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EDITED BY
Meilin Wu,
South China Sea Institute of Oceanology (CAS), China

REVIEWED BY
Liancong Luo,
Yunnan University, China
Lijie Duan,
Sun Yat-sen University, China
Qiao Yongmin,
Jinan University, China

*CORRESPONDENCE
Yutao Lei
leiyutao@scies.org

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Spatial distribution and risk assessment of heavy metals in seawater and sediments in Jieshi Bay, Shanwei, China

Wen Zhou[1], Qian Cao[1], Man Hong[1], Yutao Lei[1]*, Ding Wen[1] and Dengfeng Zhang[2]

[1]South China Institute of Environment Sciences, Ministry of Environment Protection of People's Republic of China (PRC), Guangzhou, China, [2]College of Resource and Environment, Huazhong Agricultural University, Wuhan, China

The contamination of heavy metals due to human activities has attracted great attention and may lead to serious environmental problems. This research was performed on seawater, sediments, and organisms in Jieshi Bay, China. The level of Cu, Pb, Zn, Cd, Hg, and As, respectively, was measured in each environment to acquire a comprehensive understanding of their sources and distribution and to accomplish a risk assessment. The results showed that the concentration of heavy metals in autumn was higher than those in spring, and surface water has a higher heavy metal content than bottom water. The main sources of these heavy metals could be surface runoff and industrial wastewater discharge in the said bay. Hg was the main pollutant in the seawater. Hg, Cu, Zn, and Cd could easily accumulate in organisms than other heavy metal contents. In addition, even though the concentration of Pb did not exceed the Seawater Quality Standard, decision-makers should still be attentive to the Pb content in fish because of bioaccumulation from seafood product consumption.

KEYWORDS

risk assessment, heavy metal, coastal contamination, bay, marine environment quality

1 Introduction

Heavy metal is one of the many global concerns, and the increasing quantity of heavy metals in seafood has threatened human health. Heavy metals (HMs) are characterized by high toxicity, difficult degradation, and easy biological enrichment. Some heavy metals are also important components of environmental endocrine disruptors, which are considered as one of the main characteristic pollutants harmful to water organisms and are typical marine priority control pollutants (Jiang et al., 2012; Wang et al., 2020). Once a heavy metal enters the ocean, it bioaccumulates and bio-amplifies in food webs, thereby harming ecosystems and human health (Jiang et al., 2012; Wang et al., 2018). Heavy metals may come from the natural weathering and erosion of rocks, many dispersed pollution sources, polluted rivers, domestic wastewater, industrial and agricultural sewage discharge, *etc*. (Birch et al., 1996; Matthiessen et al., 1999). The discharge of heavy metals into seawater environments may lead to bioaccumulation and bio-amplification in predators and, eventually, harm human health through the food chain (Bosch et al., 2016; Baltas et al., 2017; Liu et al., 2021; Liu et al., 2022).

Since the industrial revolution, the industrial construction and population boom in a bay's watershed has caused a sharp rise in heavy metal pollution. This dramatic increase in heavy metals flowing into the bays has overburdened the waterbody (Zhen et al., 2016; Zhixin et al., 2016). Some bays in China were severely polluted by heavy metals—for example, Cd and Hg pollution in Bohai Bay (Wei et al., 2008; Hu et al., 2013a), Liaodong Bay (Hu et al., 2013b; Zhang A. et al., 2017), and Hangzhou Bay (Pang et al., 2015; Fang et al., 2016) and Cu and Zn pollution in Xiangshan Bay and Quanzhou Bay (Sun et al., 2014; Liu et al., 2019; Yan et al., 2020; Zhang M. et al., 2020). The Jieshi Fishing Port is one of the top 10fishing ports in Guangdong, with vast fishing grounds and rich resources, and the protection of its ecological environment is particularly important. Although in the recent years there has been abundant research on the geochemical characteristics of heavy metals in the sediments of the South China Sea, investigations on heavy metals in Jieshi Bay are quite limited; most scholars have focused on the distribution characteristics of heavy metals and have only considered the water quality or sediment without studying the ecological risks of heavy metals from the perspective of the ecosystem. This paper investigated the spatial distribution of heavy metals in Jieshi Bay in spring and autumn, identified the degree of pollution of heavy metals, analyzed the possible sources and correlations of various heavy metals, and evaluated the ecological risks and potential threats to human health from heavy metals in this area. We hope that these studies will contribute to a comprehensive understanding of the distribution, source, accumulation, transport, and ecological risks of heavy metals in the various media of marine ecosystems and help the government to scientifically supervise the protection of coastal and marine ecosystems and public health.

