Research Article

Spatial Variations and Potential Risks of Heavy Metals in Seawater, Sediments, and Living Organisms in Jiuzhen Bay, China

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Received 18 October 2019; Revised 23 December 2019; Accepted 9 January 2020; Published 3 February 2020

Guest Editor: Yuhe He

Coastal waters are polluted by heavy metals to varying degrees, posing potential risks to marine ecology and human health. In May 2006, the pollution levels, sources, and ecological risks of heavy metals (Cu, Pb, Zn, Cd, Hg, and As) in seawater, surface sediments, and living organisms were studied in Jiuzhen Bay in Fujian, China. This study identified Hg (0.26–0.72 µg/L) and As (20.3–31.5 µg/L) pollution in seawater of Jiuzhen Bay. In sediments, heavy Pb pollution (946 µg/g dw) was only detected at one station at a level posing very serious potential risk, while Hg pollution (0.052–0.087 µg/g dw) was observed at three stations at a level posing serious potential risk. No heavy metal pollution was detected in sediments at other stations. The concentrations of five heavy metals (Cu, Zn, As, Cd, and Pb) exceeded the corresponding National Quality Standards for oysters, indicating heavy pollution, based on an ecological risk assessment. In clams, two heavy metals (Pb and As) exceeded the standards, indicating light pollution, based on an ecological risk assessment. No heavy metal pollution was found in fish or shrimps. The heavy metals in the seawater and sediments of Jiuzhen Bay are mainly derived from the river discharges of Luxi and Wujiang Rivers although sewage discharge along the coast of Jiuzhen Bay is another source of heavy metal pollution at some stations. Given the pollution of Pb, Hg, and As in seawater and sediments at some stations within the bay, the potential risks of Pb, Hg, and As in living organisms to both the marine ecology and human health deserve increased attention.

1. Introduction

Over the past 40 years, because of rapid industrialization and economic development in China, the problem of soil contamination by heavy metals has become increasingly serious [1], with large amounts of these soil pollutants being discharged into coastal and estuarine environments by rivers and other pathways [2, 3]. Compared with other pollutants, heavy metals are persistent and have toxic as well as bio-accumulation effects, which severely damage marine environments [4]. Therefore, heavy metal contamination has attracted much attention and become the focus of global concern in recent years. Estuaries and coastal regions (such as bays) are important components of coastal systems, receiving large amounts of heavy metals via riverine input [5–7].

Estuaries are zone of complex interaction between fluvial and marine ecosystems in which many critical environmental processes including sediment deposition, fresh water-salt water interaction, delta accretion, pollutant retention, and material-energy exchanges occur. Particularly, the mixing of continental river water and marine salt water usually leads to flocculation and accumulation processes of heavy metals [8]. Although metal-aquo chemistry is the main factor affecting the removal and transformation of heavy metals, sediment type also has a major effect on transport and accumulation of heavy metals. Therefore, the spatial patterns of heavy metals are
closely related to sediment types [9]. The spatial distributions of heavy metals are often consistent with those of fine-grained sediments [10]. Most heavy metals become part of the sediment through different biogeochemical processes, which trigger precipitation and deposition [11]. Heavy metals do not behave conservatively, and they are also affected by changing physicochemical conditions, such as salinity, pH, and redox conditions [12]. When sedimentary environments change, the heavy metals in the sediment may be released back into the water body through various processes of remobilization, producing secondary pollution within the environment [13]. Therefore, marine sediments are considered an important sink of heavy metals, especially in coastal areas.

Heavy metals in seawater and marine sediments can be directly absorbed by organisms and then accumulated and transformed within their bodies. Accumulation may be magnified via the food chain, thereby threatening the marine ecosystem and human health [14–16]. Zhao et al. [17] reported that some seafood from Xiangshan Bay might pose noncarcinogenic risks to both adults and children. Although consumption of most common types of seafood from Xiamen markets does not pose a noncarcinogenic risk, some types, such as yellowfin bream (Sparus latus), oyster, and the Red alga (nori; Porphyra tenera), could form a carcinogenic risk [18]. The levels of five metals in marine fish and shellfish from China are generally low, based on published data. However, some findings suggest that there are health risks from exposure to Cd and As in some shellfish [19]. Considering the pollution of heavy metals in seawater and sediments in coastal areas, the risk of heavy metal pollution in seafood deserves greater attention.

