CONCENTRATION DISTRIBUTION OF HEAVY METALS IN MANILA CLAM (*RUDITAPES PHILIPPINARUM*) AND POTENTIAL HEALTH RISK IN THE COASTAL AREAS OF LIAODONG BAY, CHINA

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Abstract. Arsenic (As), Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb), and Zinc (Zn) concentrations in Manila clam from Liaodong Bay (China) were determined by inductively coupled plasma mass spectrometry. Tissue-specific bioaccumulation, geographical variability (using metal pollution indices) and health risks (using target hazard quotients and maximum daily consumption values) were assessed. Cd concentrations were high at all sampling sites, particularly S6 and S7, where it reached Category III. Other metal concentrations were high at S7 (As), S1 (Cr), S5 (Pb), and S5 and S7 (Zn). As, Cr and Cu accumulated to a higher degree in the viscera than muscles, but Cd accumulated more in muscles than viscera. Heavy metal (HM) contamination was worse in the west than that in the east. The Cd target hazard quotients were greater than 1, but the other HM target hazard quotients were less than 1 at all sites, indicating Cd posed the greatest health risks to humans. The maximum daily consumption values also indicated Cd poses health risks.
Keywords: *heavy metals, clam, bioaccumulation, geographical variability, health risk estimate, Liaodong Bay*

Introduction

Liaodong Bay is one of the three main bays in the Bohai Sea. Water exchange between Liaodong Bay and the open sea is slow, so pollutants are not readily transported out of Liaodong Bay. Many rivers (e.g., the Dalinghe River, Liaohe River, and Xiaolinghe River) flow into the bay area. There are cities of different sizes around Liaodong Bay, and large parts of the coast are occupied by harbors, industrial areas and aquaculture facilities. Large amounts of domestic sewage and industrial effluent containing various pollutants are discharged into Liaodong Bay (Zhang et al., 2016; Naser, 2013; Gargouri et al., 2011; Wan et al., 2008a). These wastewaters also contain large amount of heavy metals (HMs). HM contamination is a serious issue around the world (Zhang et al., 2016; Gao and Chen, 2012; Hu et al., 2013).

HMs are persistent, stable, toxic, non-biodegradable, and bioaccumulative. HMs can accumulate in organisms and can cause many diseases (Li et al., 2015). HMs in the marine environment therefore pose health risks to aquatic organisms and humans consuming aquatic organisms. It is necessary to determine HM concentrations in aquatic media and organisms to gain an understanding of the health risks posed by contaminated seafood to humans consuming (Wei et al., 2014; Li et al., 2015).

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Bivalves as benthic species can accumulate many pollutants, including HMs, to which they are very tolerant. Bivalves are therefore used as bioindicators of HM contamination in aquatic systems (Li and Gao, 2014; Won et al., 2016; Shoults-Wlison et al., 2015). The bivalve Manila clam (*Ruditapes philippinarum*, *Rud*) is generally found in sediment and tends to accumulate HMs present in the sediment (Zhao et al., 2012). *Rud* is easily collected in Liaodong Bay. *Rud* is considered to be delicious, and is consumed in large quantities. Determining the concentrations of HMs in *Rud* in Liaodong Bay will allow HM pollution of the aquatic environment and health risks posed to humans consuming contaminated seafood to be assessed (Liang et al., 2004).

In previous studies of HM in the Bohai Sea, most sampling sites were located along the Bohai Bay coast (Li et al., 2015; Zhang et al., 2016; Wang et al., 2005) and in Laizhou Bay (Liu et al., 2017). Few sampling sites in Liaodong Bay were used. In this study, we determined the concentrations of Arsenic (As), Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb), and Zinc (Zn) in muscles and viscera from *Rud* collected from seven sites along the Liaodong Bay coast. The main objectives were 1) to determine HM concentrations in *Rud* from the different sampling sites, 2) to assess tissue-specific HM bioaccumulation by *Rud* 3) to assess geographical variability in HM concentrations in *Rud* using metal pollution indices (MPIs), and 4) to assess the risks posed by HMs in *Rud* to human health using national and international guidelines, target hazard quotients (THQs), and maximum daily consumption rates (CR_{max}s).

