Front. Earth Sci. 2019, 13(2): 361–370
https://doi.org/10.1007/s11707-018-0724-x

RESEARCH ARTICLE

Metal accumulation in Asiatic clam from the Lower Min River (China) and implications for human health

Yue ZENG[1,2,3], **Zhongtao LI**[1], **Qianfeng WANG** (✉)[1,2,3], **Changcheng XU**[1,2,3], **Yunqin LI**[1], **Jia TANG**[1,2,3]

1 College of Environment and Resources, Fuzhou University, Fuzhou 350116, China
2 Key Laboratory of Spatial Data Mining & Information Sharing, Ministry of Education of China, Fuzhou 350116, China
3 Fujian Provincial Key Laboratory of Remote Sensing of Soil Erosion, Fuzhou University, Fuzhou 350116, China

Abstract Considering growing concerns regarding polluted estuaries and their adverse effects on public health, this study aimed to identify concentrations of metal (Zn, Fe, Cr, Ni, Cd, Mn, As, Cu, and Pb) in Asiatic clams sampled along the Lower Min River, China. Multivariate methods were used to identify and apportion pollution sources. Noncarcinogenic and carcinogenic health risk assessments were performed to gauge adverse consumer health effects. Results showed that Cr, Pb, and Zn concentrations were higher than the limits prescribed in Chinese government guidelines. In comparison with concentrations of selected metals in other rivers, Cr, Pb, Zn, and As concentrations in clams were generally higher. Pollution assessment using the metal pollution index showed that sampling sites surrounding developing industrial and residential areas were the most polluted. Principal component analysis indicated significant anthropogenic metal contributions in clams. Health risk assessment indicated significant risk for clam consumers along the Lower Min River in terms of hazard quotient and carcinogenic risk and, thus, clam consumption from the study area should be avoided. The present findings would help in establishing environmental monitoring plans and contribute to preserving public health as well as the development of water conservation strategies to alleviate the metal pollution.

Keywords metal accumulation, Asiatic clam, source identifications, health risk, Min River

1 Introduction

Metal pollution in the aquatic environment is of significant concern worldwide because of its high toxicity, persistence, and bioaccumulation (Förstner and Wittmann, 1979; Marsden and Rainbow, 2004). Arsenic (As), lead (Pb), and cadmium (Cd) are particularly toxic at relatively low doses (Bruins et al., 2000). Several other metals, such as iron (Fe), zinc (Zn), manganese (Mn), nickel (Ni), chromium (Cr), and copper (Cu) are essential for life but are toxic at high concentrations (Marschner, 1995; Türkmen et al., 2008). Metals may come from natural sources such as precipitation, erosion, and geological weathering, or anthropogenic sources including industrial and agricultural practices, vehicular traffic, domestic wastewater discharge, and city construction (Nriagu, 1996; Krishna et al., 2009; Wei et al., 2015). Human activities are major contributors to increasing environmental metal concentrations because of increasing urbanization and industrialization.

Bivalve mollusks, such as the Asiatic clam (*Corbicula fluminea*), accumulate a wide range of metals in their soft tissues. The habitat of this benthic species is relatively stable because they live buried within superficial sediment layers. Thus, clams are an important tool for the biomonitoring of environmental pollution in aquatic environments (Farrington et al., 1983; Liu et al., 2011). The reliability of clams as biomonitors of pollutant metal contamination has been demonstrated in a number of studies (Cairns and Pratt, 1993; Marie et al., 2006; Lewbart et al., 2010; Idriss and Ahmad, 2015). In field studies, Leland and Scudder (1990) and Reis et al. (2014) have found that metal accumulation in clams was several orders of magnitude higher than in relevant matrices, such as water and sediments. The characteristics of metal accumulation in clams have positive correlations with environmental metal distributions (Arini et al., 2014; Kong et al., 2016).

Asiatic clams are native to many parts of Africa, Asia, and Australia, inhabiting estuaries as well as freshwater systems, with temperature and salinity for growth at 9°C–32°C and < 8‰, respectively (Morton, 1986; Smaal, 2002; Zhang et al., 2007). As Asiatic clams are an edible shellfish

Received January 9, 2018; accepted July 2, 2018

E-mail: wangqianfeng@fzu.edu.cn

species favored by local residents and also marketed commercially, their high toxic pollutant concentrations yield them toxic to humans (Hussein and Khaled, 2014; Idriss and Ahmad, 2015). Measurements of metal concentrations in clam soft tissues and human health risk assessments have become increasingly significant because metals are nonbiodegradable and cannot be metabolized into harmless forms.

The Min River, which is the third largest river in Southeast China, is located in Fujian Province, adjacent to the East Sea. Fuzhou City, as the provincial capital, is located downstream on the Min River. Fuzhou City is a special coastal zone that can serve as a metal pollutant sink for runoff from upstream areas and surrounding lands and a secondary pollution source for the adjacent marine environment. The Lower Min River is an important drinking water supply for 2.3 million residents in the capital city of Fujian Province. Local residents customarily eat clams from the Min River. However, the river has been subjected to metal pollution for decades; metal sources in the clams were attributed to human and industrial activities in a previous study (Song and Huang, 1991). In recent years, with increasing expansion of urbanization and industries, pollution of clams and water by metals may result in adverse effects on human health.

However, limited information is available about the trend of metal contamination in clams in recent decades. So far, metal risk assessment on human health has not been carried out in this drinking water source area. Therefore, it is necessary to emphasize the sources of pollution and determine the potential threats of human exposure to toxic metals.

The present study aims to analyze the presence of metals in the soft tissues of Asiatic clams from the Lower Min River. The goals of the research were to (i) characterize metal contamination by determining metal concentrations in soft tissues of Asiatic clams from the Lower Min River, (ii) identify the natural and/or anthropogenic sources of metals, and (iii) evaluate the health risks to exposed humans using a hazard quotient (HQ) and carcinogenic risk (CR) via a health risk assessment model. This study supported future development of Min River conservation strategies to alleviate metal pollution and help residents take protective measures for minimizing potential adverse health effects.