Jiuzhen Bay is located on the southern coast of Fujian, between Gulei and Liu’ao Peninsulas. It is a concave, semienclosed shallow bay, forming an estuary. Its coastline is 45.97 km in length, with a total embayed area of 69.64 km². The entire bay is occupied by an intertidal shoal, except for its tidal channel. The intertidal area accounts for about 80% of the bay area. The tidal inlet runs from the bay’s mouth to the Luxi River outlet [20]. Jiuzhen Bay has rich marine biological resources and is an important marine aquaculture base in Fujian. The main aquaculture species are shellfish. Along the coast of Jiuzhen Bay, there are multiple industries, such as wind power generation, shipbuilding, placer mining, seafood processing, and shipping. In addition, the nearby Gulei Port Economic Development Zone is one of the seven petrochemical bases in China. The pollutant discharges from these various industrial enterprises may be causing serious environmental impacts on Jiuzhen Bay.

Given the environmental and sedimentation characteristics of Jiuzhen Bay, most of the heavy metals entering Jiuzhen Bay accumulate within the bay. To date, there has been no research on the distribution and ecological risk of heavy metals in seawater, sediments, or seafood of Jiuzhen Bay. The purpose of this study was to (1) determine the contents and distributions of various heavy metals in seawater, sediments, and living organisms within the bay; (2) analyze the sources of these heavy metals; and (3) evaluate the degree of heavy metal pollution, using the single-factor pollution index, comprehensive index evaluation, and potential ecological risk index.

The data provided here will assist the local government to monitor the change in the heavy metal pollution status of this area and implement targeted control measures.

2. Materials and Methods

2.1. Sample Collection. A total of nine stations were investigated during the survey of May 2006, as shown in Figure 1. A GO-FLO water sampler (General Oceanics, Inc., Miami, FL, USA) was used for sample collection. Water only within the surface layer was collected at stations, where water depths were less than 5 m (A1, B1, and B2), while water samples from surface (Sur) and bottom (Bot) layers were collected at stations with water depths of 5–20 m (B3, C1, C2, and E1–E3). Water samples were preserved at −20°C for later analysis. Sediment samples were collected using a grab bucket-type bottom sampler; sediments from the 0–2 cm surface layer were placed in a polyethylene sealing bag and preserved at −20°C for later analysis.

Biological samples were collected by trawling, and strict criteria were used to select adults of local representative biological species for the study. Specimens were preserved at −20°C for later analysis.

2.2. Laboratory Analysis. Six heavy metals—Cu, Pb, Zn, Cd, Hg, and As—in seawater, sediments, and living organisms were analyzed in this study.

All water samples were filtered using a hybrid cellulose ester Millipore filter, which was dipped before filtering in acid. The filtrate was acidified to pH < 2 with HNO₃ and stored in a Teflon bottle.

Before analysis of the sediment samples, they were freeze-dried. All large debris was removed, followed by grinding and sieving. Dry sediment samples were dissolved in an equimolar mixture of HF, HNO₃, and HClO₄, having a volumetric ratio of 3:1:0.5, and then evaporated to dryness. The samples were redissolved with aqua regia, and three parts of this metal solution were diluted with HNO₃.

Edible parts of living organisms were removed using biomedical stainless-steel implements, smashed using a stamping machine, blended, and dried in an oven, allowing moisture content to be calculated. Dried biological samples were digested like sediments.

Cu, Pb, Zn, and Cd were analyzed using an Agilent 7500a inductively coupled plasma source mass spectrometer (Agilent Technologies, Inc., Santa Clara, CA, USA), while Hg and As were analyzed using an atomic fluorescent photometer AFS200T (Skyray Instrument, Inc., Stoughton, MA, USA).

Total nitrogen (TN), total phosphorus (TP), dissolved inorganic nitrogen (DIN), dissolved inorganic silicon (DSI), and PO₄ were measured using a spectrophotometric method, while total organic carbon (TOC), redox potential (Eh), and pH were analyzed using K₂Cr₂O₇ oxidation-reduction volumetric method and potentiometer and acid meter method, respectively. DIN is the sum of NO₃⁻, NO₂⁻, NO₃⁻.
and NH₄⁺, TN, TP, DIN, DSI, and PO₄ were analyzed using a flow injection analyzer (model: Lachat-QC8000). Redox potential (Eh) and pH were determined using an acid meter (model: pHS-3C).

2.3. Quality Control and Quality Assurance. Laboratory quality assurance and quality control methods were practiced, involving standard operating procedures, calibration using standards, and analysis of reagent blanks. All samples were analyzed in parallel at proportions of 10%. Quality control was ensured by measuring nationally certified standard reference materials (GBW07314, GBW080042, GBW080230, GBW080040, and GBW 10050). The differences in the concentrations between the certified and measured values were <5%. The recoveries of all metals were within the range of 85%–99%. The relative measurement error was less than 10%.

2.4. Data Analysis and Evaluation Methods. Pearson correlation analysis was used to quantify relationships among heavy metals. Data calculation and statistics were performed with SPSS 17.0 statistical software (IBM, Armonk, NY, USA) and Microsoft Excel (Microsoft Corp., Albuquerque, NM, USA). The figures in this paper were drawn with Surfer 8 (Scientific Software Group, Salt Lake City, UT, USA).