Materials and methods

Samples were collected from seven sites along the coastal area of Liaodong Bay (China) in July and August 2017, which is the main fishing season (*Fig. 1*). Attached mud and debris was removed from the surface of each shell. The *Rud* weights and shell lengths were measured. Muscle and viscera tissue samples were collected. Muscle tissue from three individuals were combined and homogenized to form a single sample. Viscera from 15 individuals were combined and homogenized to form a single sample to provide sufficient analytes for analysis (Liu et al., 2017). Six mixed muscle samples and six viscera samples from each sampling site were analyzed. The samples were lyophilized and ground into powder. The samples were analyzed using a previously described method (Li et al., 2015). Briefly, 0.2 g of a dry sample was placed in a polytetrafluoroethylene tube containing 3 mL HNO_3 (65%) (Suprapur; Merck, Damstadt, Germany) and 1 mL H_2O_2 (35%) (Suprapur; Merck). The tube was then placed in a Mars-5 microwave digestion instrument (CEM, Buckingham, UK) to digest the mixture. The digest was then diluted to 50 mL with ultrapure water and passed through a 0.45 μm membrane filter. The HM concentrations in the digest were determined using an Agilent 7500i inductively coupled plasma mass spectrometer (Agilent Technologies, Santa Clara, CA, USA). A standard reference material (GBW08571, mussel tissue) was analyzed to ensure the HM analysis method gave acceptable results. The mean recovery rates of the metals that were analyzed in the reference material samples were 82–112%.

The MPI was calculated for each sampling site to allow geographical variability in the HM concentrations in the Rud tissues to be assessed (Liu et al., 2017). The MPI was calculated using *Equation 1:*

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$$MPI = \left(Cf_1 \times Cf_2 \times \cdots \times Cf_n\right)^{\frac{1}{n}} \quad \text{(Eq.1)}$$

where Cf1 is the concentration of the first HM in muscle tissue (in this study) from the sampling site of interest, Cf2 is the concentration of the second HM, Cfn is the concentration of the nth HM, and n is the total number of HMs analyzed.

The risks posed by HMs in *Rud* to human health were assessed using THQs and CR_{max}s (US EPA, 2000; Li et al., 2015). The THQs and CR_{max}s were calculated from the HM concentrations in the muscle samples using *Equations 2* and *3:*

$$THQ = \frac{C \times W_{clam} \times ED \times EF}{BW \times RfD \times Atn} \times 10^{-3} \quad \text{(Eq.2)}$$

and

$$CR_{max} = \frac{RfD \times BW}{C} \quad \text{(Eq.3)}$$

where C is the mean concentration of a HM in *Rud* muscle (in mg/kg wet weight (ww)), W_{clam} is the daily *Rud* ingestion rate for an adult (38.9 g/d) (National Bureau of Statistics of China, 2009), ED is the exposure duration (74.8 y, the expected average lifetime) (MEP, 2013), EF is the exposure frequency (365 d/y), BW is the average body weight (63 kg for an adult) (National Physique Monitoring Center of China, 2012), RfD is the average oral reference dose for the HM of interest (US EPA, 2014), Atn is the average exposure time for a non-carcinogen (ED × 365 d/y), and 10^{-3} is the unit conversion factor.

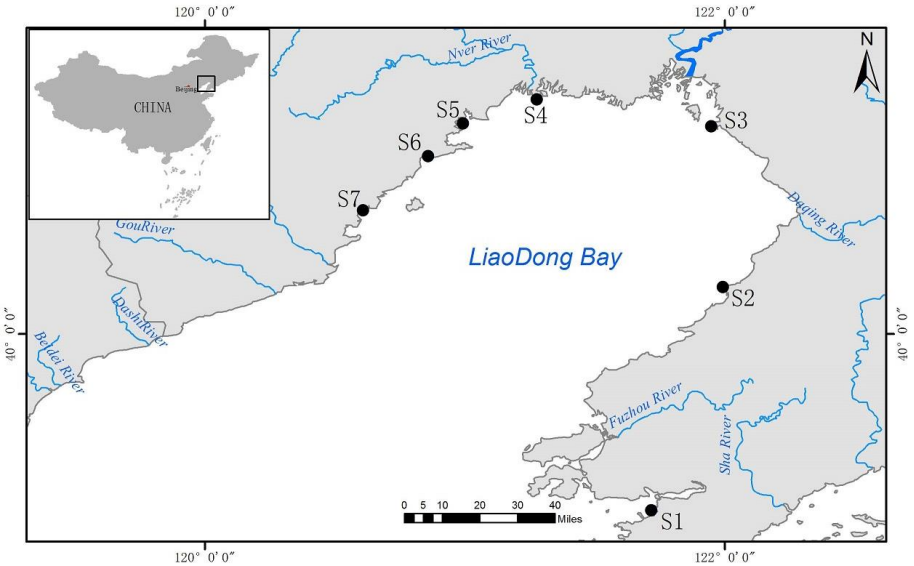

Figure 1. *Map of sampling sites at the Liaodong Bay, China. S1, Puwan (39°19'16.15785"N, 121°42'59.84795"E); S2, Bayuquan (40°10'46.84870"N, 121°59'34.03441"E); S3, Erjiegou (40°47'49.44684"N, 121°56'54.37565"E); S4, Qilihekou (40°54'2.31809"N, 121°16'37.68160"E); S5, Laohekou (40°48'33.39360"N, 120°59'32.76576"E); S6, Huludao (40°41'0.69727"N, 120°51'31.40488"E); S7, Shahousuo (40°28'27.18772"N, 120°36'32.63602"E)*