2 Materials and methods

2.1 Study area

The Min River is the third largest river in Southeast China, and flows from the western regions with hilly terrain of Fujian Province eastward into the East China Sea. The basin area is ~60,992 km^2 and the average annual runoff 6.055×10^{10} m^3. The basin lies between latitude 25°23′N–28°19′N and longitude 116°23′E–119°43′E and features a subtropical monsoon climate, with an annual average temperature of 17°C–19°C and an annual average precipitation of 1724 mm. The Lower Min River is divided into the North and South Branches where it meets Nantai Island. The drainage area of the Lower Min River is ~8000 km^2. Fuzhou City lies at the downstream end of the Min River basin and, as the provincial capital ranking in the upper-middle level cities in China regarding earnings and population, this city has an industrialized and diversified economy. The study area covers a common mix of urban land uses, including residential, commercial, agricultural, and industrial areas. Nanping and Sanming in the river's headwaters and mid-section and Fuzhou in the lower section are three major settlements along the river that discharge domestic sewage and agricultural and industrial effluents without any treatment. Thus, water quality has been a concern in the Min River Basin. A previous study reported that Cu, Pb, Cd, Ni, Cr, Hg, and As have been detected in clam soft tissues (Song and Huang, 1991).

2.2 Sampling and analytical methods

Clam samples were collected from nine locations along the Lower Min River in different land use areas surrounding the river. The most common land uses were 1) established commercial and residential areas (ECRA), where sampling sites 7 and 8 were located; 2) industrial and agricultural areas (IAA), where sampling sites 1, 4, 5, 6, and 9 were located; 3) a developing industrial and residential area (DIRA), where sampling site 3 was located; and 4) a developing residential area (DRA), where sampling site 2 was located. Sampling locations in the Lower Min River are shown in Fig. 1.

Sampling was performed from January to March 2015. About 400 Asiatic clams, having diameters of ~10 mm, were collected at each site with a net during ebbed tide to obtain a representative sample for each site. Samples were transported and stored at 4°C until processed in the laboratory. The weights (wt(s)) and lengths of the clams and total wts of the collections were measured with the wet wt ranging from 0.57 to 3.04 g (wet wt, including valves) and the shell lengths from 10–20 mm. All samples were kept away from metallic materials to avoid contamination during sample handling.

Clam samples were cleaned, rinsed with Milli-Q water to remove any remaining sand and other particles, dissected into shell and soft tissues with a clean scalpel blade, stored in sterile polythene bags, and refrigerated at −20°C.

Soft tissues (wet wt) of clams were analyzed for Zn, Fe, Cr, Ni, Cd, Mn, As, Cu, and Pb, which might pose a risk to consumers (Bruins et al., 2000; Nkpaa et al., 2016). Approximately 1.0 g of soft tissues was used for digestion, which involved adding 6 mL of HNO_3 (65%) and 2 mL of

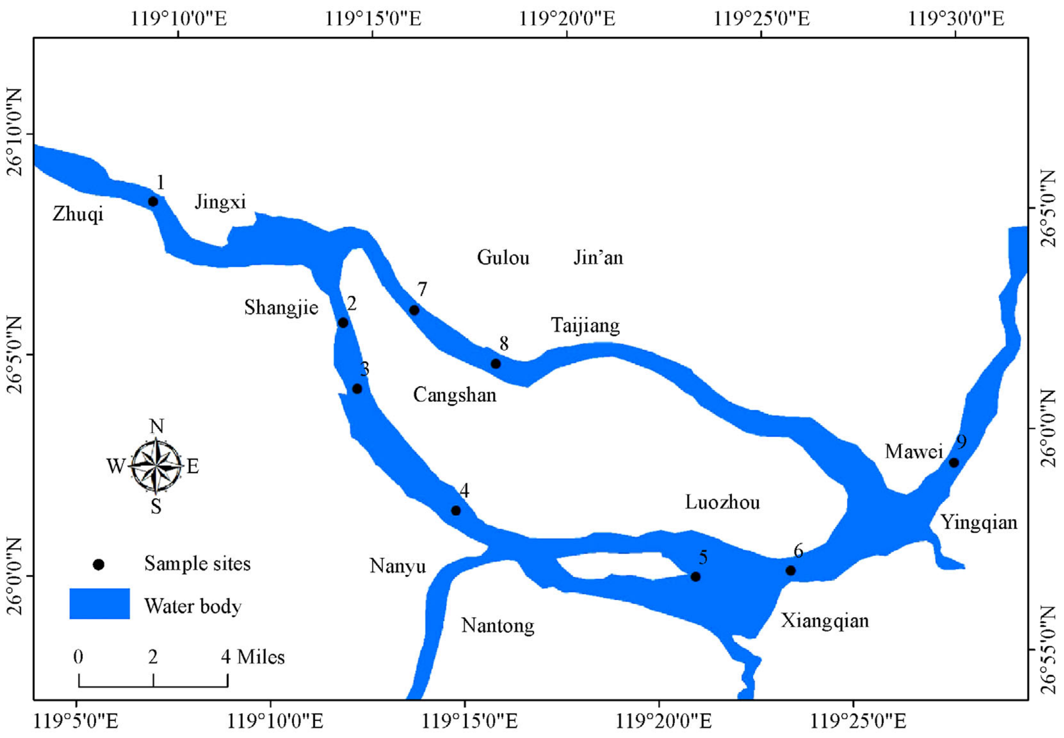

Fig. 1 Sample site locations along the Lower Min River, Southeast China.

H_2O_2 (30%) to samples prior to digestion in a microwave digestion oven. Microwave digestion consisted of three steps: 5.5 MPa and 120°C for 3 min, 120°C–160°C for 3 min, and 160°C–190°C for 25 min. After cooling for at least 1 h, digested clam samples were heated to 140°C using an electric heating plate, until almost all HNO_3 was evaporated and then diluted to 50 mL with Milli-Q water. The clam soft tissues were dried in a drying oven at 80°C until constant wt and recorded for dry/wet wt ratio determination. Digestion of a blank control was performed in the same manner.