The single-factor index method was used to evaluate heavy metals in seawater, using the formula below:

\[ P_i = C_i \times S_{ij}, \]  

where \( P_i \) is the heavy metal pollution index, \( C_i \) is the concentration value of a given heavy metal, and \( S_{ij} \) is the standard value of a given heavy metal. The Type I Standard of the National Seawater Quality Standard (China) was used to obtain standard values of heavy metals in seawater, yielding values for Cu (5 μg/L), Zn (20 μg/L), As (20 μg/L), Cd (1 μg/L), Hg (0.05 μg/L), and Pb (1 μg/L).

Biotoxicity or biological risks of heavy metals in sediments were evaluated using the potential ecological risk index proposed by the Swedish scholar Hakanson [21], using the formula below:

\[ E'_r = T'_r \times C'_r = T'_r \times \frac{C_i}{C_n}, \]

\[ RI = \sum_{i=1}^{n} E'_r, \]

where \( T'_r \) is the toxicity response coefficient of a heavy metal and those of Hg, Cd, As, Cu, Pb, and Zn are, respectively, set as 40, 30, 10, 5, 5, and 1 [22, 23]; \( C'_r \) is the pollution index of a given heavy metal, also called its enrichment coefficient; \( C_i \) is the measured concentration of the given heavy metal in sediments; \( C_n \) is the background value of the given heavy metal; \( E'_r \) is the potential ecological risk index of the given heavy metal; and \( RI \) is the total potential ecological risk index related to multiple heavy metals. The degrees of ecological risk corresponding to \( E'_r \) and \( RI \) values are given in Table 1.

The single-factor evaluation method and comprehensive index evaluation method were combined to evaluate heavy metals in living organisms. In this case, the National Quality Standard was a bivalve, referred to as the Type I Standard in Marine Biological Quality (China) (GB18421-2001). Standard values for crustacea and fish are taken from previously determined values from monitoring and evaluation of heavy metal pollution elements in offshore areas [24].

The formula for the comprehensive index evaluation method is given below:

\[ P_{ij} = \sqrt{\left( \text{max} P_i \right)^2 + \left( \text{ave} P_i \right)^2}, \]

where \( P_{ij} \) is the comprehensive quality index, \( P_i \) is the single-factor pollution index, \( \text{max} P_i \) is the maximum value of the single-factor pollution index for a given living organism, and \( \text{ave} P_i \) is the average value of the single-factor pollution index of a given living organism.

The degree of heavy metal pollution in marine living organisms that correspond to values obtained from the comprehensive index method are given in Table 2.

In this paper, the bioconcentration factors (BCFs) and biota-sediment accumulation factors (BSAFs) were calculated to analyze the bioaccumulation of heavy metals from seawater and sediments in living organisms. The BCFs indicate the status of organisms enriched in heavy metals from the surrounding waters, while the BSAFs evaluate the equilibrium relationship of heavy metal contents in benthic organisms and sediments, i.e., the indirect absorption of heavy metals by benthic organisms from their living environments [25, 26].

The calculations are defined as follows:
Heavy metal contents in seawater were measured in Jiuzhen Bay. Distribution characteristics of heavy metals in seawater and their vertical distributions were examined, and the results showed that the mean concentration of the metal in the sediment is the horizontal distribution characteristic of heavy metals. Changes in environmental factors such as salinity, hydrodynamics, pH, redox, and biological effects related to heavy metal dissolutions in suspended particles were considered. Analysis of their vertical distributions show that the average values of heavy metals—Cd and Pb—in seawater from the surface layer were higher than those in the bottom layer, while other heavy metal contents (Cu, Zn, As, and Hg) in the surface layer were lower than those in the bottom layer. Zn concentrations in the bottom layer were twice those in the surface layer.

The overall horizontal distribution characteristics were consistent, both layers showing a decrease from the outer to inner parts of the bay, with highest values within the bay’s mouth and outer areas. There was a distinct difference in the distribution of Pb in the surface and bottom seawater layers. An area having a low Pb content occurred on the inner side of the mouth. The Pb content gradually increased in surrounding areas, resulting in a high value at station B1 on the west side of the bay. In the bottom seawater layer, high levels of Pb were detected at stations B3 and C2, while Pb gradually decreased from east to west. An overall decrease from the bay’s mouth to outer areas was observed for Cd in both the surface and bottom seawater layers, with the highest values within the mouth area. The distribution of Zn varied greatly in the surface and bottom seawater layers. Although Zn displayed a consistent trend, like those of Cu, As, and Hg in surface seawater, its concentration at station C2 in the bottom seawater layer was 123 µg/L, which was seven times that of surrounding stations. Distributions of all six heavy metals within Jiuzhen Bay are shown in Figure 2.