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Data analyses were performed using SPSS 18.0 software (IBM, Armonk, NY, USA). Each result is expressed as the mean ± standard deviation. ANOVA, SNK, and post hoc multiple comparisons, etc. were used to detect differences between groups. Differences between results for groups of samples were considered to be statistically significant at $P < 0.05$.

Results

Heavy metal concentrations in Rud

The HM concentrations found in the *Rud* muscle and viscera samples from each sampling site are shown in *Figure 2*. The Cd concentrations were quite high in the samples from all of the sites, particularly S6 and S7, the samples from which had Cd concentrations much higher than the Category III standard for Cd. The Cd concentrations in the samples from sites S1, S3, and S4 exceeded the Category II standard. The maximum permissible level for Cd, set by the World Health Organization in 1982, is 2 mg/kg ww, which is equivalent to the Category II standard. This indicated that attention needs to be paid to Cd pollution in Liaodong Bay. The As concentrations were higher in the samples from site S7 than in the samples from the other sites. The As concentration in the viscera samples from site S7 exceeded the Category II standard for As. The Cr concentrations were higher in the samples from site S1 than in the samples from the other sites but lower than the Category II standard for Cr. The Cu concentrations in all of the samples were low and were lower than the Category I standard for Cu. The Pb concentrations were higher in the samples from site S5 than in the samples from the other sites but were lower than the Category II standard for Pb. The Pb concentrations in the samples from the other sites were lower. The Zn concentrations in the samples from sites S5 and S7 exceeded the Category II standard.

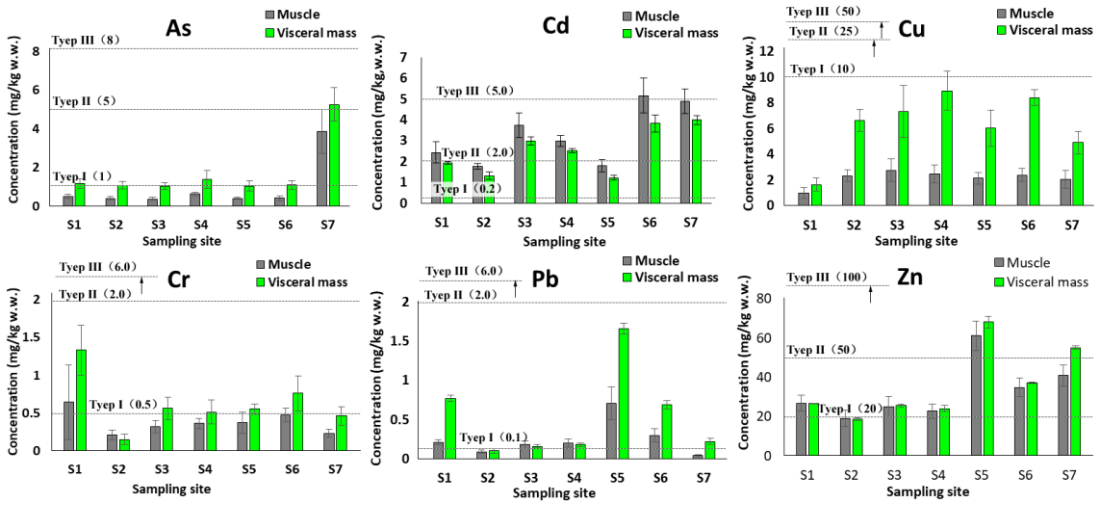

Figure 2. *Concentration ranges of heavy metals in different tissues at different sampling sites*

According to the standard (*Table 1*; *Fig. 2*), the concentrations of Cd, Pb and Zn in *Rud* have exceeded the allowable limits. Moreover, compared with previous studies (*Table 1*), the concentration of Cd was the highest and the others were present at a moderate level in the Liaodong Bay.