2 Materials and methods

2.1 Study area

Jieshi Bay, located in the south of Shanwei City, Guangdong Province (22°39′–22°52′ N, 115°31′–115°49′ E), is a bay that connects to the South China Sea. The bay covers an area of 890 km^2, with an average depth of 5–18 m and a maximum depth of 20.5 m. The mouth of the bay faces south, having a width of 27 km and a depth of 18 km. It gradually becomes shallow from the mouth to the inside of the bay. The climate in Jieshi Bay is a typical South Asian tropical oceanic monsoon climate, with an annual average temperature of 22.8°C and an annual average precipitation of 2,019.8 mm. The irregular diurnal tide has affected the hydrodynamic condition of Jieshi Bay, making it special to study. The rising tide in the bay flows to the north-northwest, while the falling tide flows to the southeast. The large waves generated by northeast gales and tropical cyclones can reach 3 m in height. There are many harbors along the coast of Jieshi Bay, which has an average coastline of length 23.3 km. There are many excellent breeding harbors along the coast, with vast breeding areas and rich marine fishery resources, and it has a long history of seawater aquaculture.

The region is rich in seawater, port, and fishery resources and has significant ecological and economic value. In recent years, industry, mariculture, and tourism in this region have developed rapidly, and human activities have had a certain impact on the marine environment. To comprehend the spatial distribution and evaluate the ecological risks of HMs in the bay's semi-closed estuary and marine ecosystem, the presence of Cu, Pb, Cd, Hg, As, and Zn in seawater, sediments, and organisms was investigated. The results of the present research may contribute to scientifically regulating enterprises and managing government policies.

2.2 Sample collection and analysis

In April 2021 (spring) and August 2021 (summer), 20 seawater sampling sites, 10 sediment sites, and 12 ecological sites were selected in Jieshi Bay, China (Figure 1). All the samples were collected and preserved according to the *Specification for Marine Monitoring* (GB17378.3-2007) (Ma et al., 2008). Since the near-shore seawater depth ranged from 0 to 20.5 m, the bottom seawater sample was collected from both 0.5 m below the water surface and 2 m above the bottom for a water depth range of 10 to 20.5 m.

Seawater sample (500 ml) was collected and stored at 4°C. The sediment sample was taken using a grab sampler (China Qingdao Juchuang Environmental Protection Group Co., Ltd., JC8000S) from 0 to 5 cm above the bottom and stored in a polyethylene bag at 0–4°C. A 1.5-kg biological sample was grabbed from the selected site and then washed with seawater and cryopreserved. A 5-L acrylic sampler was used for surface water sampling. The surface seawater was filtered immediately once received and stored at 4°C. All sampling methods were performed in accordance with GB17378.3-2007. The seawater sample was filtered with a 0.45-μm filter and then acidified to pH<2 with sulfuric acid. The sediment sample was stored in a dry and ventilated environment, ground with an agate mortar, and then sifted through 160-mesh sieves. The biological samples