The contents of heavy metals in seawater measured in the present study were compared with historical data and studies of other regions in China (Table 4). In comparison with historical data [20], the contents of Cu and Cd have increased markedly. Except for the much higher contents of Hg and As, the heavy metal contents determined in the present study were basically consistent with those of the southern Gulf of China [13, 17, 27–29]. This comparison indicates that heavy metal content of the seawater of Jiaozhou Bay was the highest among all documented bays [30].

### 3. Results and Discussion

#### 3.1. Distribution Characteristics and Evaluation of Heavy Metal Pollution in Seawater

**3.1.1. Distribution Characteristics of Heavy Metals in Seawater.** Heavy metal contents in seawater were measured in samples from all stations within Jiuzhen Bay, as shown in Table 3. Analysis of their vertical distributions show that the average values of heavy metals—Cd and Pb—in seawater from the surface layer were higher than those in the bottom layer, while other heavy metal contents (Cu, Zn, As, and Hg) in the surface layer were lower than those in the bottom layer. Zn concentrations in the bottom layer were twice those in the surface layer.

The overall horizontal distribution characteristics were as follows. The distribution of heavy metals (Cu, As, and Hg) in the surface and bottom seawater layers were relatively consistent, both layers showing a decrease from the outer to inner parts of the bay, with highest values within the bay’s mouth and outer areas. There was a distinct difference in the distribution of Pb in the surface and bottom seawater layers. An area having a low Pb content occurred on the inner side of the mouth. The Pb content gradually increased in surrounding areas, resulting in a high value at station B1 on the west side of the bay. In the bottom seawater layer, high levels of Pb were detected at stations B3 and C2, while Pb gradually decreased from east to west. An overall decrease from the bay’s mouth to outer areas was observed for Cd in both the surface and bottom seawater layers, with the highest values within the mouth area. The distribution of Zn varied greatly in the surface and bottom seawater layers. Although Zn displayed a consistent trend, like those of Cu, As, and Hg in surface seawater, its concentration at station C2 in the bottom seawater layer was 123 µg/L, which was seven times that of surrounding stations. Distributions of all six heavy metals within Jiuzhen Bay are shown in Figure 2.

**3.1.2. Main Factors Affecting Dissolved Heavy Metal Distributions in Seawater.** According to the correlation analysis between the heavy metals in seawater and environmental factors (Table 5), significant correlations were determined between heavy metals (As, Hg, and Cu) and environmental factors (PO4, Si, and DIN) ($R > 0.605, P < 0.05, N = 15$), indicating certain similar sources of the substances. In fact, the sources of heavy metals (As, Hg, and Cu) in seawater are indeed similar to those of PO4, Si, and DIN, mainly being river water and domestic and industrial wastewater along the coast of Jiuzhen Bay. The distribution trend of heavy metals (As, Hg, and Cu) increased gradually from the mouth of the bay out to sea. The higher concentrations at the mouth were results from the changes in environmental factors such as salinity and redox factors. Specifically, the heavy metals adsorbed by suspended particles in the river water dissolved out, resulting in high contents of As, Hg, and Cu in the seawater at the mouth. This is consistent with the conclusions of Zhang et al. [31], in which study it was proposed that the content of heavy metals in the downstream of the Yellow River was lower than that at the estuary, and the content of dissolved heavy metals was closely related to the adsorption and desorption of heavy metals in suspended particles.

In the seawater of bays and estuaries, heavy metal content is affected by changes in environmental factors such as salinity, hydrodynamics, pH, redox, and biological effects [32, 33]. The investigation sea area is located at the mouth of
Jiuzhen bay, which is the mixing region of river water and seawater. Therefore, the environmental factors vary significantly, posing different degrees of impact on different heavy metals, which is the main reason for the poor correlation between heavy metals and environmental factors.

The maximum value of Pb content in surface seawater was located at station B1 of shallow water, which is consistent with the location of the high-value zone in the sediment, indicating that the concentration of Pb in seawater at station B1 was affected by the content of sediment. Upon changes of environmental conditions, the equilibrium between the overlying water and the sediment is broken, resulting in the migration of heavy metal pollutants from the sediment to the water body [12]. The high value area of Pb in bottom seawater was at the east side of the tidal channel, i.e., the Liu’ao Peninsula and the Liu’ao Port. The population on the Liu’ao Peninsula is mainly concentrated in Liu’ao Town. Large amount of sewage discharge from human domestic and industrial activities to the Jiuzhen Bay resulted in the high Pb content on the east side of the tidal channel. The maximum concentration of Zn detected at the C2 station in bottom seawater was also caused by the discharge of domestic and industrial sewage from Liu’ao Town.