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Table 1. *Comparison of heavy metal concentrations in the soft tissues of shellfish from the Liaodong Bay, other areas in China, and world marine waters*

Site	Species		Year	As	Cd	Cr	Cu	Pb	Zn	Reference
Liaodong Bay	Ruditapes philippinarum	w. w.	2017	0.92	3.27	0.37	2.15	0.25	32.82	This study
Shanghai	Mactra chinensis	d. w.	2008 2009	0.2	0.03	0.1		0.13	14.8	Lei et al., 2013
A site of Bohai Bay	Mactra veneriformis	w.w.	2008	1.44	0.27	1.37	2.34	0.37	11.57	Li et al., 2015
B site of Bohai Bay				1.51	0.45	1.95	1.98	0.56	14.56	
C site of Bohai Bay				2.51	0.47	0.49	1.05	0.17	20.65	
Laizhou Bay	Ark shell	w.w.	2011	30.4	1.3	0.25	1.17	0.1	17.53	Liu et al., 2017
	Surf clam			1.16	0.22	0.48	1.11	0.21	9.89	
	Manila clam			3.08	0.26	0.38	1.78	0.22	21.35	
Daya Bay	Perna viridis	w.w.	2015	0.54	0.006	1.07	0.69	0.07	10.76	Gu et al., 2016
Catania Gulf	D. trunculus	w.w.	2012	1.53	0.01	0.25		0.07	7.63	Copat et al., 2013
Adriatic coastal area	Mytilus galloprovincialis	d.w.	2009		1.72		5.31	3.79	111.2	Jovic and Stankovic, 2014
Venezuelan coast	Crassostrea rhizophorae	d.w.	2008 2009		2.5	1.2	50.4	2.6	563.3	Alfonso et al., 2013
Grade I.	Shellfish	w.w		1.0	0.2	0.5	10	0.1	20	SEPA, 2001
Grade II.	Shellfish	w.w		5.0	2.0	2.0	25	2.0	50	SEPA, 2001
Grade III.	Shellfish	w.w		8.0	5.0	6.0	50	6.0	100	SEPA, 2001

w.w.: wet weight, d.w.: dry weight

Tissue-specific bioaccumulation of HMs in Rud

The results of one-way analyses of variance in the HM concentrations in the muscle and viscera samples are summarized in *Table 2*. The concentrations in the muscle and viscera samples were significantly different for As, Cd, Cr, and Cu ($p < 0.05$) but not for Zn and Pb ($p > 0.05$). Different tissue-specific bioaccumulation patterns were found for different metals (*Fig. 3*). The Cd concentrations were significantly higher in the muscle samples than the viscera samples ($p < 0.05$). The As, Cr, and Cu concentrations were significantly higher in the viscera samples than the muscle samples ($p < 0.05$).

Table 2. *One-way ANOVA results running on the heavy metal concentrations (mg/kg wet wt) in the muscles and visceral masses of Rud from Liaodong Bay*

HM	Cd	Cr	As	Cu	Zn	Pb
F	4.29	10.93	4.59	65.76	0.73	9.40
p	0.043	0.002	0.036	0.002	n.s.	n.s.

n.s.: no statistical difference

Geographical variability in the HM concentrations in Rud

HM concentrations in samples from different areas

Comparison of the HM concentrations (mg/kg dry wt) in the muscles of *Rud* in different sampling regions of Liaodong Bay are summarized in *Figure 4*. The west coast is S4–S7, and the east coast is S1-S3. The As, Cd, and Pb concentrations were significantly higher along the west coast than along the east coast ($p < 0.05$). The Cr, Cu, and Zn concentrations along the east and west coasts were not significantly different ($p > 0.05$).

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Rud MPIs in different areas

MIPs were calculated according to *Equation 1*. The MPIs for the different sampling sites are summarized in *Table 3*. The MPIs for the sampling sites decreased in the order S4 > S5 > S6 > S7 > S3 > S1 > S2. The MPIs were higher for the west coast (sites S4–S7) than the east coast (sites S1–S3). These results indicated that HM contamination is more serious along the west coast than the east coast.