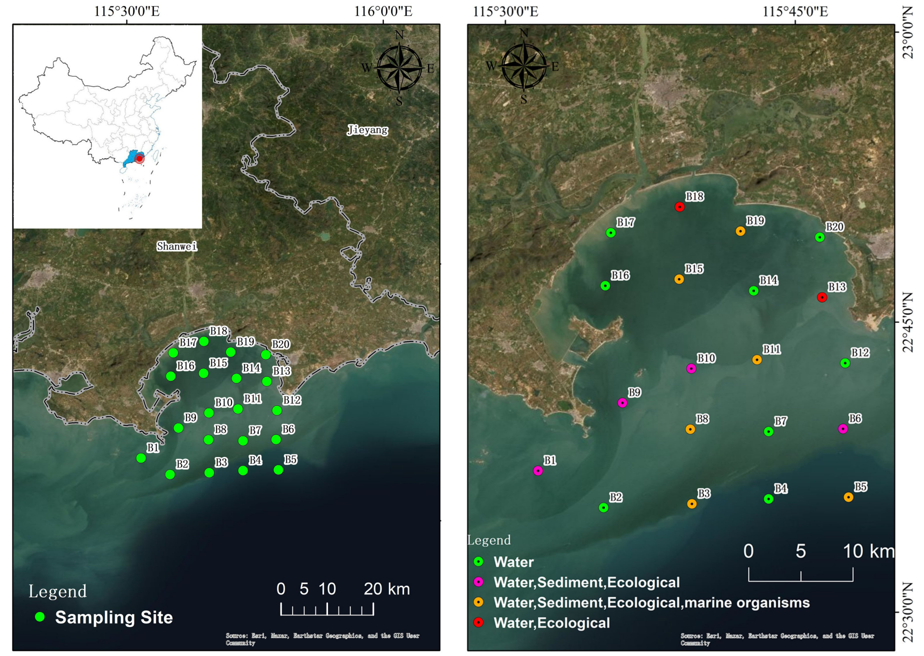

FIGURE 1
Location of sampling sites in Jieshi Bay, China.

were dissected and freeze-dried for 24 h. A 200-mg muscle tissue sample was extracted and then acidified with HNO_3–H_2O_2 (4:2) solution and heated to 120–140°C. Hg and As were tested with atomic fluorescence spectroscopy (Beijing Titan Instruments Co., Ltd, AFS-930). Cu, Pb, Zn, and Cd were measured with atomic absorption spectroscopy 10 times and measured with UV–visible spectrophotometry (The PinAAcle series of atomic absorption spectrometers, 900Z). The samples were analyzed using the national standard (GBW07309). The quality control of the standards was performed by using the national standard (GBW07309), the error of the parallel samples was less than 5%, and the recoveries of the standards were between 95% and 110%.

The species of biological samples from which muscle tissues were extracted were fish, crab, and other invertebrates. The study involving animals was reviewed and approved by the Ethics Committee of South China Institute of Environment Sciences.

2.3 Analytical assessment method

2.3.1 Pollution assessment

Three methods to evaluate the degree of pollution in the study area were applied, and they were water quality index assessment, coefficient of variation method, and principal component analysis.

As an indicator of water quality, the Water Quality Index (WQI) is widely applied in water quality assessments (Tyagi et al., 2013; Kükrer and Mutlu, 2019), turning complex water quality data into information that is understandable and useable to the public. It provides a single number that expresses the overall water quality at a certain location and time based on several water quality parameters. The calculation formula is as follows:

$$\mathrm{WQI} = \frac{1}{n}\sum_{i=1}^{n}\frac{C_i}{C_{i0}} \tag{1}$$

where C_i is the measured concentration of HM, C_{i0} is the first-class standard concentration of HM in the *Seawater Quality Standard* (GB3097-1997), and n is the number of water quality elements that were considered. In this assessment, Cu, Pb, Zn, Cd, Hg, and As were the HMs involved. WQI< 1 means that the pollution in seawater was slight and could be ignored, 1 ≤ WQI< 2 means that the seawater pollution is small, 2 ≤ WQI< 3 means that the seawater pollution is medium, 3 ≤ WQI< 5 means that the seawater pollution is strong, and WQI ≥ 5 means that the seawater is seriously polluted.

To reflect the volatility of the analytical chemistry data, coefficient variation (CV) (Brown, 1998; Bedeian and Mossholder, 2000) was used, which is a standardized measurement of the dispersion of a probability distribution or frequency distribution. The calculation formula is as follows:

$$CV = \frac{\sigma}{\mu} \tag{2}$$

where σ is the standard deviation of the measured concentration of the elements, and μ is the mean of the measured concentration of the elements.

Principal component analysis (PCA) (Shin and Lam, 2001; Li et al., 2006) is one of the most popular multivariate statistical techniques, describing chemical compound properties in order to extract and express information from the data.

2.3.2 Risk assessment

In this paper, the potential ecological hazard index method established by Swedish scholar Hankanson was used to evaluate the pollution level of heavy metals in sediments (Hakanson, 1980). Compared with the single-factor pollution index method, the potential ecological hazard index method more comprehensively considers the synergism between different heavy metals and the difference in pollution level and toxicity (Rezaee Ebrahim Saraee et al., 2011; Liu et al., 2021) as calculated in Equation (3):

$$\mathrm{E}_f^i = T_f^i \times C_f^i = T_f^i \times \frac{C^i}{C_n^i} \tag{3}$$

where E_f^i is the potential ecological hazard coefficient of the metal, T_f^i is heavy metal toxicity response coefficient, C_f^i is the pollution coefficient of the metal, C^i is the measured concentration of the heavy metal, and C_n^i is the evaluation standard for the metal (this paper adopts the national standard for class I sediments).