Metal concentrations were determined by inductively coupled plasma optical emission spectrometry (Optima 7000 DV, Perkin Elmer, Inc., Waltham, MA, USA) with an analytical precision of 0.34%–2.36%. Analytical blanks were included in the analyses.

2.3 Pollution evaluation of metals in Asiatic clam

The metal pollution index (MPI) was adopted to compare the total content of metals at different sampling sites (Usero et al., 1997). MPI was obtained using Eq. (1)

$$\mathrm{MPI} = (C_1 \times C_2,\ldots,C_n)^{\frac{1}{n}}, \tag{1}$$

where C_1 is the concentration of the first metal; C_2 the concentration of the second metal; C_n the concentration of metal n; and n the total number of investigated metals in the sample.

2.4 Health risk assessment model

The CR and non-CR values for each metal at each location were determined to better estimate the risks to human health from consumption of these clams (US Environmental Protection Agency [USEPA], 1991). In this study, direct oral ingestion was the only exposure route considered and the exposure dose for carcinogenic effects considered as a lifetime exposure for adults.

The dose absorbed through ingestion was calculated using Eq. (2) modified from the USEPA (1991)

$$\mathrm{CDI}_{\text{ingestion}} = \frac{C_i \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}, \tag{2}$$

where $\mathrm{CDI}_{\text{ingestion}}$ is the average daily dose by ingestion, in mg/kg/d; C_i the average metal concentration in clam tissue, in mg/kg; IR the ingestion rate, in g/d; EF the exposure frequency, in d/yr; ED the exposure duration, in yr; BW the body wt, in kg; and AT the average time, in d.

The CR was evaluated using Eq. (3). It represented the probability of an individual developing cancer over a lifetime as a result of exposure to a carcinogen. In general, a CR that is acceptable or tolerable according the USEPA lies within the range of 10^{-6} to 10^{-4}. A CR lower than 1×10^{-6} can be regarded negligible and CR higher than 1×10^{-4} regarded as harmful to human beings, using

$$\mathrm{CR} = \mathrm{CDI}_{\text{ingestion}} \times \mathrm{SF}, \tag{3}$$

where CR is the carcinogenic risk and SF the slope factor that causes cancer.

The HQ can be used to indicate non-CR and is defined by the ratio of CDI to the oral reference dose (RfD_i) for the metal i. HQ is calculated using Eq. (4) (USEPA, 1991). When $HQ \leqslant 1$, it is considered that there will be no adverse health effects, while when $HQ > 1$, adverse health effects will be likely to occur. The hazard index (HI) was used here, as the sum of the HQs of total metals (Eq. (5)), to evaluate the cumulative health risks. $HI > 10^{-6}$ indicates the potential for adverse effects on human health, using

$$\mathrm{HQ}_i = \frac{\mathrm{CDI}_{\text{ingestion}} \times 10^{-6}}{\mathrm{RfD}_i}, \tag{4}$$

$$\mathrm{HI} = \sum \mathrm{HQ}_i, \tag{5}$$

where RfD_i is the oral reference dose, in μg/kg/d.

2.5 Statistical analysis

Correlation analysis was used to assess viable relationships among metal concentrations in these clams. Principal component analysis (PCA) was conducted to cluster metals that behaved similarly for identifying potential pollution sources. PCA with varimax rotation was conducted to reduce the data with a high loading on each component that could facilitate the interpretation of results (Acosta et al., 2010). In this study, Zn, Fe, Cr, Ni, Cd, Mn, As, Cu, and Pb in clam soft tissues were analyzed by PCA using SPSS Statistics 17.0 for Windows.

3 Results and discussion

3.1 Metal concentrations in Asiatic clams from the Lower Min River

Characteristics of metals in clam soft tissues from along the Lower Min River showed that, in general, the mean metal concentrations were, in decreasing order, Zn > Fe > Cr > Mn > Pb > Cu > As > Ni ≈ Cd (Table 1). The most abundant metals were Zn, Fe, and Cr (184, 112, and 42.1 mg/kg, respectively), whereas the least abundant metals were Ni and Cd (0.55 mg/kg). Most metals, particularly Zn, Mn, Fe, and Pb, exhibited wide variation as evidenced by large standard deviations and indicative of chemical-physical properties and anthropogenic addition at the sampling sites (Table 1).

When compared with the Chinese Marine Biological Quality Standards (CSOA, 2001; Table 1), the maximum concentrations of Cd, As, and Cu were lower than guideline values and the average concentrations of Cr, Pb, and Zn were 21.0, 4.0, and 3.7 times higher than the permissible limits, respectively. Even the minimum concentrations detected in this study were higher than guideline values. These results indicated that pollutant influences on the aquatic environment were serious.

The metal concentrations in clams from different rivers are listed in Table 2. The mean Cd and Ni concentrations in the present clam samples were comparable to those measured in Taihu Lake and the Yangtze River. The mean Cu concentration in the present clam samples was lower than measured in the Yangtze River and similar to that measured in Taihu Lake. The mean Fe and Mn concentrations were comparable to those measured in the US. However, the mean Cr, Pb, Zn, and As concentrations in clams from the Lower Min River were higher than the concentrations reported in clams from other areas inside or outside China. The high observed concentrations of these metals indicated a strong association with pollution from significant anthropogenic activities. Therefore, these results indicated that further investigation is needed to identify the sources of metal pollution in the Lower Min River regions.

Compared with a previous study of the same sampling sites (Fe, Zn, and Mn are unavailable; Table 3; Song and Huang, 1991), the mean levels of Cr, As, and Pb in the present study were 43.4, 2.3, and 1.8 times higher, respectively, than those measured three decades ago. Elevated concentrations of Cr, As, and Pb indicated significant anthropogenic disturbances. The mean Cd concentration in this study was also observed to be approximately two times lower than that of Song's study. As fertilizer application can be a major source of Cd (Li et al., 2001), increasing urbanization and industrialization and subsequent reduction of agricultural land in the Lower Min River in recent decades might explain why mean Cd concentration has decreased while concentrations of Cr, As, and Pb increased.