Table 3. *MPI values of Rud in different sampling sites of Liaodong Bay*

Eastern site	MPI	Western site	MPI
S1	3.36	S4	4.61
S2	2.70	S5	4.50
S3	3.71	S6	4.45
		S7	3.93
Mean value	3.26	Mean value	4.37

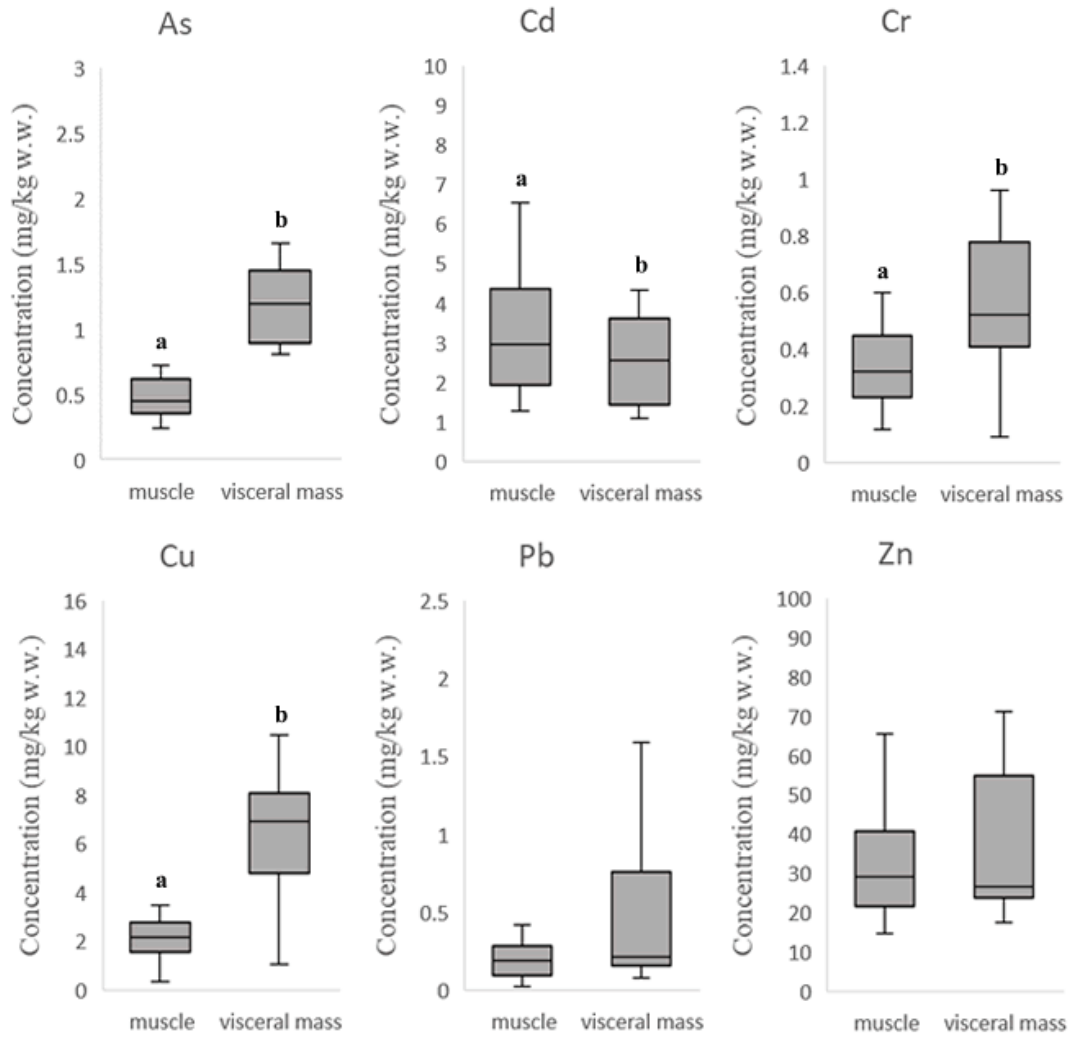

Figure 3. *Boxing plotting of the heavy metal concentrations (mg/kg dry wt) in the tissues of Rud from Liaodong Bay. Different superscript letters indicate significant differences at $p < 0.05$ ($n = 6$)*