The heavy metals in the seawater of the investigation area are mainly originated from the water of Luxi and Wujiang River, as well as the domestic and industrial wastewater along the coast of the Jiuzhen Bay. Moreover, the dissolution of heavy metals from sediments is another source of heavy metals in seawater.

### 3.1.3. Evaluation of Heavy Metal Pollution in Seawater

Values of the single-factor pollution indices of various heavy metals at each station are given in Table 6, and their respective ranges were Cu (0.67–1.27), Zn (0.45–6.13), As (1.02–1.58), Cd (0.24–0.50), Hg (5.29–14.33), and Pb (0.25–0.54). Typically, the single-factor pollution indices for both Hg and As in Jiuzhen Bay were greater than 1, indicating that both metals exceeded the Type I National Seawater Quality Standard at all stations. Cu and Zn exceeded the type I standard at some stations, at rates of 33% and 13%, respectively. Neither Cd or Pb exceeded the type I threshold. According to average values of single-factor pollution indices for heavy metals, their pollution degree was Hg > As > Zn > Cu > Pb > Cd.

### 3.2. Distribution Characteristics and Evaluation of Heavy Metal Pollution in Sediments

#### 3.2.1. Distribution Characteristics of Heavy Metals in Sediments

Average heavy metal contents of sediments within Jiuzhen Bay are given in Table 7. Pb had highest values in sediments along the northern coast and in the northwest part of the mouth of the bay; they progressively declined eastward. A high value area of all five heavy metals (Cu, Zn, Cd, Hg, and As) occurred on the northeast side of Jiuzhen Bay. Heavy metal distributions in sediments are shown in Figure 3.

The heavy metal contents in sediments measured in this present study were compared with historical data and studies of other regions (Table 8). Compared with historical data [20], Pb contents have increased markedly, while other heavy metal contents have remained unchanged. In this paper, the heavy metal contents in sediments were lower than those of other marine areas and bays in China [10, 17, 28, 36, 37], except for Pb content, which was much higher than in all other locations.

#### 3.2.2. Main Factors Affecting Heavy Metal Distributions in Sediments

According to our correlation analysis of heavy metals and environmental factors in the sediments (Table 9), a significant positive correlation was determined for Cu, Zn, Cd, Hg, and As ($R = 0.737; P < 0.05, N = 9$). This suggests these metals have the same mode of accumulation in surface sediments of Jiuzhen Bay. The main source of these heavy metals was land-based pollution. Regarding the effect of environmental factors, the correlation coefficients for total organic carbon (TOC), TP, total nitrogen (TN), and redox potential (Eh) with Cu, Zn, Cd, Hg, and As were all relatively high, showing strong correlations. The reproduction of plankton removes nutrients from fresh water inputs, forming a large amount of organic suspended matter, just like the natural large-scale ecological beds resulting from sewage treatment in Jiuzhen Bay [38]. The heavy metals combine with organic matter and remain suspended in water mixing zones water, before being precipitated. This may be the cause for the high value areas of all five heavy metals (Cu, Zn, Cd, Hg, and As) on both sides of the tidal channel, as well as the higher contents of heavy metals in sediments within the mouth area, compared with further offshore.

Based on a comparison of historical background values [20], the values of Pb in sediments were far higher than those of other heavy metals. In terms of their distributions, the correlations of Pb with other heavy metal elements were nonsignificant. For example, the Pb content at station B1 was tens of times more than those at other stations with serious pollution, indicating that increasing Pb content was related to pollution discharge from land areas on the west side of the bay.

### Table 3: Heavy metal contents in seawater layers of Jiuzhen Bay (μg/L).

<table>
<thead>
<tr>
<th>Heavy metal type</th>
<th>Total average value (range)</th>
<th>Average value at the surface layer (range)</th>
<th>Average value at the bottom layer (range)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>4.82 (3.37–6.33)</td>
<td>4.74 (3.37–5.88)</td>
<td>4.94 (4.05–6.33)</td>
</tr>
<tr>
<td>Zn</td>
<td>22.0 (8.9–123)</td>
<td>16.7 (11.5–29.2)</td>
<td>30.1 (8.9–123)</td>
</tr>
<tr>
<td>As</td>
<td>27.0 (20.3–31.5)</td>
<td>26.3 (20.3–29.8)</td>
<td>27.9 (25.0–31.5)</td>
</tr>
<tr>
<td>Cd</td>
<td>0.34 (0.24–0.50)</td>
<td>0.34 (0.24–0.50)</td>
<td>0.32 (0.29–0.35)</td>
</tr>
<tr>
<td>Hg</td>
<td>0.46 (0.26–0.72)</td>
<td>0.44 (0.26–0.72)</td>
<td>0.50 (0.39–0.67)</td>
</tr>
<tr>
<td>Pb</td>
<td>0.37 (0.25–0.54)</td>
<td>0.37 (0.25–0.45)</td>
<td>0.36 (0.25–0.54)</td>
</tr>
</tbody>
</table>