Table 1 Metal concentrations (mg/kg wet wt) in Asiatic clam tissues along the Lower Min River

Item	Fe	Zn	Cd	Mn	As	Cu	Ni	Pb	Cr
Min	66.9	51.5	0.25	3.93	0.71	3.51	0.37	2.03	31.2
Max	154	837	1.33	17.0	2.02	7.03	1.11	30.5	49.3
Mean	112	184	0.55	9.28	1.54	5.79	0.55	7.95	42.1
SE	31.1	249	0.33	38.8	0.45	1.11	0.25	9.31	5.73
Guideline[a)]	–[b)]	50	2.0	–	5.0	25	–	2	2

a) Guideline: Chinese Marine Biological Quality Standard (Chinese State Oceanic Administration, 2001); b) –: unavailable.

Table 2 Metal concentrations (mg/kg, dry wt) in Asiatic clams in different areas

Study area	Concentrations of metals/($\mathrm{mg\cdot kg^{-1}}$, dry wt)									Reference
	Fe	Zn	Cd	Mn	As	Cu	Ni	Pb	Cr	
This study[a]	334–757 (563)[b]	258–4.18×10^{3} (918)	1.25–6.65 (2.75)	19.6–85.1 (46.4)	3.55–10.1 (7.4)	17.6–35.2 (28.95)	1.85–5.55 (2.75)	10.2–153 (39.75)	156–246 (210)	
Georgia, USA	– c)	131	2.55	51.8	4.75	70	3.9	0.55	–	Shoults-Wilson et al. (2010)
Taihu Lake, China	–	79.3–291	2.8–16.8	–	–	23.3–68.4	3.8–20.2	1.5–12.3	nd[d]	Fan et al. (2014)
Yangtze River, China	–	143–209	2.13–7.79	–	–	40.0–93.2	–	0.98–3.28	1.35–2.55	Sun and Wang (2004)
Plata Estuary, Argentina	–	118–316	0.5–1.9	–	–	28–89	1.3–6.4	–	0.5–1.9	Bilos et al. (1998)
Northwest Iberian Peninsula	–	136–161	1.1–2.3	–	–	34–71	5.8–11	0.45–1.3	1.0–1.8	Reis et al. (2014)
Shatt al-Arab River, Iraq	–	31–83	2.2–70	–	–	40–1065	4.7	–	–	Abaychi and Mustafa (1988)
Cazaux-Sanguinet Lake, France	–	20–29	0.15–4.2	–	–	–	–	–	–	Marie et al. (2006)
Georgia Piedmont Basin, USA	–	189–544	0.8–4.1	–	2.1–5.0	32.0–87.7	–	–	–	Peltier et al. (2008)
Florida and North Carolina, USA	225–1900	55–115	0.7–1.95	10–85	nd	28.5–100	–	nd–12.5	–	Lewbart et al. (2010)

a) Unit conversion of metal concentrations of clams in this study from wet wt to dry wt was 5/1; b) numbers in parentheses denote average metal concentrations; c) –: unavailable; d) nd: not detected.

Table 3 Metal ion concentrations (mg/kg wet wt) in Asiatic clams in 1987 and 2015

Time	Sampling site	Fe	Zn	Cd	Mn	As	Cu	Ni	Pb	Cr
2015	1	151	98.9	0.5	8.31	1.12	6.39	0.78	5.69	40.5
1987		–[a]	–	0.69	–	0.47	5.21	0.54	1.61	0.28
2015	4	92.7	51.5	0.42	10.8	1.58	5.28	0.38	3.03	49.3
1987		–	–	0.93	–	0.74	7.35	0.72	3.41	1.48
2015	7	154	101	0.34	9.52	1.95	7.03	0.38	2.06	48.5
1987		–	–	0.95	–	0.63	7.09	0.73	3.71	1.45
2015	8	108	92.8	0.25	5.29	1.73	3.51	0.37	2.03	46.4
1987		–	–	0.88	–	0.6	7.16	0.59	3.77	1.26
2015	9	81.4	89.6	0.75	3.93	0.71	6.91	1.11	13.5	43.1
1987		–	–	0.68	–	0.622	5.24	0.53	2.47	0.8
2015	Mean			0.45		1.42	5.82	0.60	5.26	45.6
1987				0.83		0.61	6.41	0.62	2.99	1.05

a) –: unavailable.

3.2 Spatial distribution characterization of metals

The MPI values in different land use areas were investigated to identify hot spots and potential sources. The spatial distribution of metals showed that the MPI values in different surrounding land-use areas fell into the following order: DIRA (10.17) > DRA (8) > IAA (6.94) > ECRA (5.94, Fig. 2). This finding indicated the strong influence of construction activities in development areas despite the short history of land use. The lowest MPI occurred in ECRA, which might have been explained by decreasing sewage discharges with the improvement of urban sewage treatment facilities.

The spatial distribution trends of Cd, Cu, and Pb were generally similar despite of different concentrations among these metals, which indicate that these metals might come from the same source (Fig. 2). The highest Cd and Pb concentrations were observed to the west of the South Branch and the highest Cu concentration observed to the west of the North Branch. Cd, Cu, and Pb hot spots were