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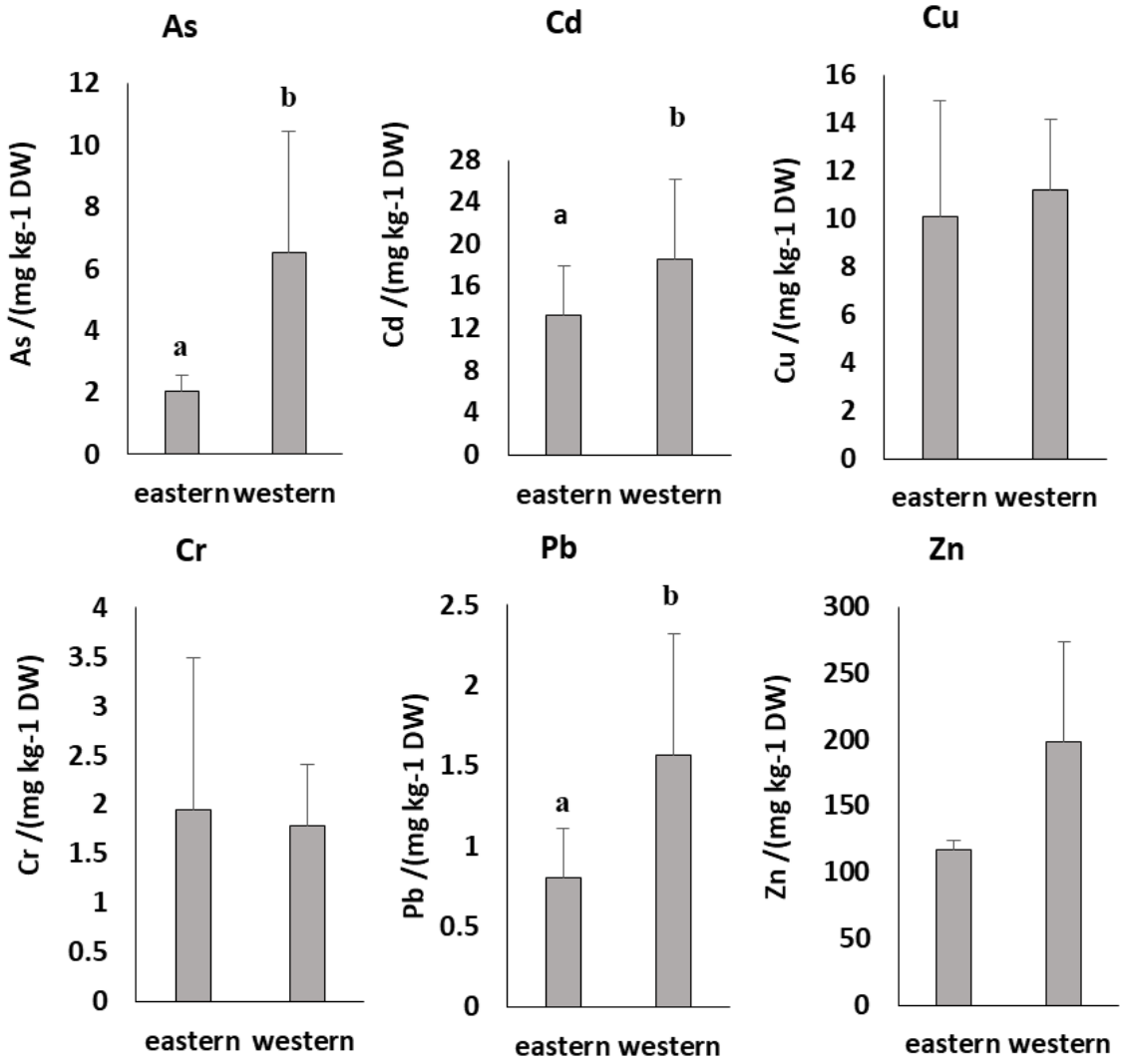

Figure 4. *Comparison of the heavy metal concentrations (mg/kg dry wt) in the muscles of Rud in different sampling regions of Liaodong Bay. Different superscript letters indicate significant differences at $p < 0.05$ ($n = 6$)*

Health risks posed to humans by HMs in Rud

The HM concentrations in the muscle tissues and national and international guidelines for the HMs are summarized in *Table 4*. The Cd concentrations in the muscle tissue samples from sites S1, S3, S4, S6, and S7 (2.45–5.18 mg/kg ww) exceeded the relevant limits for Cd (1–2 mg/kg ww). The As concentrations in the samples from sites S4 and S7 (0.6 and 3.84 mg/kg ww, respectively) exceeded the Chinese national guideline of 0.5 mg/kg ww. The Cr, Cu, Pb, and Zn concentrations (0.21–0.64, 0.98–2.76, 0.04–0.71, and 18.94–60.97 mg/kg ww, respectively) were within the relevant limits (2, 50, 1.0–1.5, and 100 mg/kg ww, respectively).

The potential health risks posed by HMs in *Rud* to humans were estimated by calculating THQs and CR_{max}s according to *Equations 2* and *3*. The results are summarized in *Tables 5* and *6*. Inorganic As is more toxic than organic As, and the calculations were performed assuming that inorganic As contributed 3% of the total As concentrations (Li et al., 2015).

