
		n	2	2	2	2	2	2	2
Site 3	Dry	mean	82.8	9.5	0.3	0.7	0.4	0.05	0.5
		range	70.3–95.2	9.2–9.8	0.12–0.40	0.68–0.79	0.31–0.45	0.04–0.06	0.35–0.65
		n	2	2	2	2	2	2	2
	Rainy	mean	143.1	18.7	0.3	1.4	0.3	0.035	0.6
		range	126.7–159.4	17.8–19.5	0.19–0.42	0.57–2.16	0.28–0.40	0.03–0.04	0.40–0.78
		n	2	2	2	2	2	2	2
Site 4	Dry	mean	79.7	10.2	0.6	1.8	0.5	0.056	0.8
		range	79.0–80.4	9.1–11.3	0.42–0.68	1.05–2.48	0.32–0.77	0.05–0.06	0.52–0.98
		n	2	2	2	2	2	2	2
	Rainy	mean	137.0	17.9	0.3	0.4	0.4	0.065	0.6
		range	120.3–153.8	17.0–18.9	0.08–0.52	0.29–0.59	0.23–0.50	0.06–0.07	0.30–0.87
		n	2	2	2	2	2	2	2
Legal limits* (wet weight)			50	20	–	1.5	2.0	0.5	10
Legal limits**(wet weight)			–	–	–	–	1.0	0.5	1.5

Notes: *: Compilation of legal limits for hazardous substances in fish and fishery products, FAO (1983); **: Commission regulation (EC) No. 1881/2006.

Table 5. Length and weight of oysters

	Site 1		Site 2		Site 3		Site 4	
	Dry season							
	Length (cm)	Weight (g)	Length (cm)	Weight (g)	Length (cm)	Weight (g)	Length (cm)	Weight (g)
Mean ± SD	12.1 ± 0.5	170 ± 29	11.8 ± 0.8	144 ± 17.2	12 ± 0.4	157 ± 15.6	12 ± 0.6	164 ± 14.1
n	18	18	18	18	18	18	18	18
	Rainy season							
mean±SD	11.7 ± 1	180 ± 27	11.2 ± 0.8	154±13.8	12 ± 1.1	147 ± 12	11.9 ± 1	157 ± 15.8
n	20	20	23	23	23	23	15	15

Concerning the food safety criteria, metal concentrations in *Crassostrea belcheri* were compared with the standard values from European legislation (EC, 2006), FAO (1983). Zinc levels in *Crassostrea belcheri* were almost higher than the permissible value of FAO (50 mg/kg in fresh weight) for all of sites in both seasons. Highest copper level was found to exceed the legal limit of FAO (20 mg/kg fresh weight) in dry season at site 2 - Ha Thanh. All of contents of Pb, Cd, Hg did not exceed the standard values of European regulation (1.5 mg/kg for Pb, 1.0 mg/kg for Cd in fresh weight). The highest As content was recorded to exceed the legal limit of FAO (1.0 mg/kg fresh weight) at site 2 - Ha Thanh in dry season. These are no regulations for chromium in these legal documents. Chromium is considered an essential element for the bivalves, but this metal can cause the risks for human consumption.

Correlation of heavy metals in oyster and environment

Zinc is an essential element, in present work, this metal was usually accumulated with higher levels in tissues than those in water and sediment and the other metal levels, because Zn is necessary for enzymes of metabolic activities. This result was similar to other studies, Rebelo et al., (2003) reported that oyster can accumulate more Zn contents than in the ambient environment [15]. Copper is also an essential element, Cu contents in oyster tissues were higher than those in water ($p < 0.05$) in both seasons, accumulated for use in their metabolisms. Cu levels in oyster and water were not correlated, it is similar to results in study of Birch et al., (2014) on oyster *Saccostrea glomerata* [16]. The trend of seasonal variation of Pb levels in oyster was

similar to those in water. However, Pb levels in oyster were significant lower than in surrounding environment ($p < 0.05$), it may be concluded that oyster *Crassostrea belcheri* in present study had no obvious tendency to accumulate Pb. Phillips et al., (1982) and Riget et al., (1997) also reported that lead was not significantly accumulated by marine biota [17], [18]. Contents of As, Cd, Hg were significantly lower than those in environment ($p < 0.05$). Moreover, Kargin et al., (1998) proved that levels of non-essential metals in aquatic organisms depend on the degree of presence in environment [19]. It may be suggested that low levels of Cd, Pb, Hg and As in oyster tissues were affected by the poor contents of these metals in the ambient environment. Contents of Cd, Pb in this work were slightly higher than those in oysters from Adriatic coast, while Hg levels were similar to the values which were found in the literatures by Astudillo et al., (2005); Cubadda et al., (2006); Gavrilovic et al., (2007); Laura Bille et al., (2015) [20–23]. Chromium levels in oyster were lower than in environment ($p < 0.05$), it may be explained that marine shellfish have low tendency to accumulate chromium [24], Cr levels in this study was similar to concentrations found in other study [25]. Mercury in present work exhibited the lowest levels, but it is still potential risk for human consumption by the bioamplification through food webs.

Determination of the influence of ambient environment on metal levels in oyster tissues is complex, because many sources can impact on variation of metal contents in oyster, for example, the accumulation and excretion of oysters can affect levels of the essential and non-essential metals in tissues. In the present work, essential metals were found with higher

levels than non-essential metals. Contents of metals in oyster did not clearly reflect the status of metal levels in surrounding environment.

CONCLUSIONS

Heavy metals exist in the estuaries with higher levels in comparison with those values in the tidal flats. In general, the heavy metal contents in the environment (and sediment) of western estuaries of Ganh Rai lagoon were lower than acceptable limit given by national technique regulations. However, contents of Cr and Cu in the sediment of Nga Bay (station 3) estuary and Dong Hoa (Cau Den) may cause biological effects at moderate levels (\geq LEL and $<$ SEL).

Contents of essential metals were found with higher levels than non-essential metals in oyster tissue, these values followed the order Zn > Cu > As > Pb > Cr > Cd > Hg. In addition, contents of metals in oyster did not clearly reflect the status of metal levels in surrounding environment. Concerning the food safety, Pb, Cd and Hg contents were lower than the permissible limits. Meanwhile, As, Cu and Zn contents exceeded the legal limits in compilation of FAO (1983), especially Zn contents were almost higher than the legal limit for all of sites in both seasons.

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