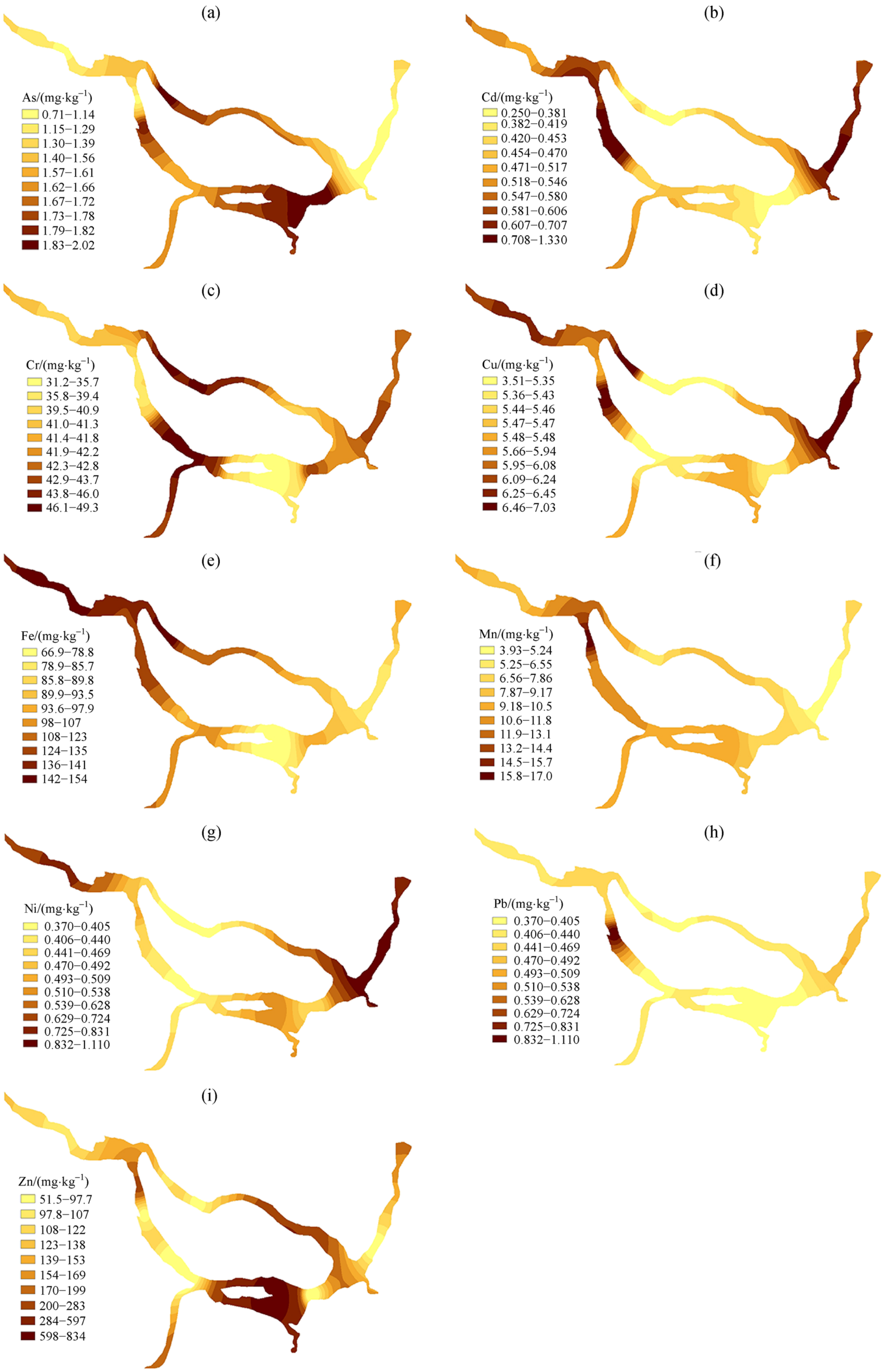

Fig. 2 Spatial distribution of metals in Asiatic clams along the Lower Min River. (a) As; (b) Cd; (c) Cr; (d) Cu; (e) Fe; (f) Mn; (g) Ni; (h) Pb; (i) Zn.

associated with areas that might have been influenced by mixed factors, such as ongoing construction, heavy traffic, tide, and industrial activities, which might have emitted Cd, Cu, and Pb.

Zn had a different distribution pattern from those of Cd, Pb, and Cu, with Zn hot spots mainly observed to the west of the South Branch along the Lower Min River. The highest Zn concentrations in the area might have been caused by wastewater discharges by processing and manufacturing activities. The As concentration was less variable, except in the spot with the highest level. Arsenic hot spots were mainly observed to the east of the South Branch along the Lower Min River. High As concentrations in the area might have been caused by automotive industrial activities. The highest Cr concentration was observed in the center of the South Branch, which might have been associated with manufacturing industries. Fe and Mn hot spots were detected upstream of the study area, which were influenced by the runoff from upstream areas.

The spatial distribution pattern of Ni was different from those of the other metals in this study. It was less variable, indicating that anthropogenic impact on Ni concentration in clams might have been slight in the study sites.

The distribution patterns of metals might have been affected by various pollution sources and natural processes such as weathering, runoff from upstream areas, tide, urban construction, and agricultural and industrial practices in the neighborhood. Thus, a more detailed statistical analysis is required to identify the contamination sources.

3.3 Metal pollution source analysis

Correlation analysis indicated a strong correlation between Cd and Pb ($p < 0.01$), which might have shared common sources. Some of the metals also exhibited inverse relationships, in which As was negatively correlated with Ni ($p < 0.01$) and Cr negatively correlated with Zn ($p < 0.05$), indicating their opposing distribution in clam samples (Table 4).

Multivariate PCA yielded three principal components (PCs) with eigenvalues > 1, which explained more than 77% of the data variance, in which PC_1, PC_2, and PC_3 contributed 32.25%, 23.19%, and 21.63%, respectively, to the total variance (Table 5). PC_1 exhibited elevated loadings of Cd, Pb, and Cu, of which Cd and Pb might have shared a common source because of their strong correlation, which was consistent with the discussion in Section 3.2 regarding the spatial distribution of metals. Cadmium and Pb were assumed to be contributed by transportation activities and urban and industrial waste (Romic and Romic, 2003; Peltier et al., 2008; Li et al., 2009; Acosta et al., 2010; Sun et al., 2013). Cu concentrations could have been influenced by agricultural activities and the paint and metal industries (Acosta et al., 2010; Sun et al., 2013). These results indicated that metals in PC_1 might have been influenced only by anthropogenic activities. PC_2 indicated higher contributions of As and Mn. Arsenic was assumed to be primarily from agricultural and industrial activities, such as mining, metal smelting and refining, and manufacturing processes, automobile exhaust, and energy and fertilizer production (Peltier et al., 2008; Li and Zhang, 2010). Spatial distribution analysis showed that As was strongly influenced by the automotive industry, which was consistent with As concentration results, which were significantly higher than those of 30 years ago. Mn is abundant in the earth and assumed to originate from natural contributions (Romic and Romic, 2003; Qishlaqi et al., 2009). The results indicated that PC_2 might have represented geologic as well as anthropogenic contributions. PC_3 exhibited the major contributions of Cr and Fe. Cr was considered to be contributed by mixed inputs of anthropogenic activities, such as industrial wastes, while natural sources were the contributors of the lithogenic components Fe and Mn (Qishlaqi et al., 2009). Meanwhile, anthropogenic inputs were more important than natural inputs in that the Cr concentration was significantly higher in this study than that of 30 years ago. The observed Fe concentrations could have resulted from natural sources, including weathering of parent material and subsequent pedogenesis (Romic and Romic, 2003;