### Table 2: Heavy metal contents in seawater layers of Jiuzhen Bay (μg/L).

3.2. (Incorporating information from the table)

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Figure 2: Continued.
The contents of Hg and As in the sediments at Futou Bay are higher than those at Jiuzhen Bay [36], related to the polluted discharge from Gulei Peninsula. Evaluation of the seawater and sediments in Jiuzhen Bay shows that Hg pollution was heavier than other heavy metals, suggesting that Futou Bay may be another source of Hg.
3.2.3. Ecological Risk Evaluation of Heavy Metal Pollution in Sediments. Evaluation results of potential ecological risks of heavy metals in sediments in the superficial layer of Jiuzhen Bay are shown in Table 10. Ranges of the ecological risk index ($E_i$) for each heavy metal were Cu (0.54–10.66), Zn (0.17–2.43), As (2.44–5.91), Cd (3–25.97), Hg (5.8–139.5), and Pb (6.3–526.7), respectively. The $E_i$ values of Cu, Zn, As, and Cd in sediments were less than 40 at all stations, representing a low degree of potential ecological risk. In contrast, the maximum $E_i$ value for Pb in sediments was 526.7 (station B1), which indicates a very serious potential ecological risk. The $E_i$ values of Pb at all other stations were less than 40, representing a low degree of potential ecological risk. $E_i$ values of Hg at three stations—A1, B3, and C2—were 83.8–139.5, indicating a relatively high degree of potential ecological risk. However, the $E_i$ values of Hg at all other stations were less than 40, representing a low degree of potential ecological risk at other sites. According to the average $E_i$ values of heavy metals, potential ecological risks related to single heavy metals were ranked: Pb > Hg > Cd > Cu > As > Zn.

The overall range of the potential ecological risk index ($RI$) was 18.9–556.1 in the study area, with a maximum value of 556.1 at station B1, indicating a relatively serious potential ecological risk. The $RI$ values of stations A1 and B3 were 222.4 and 166.7, respectively, indicating a moderate potential ecological risk. The $RI$ values of all other stations were less than 150, representing low to moderate potential ecological risks. According to average $RI$ values of all stations, potential ecological risks linked to sediments at each station were ranked: B1 > A1 > B3 > C2 > B2 > C1 > E1 > E3 > E2.
Figure 3: Distributions of various heavy metals in sediments of Jiuzhen Bay (µg/g dw): (a) Cu; (b) Zn; (c) Cd; (d) Hg; (e) Pb; (f) As.
### 3.3. Heavy Metal Contents in Living Organisms and Their Associated Ecological Risks

#### 3.3.1. Heavy Metal Content Characteristics in Living Organisms

Average heavy metal contents of representative living organisms from the study area are shown in Table 11. Cu, Zn, and Cd contents were highly variable in different kinds of living organisms. Average Cu and Cd contents in oysters were 73.19 μg/g ww and 0.748 μg/g ww, respectively; these values were much higher than those in fish (0.23 μg/g ww and 0.001 μg/g ww). The Zn content was highest in oysters (234 μg/g ww), followed by those in shrimps (25.61 μg/g ww) and fish (5.47 μg/g ww). The content of Hg was highest in fish (0.043 μg/g ww) and lowest in clams (0.008 μg/g ww). The contents of Pb and As were highest in oysters, with average values of 0.361 μg/g ww and 3.14 μg/g ww, respectively. The lowest contents of Pb and As were detected in shrimps, with average values of 0.008 μg/g ww and 1.16 μg/g ww, respectively.

Enrichment degrees of most heavy metals in shellfish were higher than those in fish, as reported by many previous researchers [29, 39, 40]. In this study, the contents of five heavy metals (Cu, Zn, As, Cd, and Pb) in oysters were higher than those in all other living organisms. Two heavy metals (Cu and Zn) had high contents in shrimps, but these ranked second to their contents in oysters. The Hg contents in fish were higher than those in other living organisms.

#### 3.3.2. Ecological Risk Evaluation of Heavy Metal Pollution in Living Organisms

Evaluation results of the single-factor pollution index \( P_i \) and comprehensive pollution index \( P_{ij} \) of heavy metals in living organisms are shown in Table 12. The evaluation index \( P_i \) ranges of various heavy metals were Cu (0.01–7.32), Zn (0.14–11.70), As (0.15–3.14), Cd (0–3.74), Hg (0.08–0.50), and Pb (0.03–3.61). The single-factor pollution index of heavy metals varied significantly in different species of living organisms. Although the heavy metal contents in fish and shrimps were all below the National Quality Standards, those in mollusks greatly exceeded the standards. Pb and As contents in clams were above the threshold values of their standards, while all heavy metals (except Hg) in oysters exceeded the standards. The ranking of the pollution indices of heavy metals in oysters was Zn > Cu > Cd > Pb > As > Hg.