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Table 4. *Heavy metal concentrations in the muscle tissues of Rud from Liaodong Bay. (mg/kg w.w.)*

	Safety guidelines	Heavy metal concentrations						
		S1	S2	S3	S4	S5	S6	S7
Cd	1[b],2[d]	2.45	1.78	3.74	3	1.82	5.18	4.91
Cr	2[a,b]	0.64	0.21	0.32	0.36	0.37	0.47	0.23
As	0.5 [a,b]	0.49	0.39	0.35	0.61	0.38	0.4	3.84
Cu	50[b]	0.98	2.33	2.76	2.45	2.13	2.38	2.04
Zn	150[e]	26.75	18.94	24.87	22.66	60.97	34.79	40.79
Pb	1.0[bd],1.5[c]	0.21	0.09	0.18	0.2	0.71	0.3	0.04

Table 5. *Estimated target hazard quotient (THQ) for Rud*

HMs	RfD (mg/kg·day)	THQ						
		S1	S2	S3	S4	S5	S6	S7
Cd	5×10^{-4}	**3.03**	**2.20**	**4.62**	**3.71**	**2.25**	**6.40**	**6.06**
Cr	3×10^{-3}	0.13	0.04	0.07	0.07	0.08	0.09	0.05
As	3×10^{-4}	0.03	0.02	0.02	0.04	0.02	0.03	0.24
Cu	4×10^{-2}	0.02	0.04	0.04	0.04	0.03	0.04	0.03
Zn	3×10^{-1}	0.06	0.04	0.05	0.05	0.13	0.07	0.08
Pb	1×10^{-2}	0.01	0.01	0.01	0.01	0.04	0.02	0.00

Table 6. *Estimated maximum consumption rates (CR_{max}) for Rud*

HMs	CR_{max} (g w.w.)						
	S1	S2	S3	S4	S5	S6	S7
Cd	12.86	17.70	8.42	10.50	17.31	6.08	6.42
Cr	295.31	900.00	590.63	525.00	510.81	402.13	821.74
As	1285.71	1615.38	1800.00	1032.79	1657.89	1575.00	164.06
Cu	2571.43	1081.55	913.04	1028.57	1183.10	1058.82	1235.29
Zn	706.54	997.89	759.95	834.07	309.99	543.26	463.35
Pb	3000.00	7000.00	3500.00	3150.00	887.32	2100.00	15750.00

A THQ ≤ 1 was taken to indicate no risks were posed to humans, and a THQ > 1 was taken to mean risks were posed to humans (Li et al., 2015). The Cd THQs (2.189–6.397) were higher than the other metal THQs for all the sampling sites. The concentrations of the HMs other than Cd in *Rud* from all of the sampling sites were found not to pose significant risks to humans (THQ < 1).

HMs can accumulate in *Rud*. Consuming certain amounts of contaminated *Rud* could therefore cause adverse health effects. CR_{max}s for theHMs in *Rud* were therefore calculated. The Pb CR_{max}s were relatively high, at 887.32–15750.00 g ww). The Cd CR_{max}s (6.08–17.70 g ww) were lower than the CR_{max}s for the other HMs, meaning the probability of health risks being posed to humans consuming *Rud* from Liaodong Bay was higher for Cd than the other HMs.

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Discussion

HMs in Rud

Liaodong Bay is the largest bay in the Bohai Sea. A complete water exchange cycle in Liaodong Bay takes 15 y (Wan et al., 2008a). Many rivers (including the Cishanhe River, Dalinghe River, Liaohe River, Shuangtaizihe River, Xiaolinghe River, and Wulihe River) discharge into Liaodong Bay. These rivers are thought to be important sources of HMs to Liaodong Bay (Wan et al., 2008b). The Cd concentrations in the samples from all of the sampling sites were quite high, and the concentration in the muscle samples from site S6 exceeded the Category III standard for Cd. This indicated that attention needs to be paid to Cd pollution in Liaodong Bay. The Cr and Pb concentrations were higher in the samples from sites S1 and S5, respectively, than in the samples from the other sites but lower than the Category II standards. The As concentrations in the viscera samples from site S7 exceeded the Category II standard for As, and the Zn concentrations in the viscera samples from sites S5 and S7 exceeded the Category II standards. These results indicated that the sites mentioned may be contaminated with these HMs. The Pb concentrations were not high. Pb and its salts can cause kidney damage and negatively affect the nervous system. Even low Pb concentrations in food cannot be ignored (Gao et al., 2014).

Compared with other domestic and international bays, Liaodong Bay has higher Cd concentration in shellfish (Lei et al., 2013; Li et al., 2015; Liu et al., 2017; Gu et al., 2016; Copat et al., 2013; Jovic and Stankovic, 2014; Alfonso et al., 2013). Cd pollution is worthy of attention.