Table 4 Pearson correlation coefficients (r) of metals in Asian clams

Metal	Fe	Zn	Cd	Mn	As	Cu	Ni	Pb	Cr
Fe	1.000								
Zn	–0.477	1.000							
Cd	0.139	–0.123	1.000						
Mn	0.348	0.198	0.250	1.000					
As	0.005	0.142	–0.204	0.042	1.000				
Cu	0.329	–0.104	0.517	0.075	–0.258	1.000			
Ni	–0.180	–0.038	0.236	–0.423	–0.833**	0.471	1.000		
Pb	0.146	–0.172	0.991**[b)]	0.197	–0.159	0.454	0.202	1.000	
Cr	0.243	–0.773*[a)]	–0.332	–0.336	0.106	–0.082	–0.184	–0.297	1.000

a)*: Significant at 0.05 probability level; b)**: significant at 0.01 probability level.

Table 5 Component matrix of metals in Asian clams

Item	Component		
	1	2	3
Fe	0.402	0.313	0.615
Zn	−0.117	0.13	−0.933
Cd	0.926	−0.078	−0.059
Mn	0.456	0.618	−0.189
Cu	0.669	−0.309	0.143
As	−0.263	0.77	0.007
Ni	0.229	−0.954	−0.099
Pb	0.896	−0.059	−0.019
Cr	−0.362	−0.025	0.875
Eigenvalue	2.745	2.106	2.085
% Total variance	30.504	23.396	23.162
Cumulative % variance	30.504	53.900	77.062

Qishlaqi et al., 2009). These results indicated that PC_3 might have had both geologic and anthropogenic contributions.

3.4 Health risk assessment of metals in clams

The values for the health risk assessment model originated from the USEPA (1991, 1999, 2015), except for IR and ED from the Fujian Province Bureau of Statistics (FPBS, 2014) and BW from the General Administration of Sports of China (DMSGASC, 2011), to localize the parameters. The HI, HQ, and CR values of each metal are presented in Table 6. Notably, the CR of As is from 3.25×10^{-4} to 5.45×10^{-4}, whereas the CR range acceptable or tolerable is 10^{-6} to 10^{-4} (USEPA, 1991). The As in clams from sampling sites posed some CR to the residents, which would cause cancers and other adverse health effects in humans with long-term exposure (Li and Zhang, 2010). The highest risk areas were ECRA and DIRA.

In this study, the mean levels of the HQs of metals ranged from 1.13×10^{-8} to 6.35×10^{-6}. The mean levels of the HQs were observed, in decreasing order, as Cr > As > Pb > Cd > Zn > Fe > Cu > Mn > Ni. These results showed that Cr, As, and Pb were the major contributors to ingestion exposure of residents, where Mn and Ni were only minor contributors. The HQs of Fe, Zn, Cd, Mn, Cu, and Ni were less than 10^{-6}, indicating that these elements posed no threat to residents. However, the HQs of Cr, As, and Pb in several sampling sites were greater than 10^{-6}, which indicated that they posed slightly adverse potential noncarcinogenic health risks to inhabitants. The HI values in different surrounding land-use areas were observed in the order of DIRA > ECRA ≈ IAA > DRA, which indicated that DIRA was the highest non-CR area. The HIs of all sampling sites were ~9.49×10^{-6} to 1.36×10^{-5}, which were greater than 10^{-6}. This finding indicated possible adverse health effects on residents, if exposed in large quantities. Compared with other references, the present findings showed more adverse health effects of metals than those in other areas (Karouna-Renier et al., 2007; Lewbart et al., 2010; Hussein and Khaled, 2014; Idriss and Ahmad, 2015). Therefore, consuming Asiatic clam from this river might present significant health risks for the local population.

The risk evaluation showed that As was the most important pollutant regarding the CR, different from the pollution evaluation results and guideline values (Table 1). Although the As concentration was lower than the national standard, it would still present potential CR for adults when excess As was consumed. At the same time, the largest contributors to non-CR were Cr, As, and Pb. Meanwhile, DIRA was the highest risk areas along the Lower Min River, similar to the result of the MPI. At the same time, ECRA should not be ignored for its low MPI because several metals had high health risks. The present findings indicated the unsuitability of these clams for edible purposes. The human consumption of clams from these polluted areas is thus not recommended. In addition, special attention should be focused on investigating these pollution sources, particularly in special function areas, and measures should be taken to sustain and promote healthy aquatic ecosystems.

Table 6 Reference dose, CR, and HQ for each metal in Asiatic clams along the Lower Min River, China

Site	HQ									HI	CR
	Fe	Zn	Cd	Mn	As	Cu	Ni	Pb	Cr		As
RfD_i/SF (μg/kg/d)/(μg/kg/d)$^{-1}$	0.7	0.3	0.001	0.14	0.0003	0.04	0.02	0.00357	0.003		1.5
ECRA	8.62E-08	1.49E-07	1.36E-07	2.44E-08	2.82E-06	6.06E-08	8.63E-09	2.64E-07	7.28E-06	1.08E-05	5.45E-04
IAA	8.80E-08	3.31E-07	2.76E-07	5.60E-08	1.68E-06	6.32E-08	1.16E-08	1.16E-06	5.82E-06	9.49E-06	3.25E-04
DRA	6.42E-08	3.53E-07	2.21E-07	2.64E-08	2.22E-06	6.77E-08	1.49E-08	7.19E-07	6.36E-06	1.00E-05	4.28E-04
DIRA	8.52E-08	1.41E-07	6.12E-07	3.81E-08	2.80E-06	7.70E-08	1.02E-08	3.94E-06	5.93E-06	1.36E-05	5.40E-04
Mean	8.09E-08	2.44E-07	3.11E-07	3.62E-08	2.38E-06	6.72E-08	1.13E-08	1.52E-06	6.35E-06	1.10E-05	4.59E-04

4 Conclusions

The metal distribution, source identifications, and health risk assessments via Asiatic clams from the Lower Min River were described in this study. Zn, Fe, and Cr were the most abundant metals in these clams, whereas Ni and Cd were the least abundant. Compared with guidelines established by the Chinese government, greater attention should be focused on Cr, Pb, and Zn. MPI assessment of the metals spatial distributions indicated the polluted river sections, which mainly concentrated in areas of DIRA.