The comprehensive pollution index \( P_{ij} \) of heavy metals in living organisms ranged from 0.13 to 9.00. Oysters displayed the highest comprehensive pollution index, being classified as heavily polluted. This was followed by clams, which were classified as lightly polluted. Heavy metal contents of fish and shrimps were not considered polluted.

#### 3.3.3. Analysis of Enrichment of Different Heavy Metals in Living Organisms

According to the BCF and BSAF values of various organisms (Table 13), the rankings of enrichment coefficients of different heavy metals in seawater according to species were oysters: \( Cu > Zn > Cd > Pb > As > Hg \), clams: \( Pb > Zn > Cu > Cd > As > Hg \), fish: \( Pb > Zn > Hg > As > Cu > Cd \), and shrimps: \( Cu > Zn > Pb > As > Hg > Cd \). Likewise, the rankings of enrichment coefficients of different heavy metals in the sediments according to species were oysters: \( Cd > Cu > Zn > Hg > As > Pb \), clams: \( Cd > As > Hg > Zn > Cu > Pb \), fish: \( Hg > As > Zn > Cu > Cd > Pb \), and shrimps.
The ranking of heavy metal contents in living organisms was not consistent with that of seawater or sediments. This indicates that, at low concentrations, the enrichment of heavy metals is not only related to the concentrations of heavy metals in seawater and sediments but also to many other factors.

In oysters, clams, and shrimps, the enrichment coefficients of Cu, Zn, Cd, and Pb were higher than those of As and Hg. This is because Cu and Zn are essential elements of life; thus, they are actively absorbed by living organisms. Therefore, the ability of marine organisms to be enriched in Cu and Zn was much higher than for nonessential metal elements [41]. Notably, the excretion of Pb and Cd is very slow in most organisms. Because shellfish are filter feeders, and both Pb and Cd complexes in suspension and in the sediments were ingested by and enriched in shellfish [42], accounting for the high enrichment coefficients of Pb and Cd in oysters, clams, and shrimps.

According to Table 13, the enrichment coefficient of Hg was higher in fish than in shellfish. This is caused by the higher capacity of fish to absorb Hg through water and food intake, as well as physiological mechanisms that produce Hg enrichment within their bodies [43].

Studies have suggested that an enrichment coefficient greater than 1000 indicates a potential cumulative problem [25]. In the present study, an enrichment coefficient greater

<table>
<thead>
<tr>
<th>Living organism</th>
<th>Cu</th>
<th>Zn</th>
<th>As</th>
<th>Cd</th>
<th>Hg</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oyster</td>
<td>73.19</td>
<td>10636</td>
<td>116</td>
<td>2200</td>
<td>54</td>
<td>976</td>
</tr>
<tr>
<td>Clam</td>
<td>15185</td>
<td>490</td>
<td>89</td>
<td>91</td>
<td>17</td>
<td>511</td>
</tr>
<tr>
<td>Fish</td>
<td>48</td>
<td>249</td>
<td>89</td>
<td>3</td>
<td>93</td>
<td>262</td>
</tr>
<tr>
<td>Shrimp</td>
<td>1589</td>
<td>1164</td>
<td>43</td>
<td>15</td>
<td>33</td>
<td>170</td>
</tr>
</tbody>
</table>

### Table 12: Values of the pollution index ($P_i$) and comprehensive pollution index ($P_i^*$) for single-factor heavy metal pollution in various living organisms.

<table>
<thead>
<tr>
<th>Living organism</th>
<th>$P_{Cu}$</th>
<th>$P_{Zn}$</th>
<th>$P_{As}$</th>
<th>$P_{Cd}$</th>
<th>$P_{Hg}$</th>
<th>$P_{Pb}$</th>
<th>$P_{Ave}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oyster</td>
<td>7.32</td>
<td>11.70</td>
<td>3.14</td>
<td>3.74</td>
<td>0.50</td>
<td>3.61</td>
<td>5.00</td>
</tr>
<tr>
<td>Clam</td>
<td>0.15</td>
<td>0.54</td>
<td>2.41</td>
<td>0.16</td>
<td>0.16</td>
<td>1.89</td>
<td>0.88</td>
</tr>
<tr>
<td>Fish</td>
<td>0.01</td>
<td>0.14</td>
<td>0.48</td>
<td>—</td>
<td>0.14</td>
<td>0.05</td>
<td>0.14</td>
</tr>
<tr>
<td>Shrimp</td>
<td>0.08</td>
<td>0.17</td>
<td>0.15</td>
<td>—</td>
<td>0.08</td>
<td>0.03</td>
<td>0.08</td>
</tr>
</tbody>
</table>

### Table 13: Bioconcentration factors (BCFs) and biota-sediment accumulation factors (BSAFs) of various living organisms.