The comprehensive potential ecological hazard index of heavy metals in a single location is the sum of the potential ecological hazard indexes:

$$E_{RI} = \sum_{1}^{n} E_r^i \tag{4}$$

The potential ecological hazard index of a single metal can be divided into five levels from low to high, and the comprehensive potential ecological hazard index can be divided into four levels. The pollution degree of single or multiple pollutants at a certain point can be evaluated through the evaluation standard of the potential ecological hazard index. The specific evaluation standard is shown in Table 1 (Hakanson, 1980; Yi et al., 2016).

TABLE 1 Potential ecological harm index evaluation criteria.

	Degree	E_{RI}	Degree
<40	Low	<150	Low
40–80	Medium	150–300	Medium
80–160	Heavier	300–600	Heavier
160–320	Heavy	≥600	Serious
≥320	Serious		

Target hazard quotients (THQs) are a method used to evaluate the risk of pollutants to human health (Gu et al., 2018), calculated as Equation (5):

$$\mathrm{THQ} = \frac{\mathrm{EF} \times \mathrm{ED} \times \mathrm{FIR} \times \mathrm{c} \times 10^{-3}}{\mathrm{RFD} \times \mathrm{WAB} \times \mathrm{TA}} \tag{5}$$

where EF is contaminant exposure frequency (365 days/year), ED is the exposure years of the pollutants (the average lifespan of man is 70 years), and FIR is the food intake rate of the human body. The statistical data of the Food and Agriculture Organization of the United Nations (FAO) were used, in which the intake of fish is 36 g/day, and the intake of crustaceans is 5.42 g/day. C is heavy metal content in seafood (mg/kg), RFD is the daily reference dose of pollutants [Hg = 0.0005 mg/(kg·day), Cd = 0.001 mg/(kg·day), Pb = 0.004 mg/(kg·day), As = 0.0003 mg/(kg·day)],WAB is the average human body weight (60 kg), TA is the mean exposure time (365 days/year × ED) to non-carcinogenic sources (Storelli, 2008), and TTHQ is the sum of the hazard quotients of various heavy metals in seafood. If the hazard quotient is less than 1, it means that the seafood does not pose a health risk to human beings. On the contrary, when the hazard quotient is greater than 1, it indicates that there is a potential risk in consumption.

To evaluate the risk of heavy metal intake to human health, it is necessary not only to evaluate the hazard quotient of seafood but also to determine the dietary intake. The World Health Organization (WHO) formulated the provisional allowable weekly intake (PTWI) scale for heavy metals. The allowable weekly intakes of Hg, Cd, Pb, and As are 5, 7, 25, and 15 µg/(kg·body weight), respectively (Agusa et al., 2007). The formula for the weekly assessed intake of heavy metals (EWI) is as follows:

$$\mathrm{EWI} = \frac{\mathrm{c} \times \mathrm{FIR} \times 7}{\mathrm{WAB}} \tag{6}$$

2.3.3 Partitioning and bioaccumulation

For the evaluation of sediments, biota-sediment and bio-accumulation factors are commonly applied to evaluate the ecological risk of sediments (Hao et al., 2013; Hao et al., 2019). The bio-accumulation factor (BAF) is used to describe the ability of marine organisms to accumulate trace metals from the surrounding medium and is calculated *via* Equation (7):

$$\mathrm{BAF} = \frac{C_{\mathrm{organisms}}}{C_{\mathrm{sediments}} \ \text{or} \ C_{\mathrm{seawater}}} \tag{7}$$

where $C_{\mathrm{sediments}}$ is the measured concentration in sediments, C_{seawater} is the measured concentration in seawater, and $C_{\mathrm{organisms}}$ is the measured concentration in organisms.

Since a marine organism and its food are exposed to similar environments, the ratio of a substance's lipid-normalized concentration in the tissue of an aquatic organism to its organic carbon-normalized concentration in surface sediment does not change substantially over time. Thus, a BAF< 100 or a biota-sediment accumulation factor< 1 indicated no significant accumulating effect between organisms and the surrounding environment.