Multivariate PCA and spatial distribution analysis revealed significant anthropogenic pollution of Cd, Pb, Cu, As, and Cr in the study areas and the major pollution sources included industrial wastes, transportation sources, and agricultural activities. Meanwhile, Mn and Fe were found to originate from natural sources.

From the human health perspective, non-CR and CR values showed that there is a slight risk in the clams in the study area. Arsenic was the most important pollutant causing noncarcinogenic and carcinogenic concerns and Cr, As, and Pb the major contributors to chronic risks. Thus, the use of clams from the study area in the human diet should be avoided.

Furthermore, other contaminants such as Persistent Organic Pollutants of health concern should be evaluated in addition to metal studies. Meanwhile, there also existed uncertainties in the present risk assessment, such as that exposure parameters used in this research were from other countries, which might not be suitable for the study region. It is necessary to carry out the epidemiologic surveys to assess real health impacts of metals on exposed populations (Kelepertzis, 2014). Therefore, further precise risk assessment approaches might be modified through investigations of local areas.

Acknowledgements The research was funded by the Natural Science Foundation of Fujian Province (Nos. 2018J01745 and 2013J01045), the China Scholarship Council (No. 201406655031), and the Science and Technology Development Foundation of Fuzhou University (No. 2013-XQ-19). We would like to thank Dr. Xiaoxu Lin and Dr. Jinliang Huang for their comments and suggestions.

References

Abaychi J K, Mustafa Y Z (1988). The Asiatic clam, *Corbicula fluminea*: an indicator of trace metal pollution in the Shatt al-Arab River, Iraq. Environ Pollut, 54(2): 109–122

Acosta J A, Faz A, Martinez-Martinez S (2010). Identification of heavy metal sources by multivariable analysis in a typical Mediterranean city (SE Spain). Environ Monit Assess, 169(1–4): 519–530

Arini A, Daffe G, Gonzalez P, Feurtet-Mazel A, Baudrimont M (2014). What are the outcomes of an industrial remediation on a metal-impacted hydrosystem? A 2-year field biomonitoring of the filter-feeding bivalve *Corbicula fluminea*. Chemosphere, 108: 214–224

Bilos C, Colombo J C, Presa M J (1998). Trace metals in suspended particles, sediments and Asiatic clams (*Corbicula fluminea*) of the Río de la Plata Estuary, Argentina. Environ Pollut, 99(1): 1–11

Bruins M R, Kapil S, Oehme F W (2000). Microbial resistance to metals in the environment. Ecotoxicol Environ Saf, 45(3): 198–207

Cairns J, Pratt J R (1993). A history of biological monitoring using benthic macroinvertebrates. In: Rosenberg D M, Resh V H, eds. Freshwater Biomonitoring and Benthic Macroinvertebrates. New York: Chapman and Hall, 10–27

CSOA (Chinese State Oceanic Administration) (2001). Marine Biological Quality Standard (GB 18421–2001) (in Chinese)

DMSGASC (Department of Mass Sports, of General State General Administration of Sport of China) of sports groups (2011). Communique on monitoring national constitutional condition in 2010. Beijing: People's Sport Publishing Press (in Chinese)

Fan W, Ren J, Wu C, Tan C, Wang X, Cui M, Wu K, Li X (2014). Using enriched stable isotope technique to study Cu bioaccumulation and bioavailability in *Corbicula fluminea* from Taihu Lake, China. Environ Sci Pollut Res Int, 21(24): 14069–14077

Farrington J W, Goldberg E D, Risebrough R W, Martin J H, Bowen V T (1983). U.S. "Mussel Watch" 1976 – 1978: an overview of the trace-metal, DDE, PCB, hydrocarbon, and artificial radionuclide data. Environ Sci Technol, 17(8): 490–496

Förstner U, Wittmann G T W (1979). Metal Pollution in the Aquatic Environment. Berlin, Heidelberg, New York, Tokyo: Springer-Verlag, 1–2

FPBS (Fujian Province Bureau of Statistics) (2014). Fujian Statistical Yearbook 2014. China Statistics Press (in Chinese)

Hussein A, Khaled A (2014). Determination of metals in tuna species and bivalves from Alexandria, Egypt. The Egyptian Journal of Aquatic Research, 40(1): 9–17

Idriss A A, Ahmad A K (2015). Heavy metal concentrations in fishes from Juru River, estimation of the health risk. Bull Environ Contam Toxicol, 94(2): 204–208

Karouna-Renier N K, Snyder R A, Allison J G, Wagner M G, Ranga Rao K (2007). Accumulation of organic and inorganic contaminants in shellfish collected in estuarine waters near Pensacola, Florida: contamination profiles and risks to human consumers. Environ Pollut, 145(2): 474–488

Kelepertzis E (2014). Investigating the sources and potential health risks of environmental contaminants in the soils and drinking waters from the rural clusters in Thiva area (Greece). Ecotoxicol Environ Saf, 100: 258–265

Kong M, Hang X, Wang L M, Yin H B, Zhang Y M (2016). Accumulation and risk assessment of heavy metals in sediments and zoobenthos (*Bellamya aeruginosa* and *Corbicula fluminea*) from Lake Taihu. Water Sci Technol, 73(1): 203–214