<table>
<thead>
<tr>
<th>Factor</th>
<th>Living organism</th>
<th>Cu</th>
<th>Zn</th>
<th>As</th>
<th>Cd</th>
<th>Hg</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>BCF</td>
<td>Oyster</td>
<td>15185</td>
<td>10636</td>
<td>116</td>
<td>2200</td>
<td>54</td>
<td>976</td>
</tr>
<tr>
<td></td>
<td>Clam</td>
<td>309</td>
<td>490</td>
<td>89</td>
<td>91</td>
<td>17</td>
<td>511</td>
</tr>
<tr>
<td></td>
<td>Fish</td>
<td>48</td>
<td>249</td>
<td>89</td>
<td>3</td>
<td>93</td>
<td>262</td>
</tr>
<tr>
<td></td>
<td>Shrimp</td>
<td>1589</td>
<td>1164</td>
<td>43</td>
<td>15</td>
<td>33</td>
<td>170</td>
</tr>
<tr>
<td>BSAF</td>
<td>Oyster</td>
<td>8.47</td>
<td>4.08</td>
<td>0.50</td>
<td>0.02</td>
<td>1.54</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Clam</td>
<td>0.17</td>
<td>0.19</td>
<td>0.38</td>
<td>0.02</td>
<td>1.54</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Fish</td>
<td>0.03</td>
<td>0.10</td>
<td>0.38</td>
<td>0.02</td>
<td>1.54</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Shrimp</td>
<td>0.89</td>
<td>0.45</td>
<td>0.18</td>
<td>0.08</td>
<td>0.54</td>
<td></td>
</tr>
</tbody>
</table>

Cu > Hg > Zn > As > Cd > Pb. The ranking of heavy metal contents in living organisms was not consistent with that of seawater or sediments. This indicates that, at low concentrations, the enrichment of heavy metals is not only related to the concentrations of heavy metals in seawater and sediments but also to many other factors.

In oysters, clams, and shrimps, the enrichment coefficients of Cu, Zn, Cd, and Pb were higher than those of As and Hg. This is because Cu and Zn are essential elements of life; thus, they are actively absorbed by living organisms. Therefore, the ability of marine organisms to be enriched in Cu and Zn was much higher than for nonessential metal elements [41]. Notably, the excretion of Pb and Cd is very slow in most organisms. Because shellfish are filter feeders, and both Pb and Cd complexes in suspension and in the sediments were ingested by and enriched in shellfish [42], accounting for the high enrichment coefficients of Pb and Cd in oysters, clams, and shrimps.
than 1000 was found for Cu, Zn, and Cd in oysters and Cu and Zn in shrimps, indicating serious accumulation of Cu, Zn, and Cd by oysters and shrimps in Jiuzhen Bay. Although the enrichment coefficients of Pb, Hg, and As were lower than 1000, the pollution levels of Pb, Hg, and As in seawater and sediments were relatively high. Therefore, the ecological and human health risks related to Pb, Hg, and As in the marine organisms of Jiuzhen Bay cannot be neglected.

4. Conclusions

This study reported the contents, distributions, pollution levels, and ecological risks of heavy metals (Cu, Pb, Zn, Cd, Hg, and As) in seawater, sediments, and living organisms of Jiuzhen Bay. The seawater was mainly polluted by Hg and As, while heavy Pb and Hg pollution was detected in sediments at a few stations. The main sources of heavy metal pollution in the marine environments of Jiuzhen Bay are from surface runoff carried by the Luxi and Wujiang Rivers and coastal discharges from local industries. The enrichment coefficients of Cu, Zn, and Cd were high in oysters and clams. According to the comprehensive pollution index values, the heavy metal pollution in oysters and clams was classified as heavy and light pollution, respectively. The heavy metal contents in fish and shrimps did not reach polluted levels. Given the pollution of Pb, Hg, and As in seawater and sediments at some stations within Jiuzhen Bay, the potential risks of Pb, Hg, and As in living organisms to both the local ecology and human health deserve increased attention. To ensure the safety of aquaculture products in Jiuzhen Bay, the sources of heavy metal pollution should be strictly controlled, and the polluted areas should be treated to restore their ecological functions.

Data Availability

No data were used to support this study.

Disclosure

Xia Sun and Bao-Shi Li are the co-first authors.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors’ Contributions

Xia Sun and Bao-Shi Li contributed equally.

Acknowledgments

This work was financially supported by the National Natural Science Foundation of China (Grant no. 41676074), the National Key Research and Development Program of China (no. 2018YFC140707600), and the Basic Scientific Fund for the National Public Research Institutes of China (Grant no. 2016Q04).

References