Tissue-specific differences in HM concentrations

In clams, HMs tend to accumulate more efficiently in viscera than muscles (Husmann et al., 2012; Sarkar et al., 2008; Liu et al., 2017). Viscera play important roles in metabolic processes related to HMs. The hepatopancreas and kidney play important roles in HM absorption, transport, storage, and excretion. HMs in muscle tissue can be removed relatively quickly. In this study, As, Cr, and Cu had similar tissue-specific accumulation characteristics. The concentrations of these HMs were higher in the viscera than the muscles. In contrast, the Cd concentrations were higher in the muscles than the viscera. Similar results have been found in previous studies (Liu et al., 2017; Tarque et al., 2012). Insufficient data were available to explain this. In aquatic environments, Cd is mainly in the dissolved phase. Cd may easily be absorbed by tissues it comes into contact with (e.g., the mantle) in *Rud*. Large amounts of Cd may therefore accumulate in the mantle (Liu et al., 2017). This could explain the Cd concentrations being higher in the muscle samples than the viscera samples. Zn is an essential element that is absorbed and eliminated through various physiological processes in different tissues. Excess Zn may accumulate in the mantle of a clam. These factors may affect Zn accumulation in the muscles and viscera and explain the Zn concentrations in the muscle and viscera samples not being different.

Geographical differences in HM concentrations

HM contamination of *Rud* was more severe in the west of Liaodong Bay (at sites S4–S7) than in the east (sites S1–S3). Geographical variations in HM concentrations in clams are generally considered to be related to the distributions and characteristics of

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local sources of pollutants. The sampling sites along the west coast of Liaodong Bay were in Jinzhou Bay, a small cove in the northwestern part of Liaodong Bay. Jinzhou Bay has been contaminated by industrial effluent from chlor-alkali plants, petrochemical plants, the Huludao zinc plant, and the Bohai shipyard (Gao et al., 2014). Water in Liaodong Bay and the open sea is exchanged quite weakly, so HMs are not quickly transported from Liaodong Bay into the open sea. HMs released into Liaodong Bay can therefore accumulate in aquatic environmental media (e.g., sediment and clams).

Assessment of the risks posed by HMs in Rud to humans

The Cd concentrations in the muscle samples from sites S1, S3, S4, S6, and S7 exceeded the relevant limit. In particular, the Cd concentrations in the samples from site S6 exceeded the limit by a factor of ~2.5. The Cd THQs for all of the sampling sites were > 1 and were higher than the THQs for the other metals. The Cd CR_{max}s (6.08–17.70 g ww) were lower than the CR_{max}s for the other HMs that were analyzed. These results indicated that Cd would pose health risks to humans consuming *Rud* from the study area.

Industrial and other human activities are the main causes of Cd entering the environment. Cd can effectively accumulate in clams. Cd is very toxic to humans and can seriously affect the structures and functions of the bones, central nervous system (including the brain), kidneys, lungs, liver, and placenta. The *Rud* in the study area were heavily contaminated with Cd. Humans living around Liaodong Bay and consuming local *Rud* for a long period may be exposed to risks posed by Cd.

Rud was the only source of HMs to humans considered in the risk assessments performed in this study. Other sources include other types of seafood, rice, vegetables, and water, but these were not included in the risk assessments. The health risks found to be posed by HMs to humans in the risk assessments would have been stronger if all possible sources of HMs were considered.

Conclusions

The Cd concentrations were quite high in the samples from Liaodong Bay. The Cd concentrations in the samples from sites S6 and S7 were in the Category III standard for Cd. The concentrations of Cr in the samples from site S1, As in the samples from site S7, Pb in the samples from site S5, and Zn in the samples from sites S5 and S7 were also quite high. The *Rud* samples had typical tissue-specific heavy metal bioaccumulation patterns. Cd tended to accumulate more efficiently in muscle than viscera, whereas As, Cr and Cu tended to accumulate more efficiently in viscera than muscles. Pb and Zn were not significantly different in the muscle and viscera samples. Heavy metal contamination of *Rud* tended to be worse in the west than the east of Liaodong Bay. The heavy metal THQs and CR_{max}s for the muscle samples and national and international guidelines indicated that Cd was the main source of health risks to humans through consuming *Rud* from Liaodong Bay.

In summary, Cd was found to have an impact on the local environment and human health. Effective measures should be taken to control Cd pollution. Distribution characteristics and the bioaccumulation of HMs in seawater, surface sediments and other marine organisms will be studied in the future in order to take effective measures to control the pollution of HMs.

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