Krishna A K, Satyanarayanan M, Govil P K (2009). Assessment of heavy metal pollution in water using multivariate statistical techniques in an industrial area: a case study from Patancheru, Medak District, Andhra Pradesh, India. J Hazard Mater, 167(1 – 3): 366–373

Leland H V, Scudder B C (1990). Trace elements in *Corbicula fluminea* from the San Joaquin River, California. Sci Total Environ, 97–98(7): 641–672

Lewbart G A, Christian L S, Harms C A, Van Wettere A J (2010). A comparison of heavy metal concentrations and health assessment in

Asian clams *Corbicula fluminea* from Florida and North Carolina. J Aquat Anim Health, 22(2): 73–77

Li J L, He M, Han W, Gu Y F (2009). Analysis and assessment on heavy metal sources in the coastal soils developed from alluvial deposits using multivariate statistical methods. J Hazard Mater, 164(2 – 3): 976–981

Li S, Poon C, Liu P S (2001). Heavy metal contamination of urban soils and street dusts in Hong Kong. Appl Geochem, 16(11 – 12): 1361–1368

Li S Y, Zhang Q F (2010). Risk assessment and seasonal variations of dissolved trace elements and heavy metals in the Upper Han River, China. J Hazard Mater, 181(1–3): 1051–1058

Liu X L, Zhang L B, You L P, Yu J B, Zhao J M, Li L Z, Wang Q, Li F, Li C H, Liu D Y, Wu H F (2011). Differential toxicological effects induced by mercury in gills from three pedigrees of Manila clam *Ruditapes philippinarum* by NMR-based metabolomics. Ecotoxicology, 20(1): 177–186

Marie V, Gonzalez P, Baudrimont M, Bourdineaud J P, Boudou A (2006). Metallothionein response to cadmium and zinc exposures compared in two freshwater bivalves, *Dreissena polymorpha* and *Corbicula fluminea.* Biometals, 19(4): 399–407

Marschner H (1995). Mineral Nutrition of Higher Plants. London: Academic Press, 3–4

Marsden I D, Rainbow P S (2004). Does the accumulation of trace metals in crustaceans affect their ecology—the amphipod example? J Exp Mar Biol Ecol, 300(1–2): 373–408

Morton B (1986). Corbicula in Asia—an updated synthesis. Am Malacol Bull, 2: 113–124

Nkpaa K W, Patrick-Iwuanyanwu K C, Wegwu M O, Essien E B (2016). Health risk assessment of hazardous metals for population via consumption of seafood from Ogoniland, River State, Nigeria: a case study of Kaa, B-Dere, and Bodo City. Environ Monit Assess, 188(1): 9

Nriagu J O (1996). A history of global metal pollution. Science, 272 (5259): 223–224

Peltier G L, Meyer J L, Jagoe C H, Hopkins W A (2008). Using trace element concentrations in *Corbicula fluminea* to identify potential sources of contamination in an urban river. Environ Pollut, 154(2): 283–290

Qishlaqi A, Moore F, Forghani G (2009). Characterization of metal pollution in soils under two landuse patterns in the Angouran region, NW Iran: a study based on multivariate data analysis. J Hazard Mater, 172(1): 374–384

Reis P A, Guilhermino L, Antunes C, Sousa R (2014). Assessment of ecological quality of the Minho estuary (Northwest Iberian Peninsula) based on metal concentrations in sediments and in *Corbicula fluminea.* Limnetica, 33: 161–173

Romic M, Romic D (2003). Heavy metals distribution in agricultural topsoils in urban area. Environmental Geology, 43(7): 795–805

Shoults-Wilson W A, Unrine J M, Rickard J, Black M C (2010). Comparison of metal concentrations in *Corbicula fluminea* and *Elliptio hopetonensis* in the Altamaha River system, Georgia, USA. Environ Toxicol Chem, 29: 2026–2033

Smaal A C (2002). European mussel cultivation along the Atlantic coast: production status, problems and perspectives. Hydrobiologia, 484 (1/3): 89–98

Song Y, Huang Y (1991). Heavy metal levels in clam (*Corbicula Fluminea*) from Minjiang River, Fuzhou area. Oceanol Limnol Sin, 22: 187–190

Sun C Y, Liu J S, Wang Y Y, Sun L Q, Yu H W (2013). Multivariate and geostatistical analyses of the spatial distribution and sources of heavy metals in agricultural soil in Dehui, Northeast china. Chemosphere, 92(5): 517–523

Sun P, Wang B (2004). Metal content and contamination assessment in *Corbicula fluminea* from the Yangtze River Estuary. Chin J Appl Environ Biol, 10: 79–83

Türkmen M, Türkmen A, Tepe Y, Ates A, Gokkus K (2008). Determination of metal contaminations in sea foods from Marmara, Aegean and Mediterranean seas: twelve fish species. Food Chem, 108(2): 794–800

USEPA (US Environmental Protection Agency) (1991). Risk Assessment Guidance for Superfund: Volume I Human Health Evaluation Manual (Part B, development of risk based preliminary remediation goals).Publication 9285.7-01B, Washington, DC: Office of Emergency and Remedial Response U.S. EPA

USEPA (US Environmental Protection Agency) (1999). Cancer Risk Coefficients for Environmental Exposure to Radionuclides. EPA/402/R-99/001. Office of Air and Radiation. Washington, DC

USEPA (US Environmental Protection Agency) (2015). Risk-Based Screening Table- Generic Tables. http://semspub.epa.gov/src/document/03/2218434, 2016-5-30

Usero J, Gonzάlez-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(3): 291–298

Wei X, Gao B, Wang P, Zhou H D, Lu J (2015). Pollution characteristics and health risk assessment of heavy metals in street dusts from different functional areas in Beijing, China. Ecotoxicol Environ Saf, 112: 186–192

Zhang H, Chen Y, Fan H, Yang M, Chang F, Niu J, Lei G, Zhang W (2007). Climatic background of modern *Corbicula fluminea* and the stable isotopes of shells from the representative areas in continental China. Marine Geology & Quaternary Geologhy, 27(3): 77–84 (in Chinese)