2.3.4 Data analysis tools

Software and statistical tools were utilized to accomplish the spatial distribution, data analysis, and correlation exploration procedures. The Kriging interpolation was completed by ArcGIS (ESRI, v10.2) to draw the spatial distribution of the study area. SSD curves were generated by Buzzlioz 2.0 in the R software. The correlation analysis figures were drawn with Origin and Excel.

3 Results and discussion

3.1 Distribution and pollution assessment of HMs in seawater

The concentration variation ranges of Cu, Pb, Cd, Hg, As, and Zn are shown in Table 2. Zn was the element with the highest concentration, while the concentration of Cd was relatively low. From highest to lowest, the mean concentrations of all the heavy metal elements in the seawater were Zn (3.84μg/L) > As (2.29μg/L) > Cu (2.04μg/L) > Pb (0.93μg/L) > Hg (0.06μg/L)> Cd (0.02μg/L), and the same rule applies to spring and autumn and the bottom layer. The dissolution of trace metals in the surface water and bottom water followed the same time change law for Cu, Pb, Cd, and As; the mean was greater than in spring. The average concentrations of Hg and Zn in autumn were greater than those in spring; this is due to differences in water temperature in different seasons. Temperature is a factor that influences the existing speciation of Hg and Zn, which, in very warm autumn weather, are released more frequently. More Cu, Pb, Cd, and As were released in spring when the water temperature was low (Gerringa et al., 2001; Gundersen and Steinnes, 2003; Nystroem et al., 2005).

According to the horizontal distribution of heavy metal content in the surface and bottom seawater of each site (Figure 2), the concentration of Hg in seawater in spring was higher than in autumn, and the concentration of pollutants in the coastal area was higher than in the offshore area, mainly because the coastal area is more affected by the discharge of industrial and domestic sewage. The water quality of the bottom seawater of Jieshi Bay is significantly better than that of the upper seawater, mainly because the upper seawater is greatly affected by aquaculture, industrial pollutant discharge, and other activities. In Jieshi Bay, heavy metal concentrations in seawater have an obvious seasonal trend, with autumn surface waters containing higher concentrations of Cu, Pb, Cd, Hg, and As than those of spring. These results could be associated with seasonal river flow changes in Guangdong. The distribution of Guangdong's rainy seasons is due to high-pressure recessions; studies show that the monsoon rainfall increases in summer but decreases in spring. The rainy season in Shanwei starts in late March, and the flood season spans from April to September

TABLE 2 Heavy metal concentrations in seawater.

Season		Index	Concentration: μg/L					
			Cu	Pb	Cd	Hg	As	Zn
Spring	Surface	Range	0.30–4.60	0.015–0.87	0.005–0.02	0.03–0.182	2.0–2.5	1.55–29.60
		Average	1.90	0.316	0.008	0.07	2.25	5.72
		Standard deviation	1.39	0.29	0.01	0.03	0.14	7.43
		Variable coefficient	72.98%	90.83%	69.60%	48.96%	6.38%	129.87%
	Bottom	Range	0.4–3.9	0.015–0.460	0.005–0.050	0.035–0.114	1.9–2.6	1.55–18.30
		Average	2.07	0.112	0.008	0.062	2.24	3.72
		Standard deviation	1.14	0.17	0.01	0.02	0.21	4.57
		Variable coefficient	55.11%	152.70%	138.29%	30.80%	9.36%	122.95%
Autumn	Surface	Range	0.9–5.0	0.43–4.62	0.005–0.260	0.017–0.141	2.1–3.0	1.55–8.60
		Average	1.99	1.71	0.040	0.055	2.38	2.46
		Standard deviation	1.07	1.19	0.08	0.03	0.20	1.97
		Variable coefficient	54.00%	69.36%	191.24%	54.79%	8.60%	91.45%
	Bottom	Range	0.6–4.1	0.46–3.15	0.005–0.450	0.016–0.114	2.0–2.6	1.55–18.30
		Average	2.19	1.571	0.040	0.048	2.29	3.45
		Standard deviation	0.94	0.85	0.11	0.03	0.15	4.62
		Variable coefficient	42.65%	54.30%	276.02%	61.61%	6.46%	133.84%
		Average	2.04	0.927	0.024	0.059	2.29	3.84
Marine Water Quality Standards (I)			≤5	≤1	≤1	≤0.05	≤20	≤20

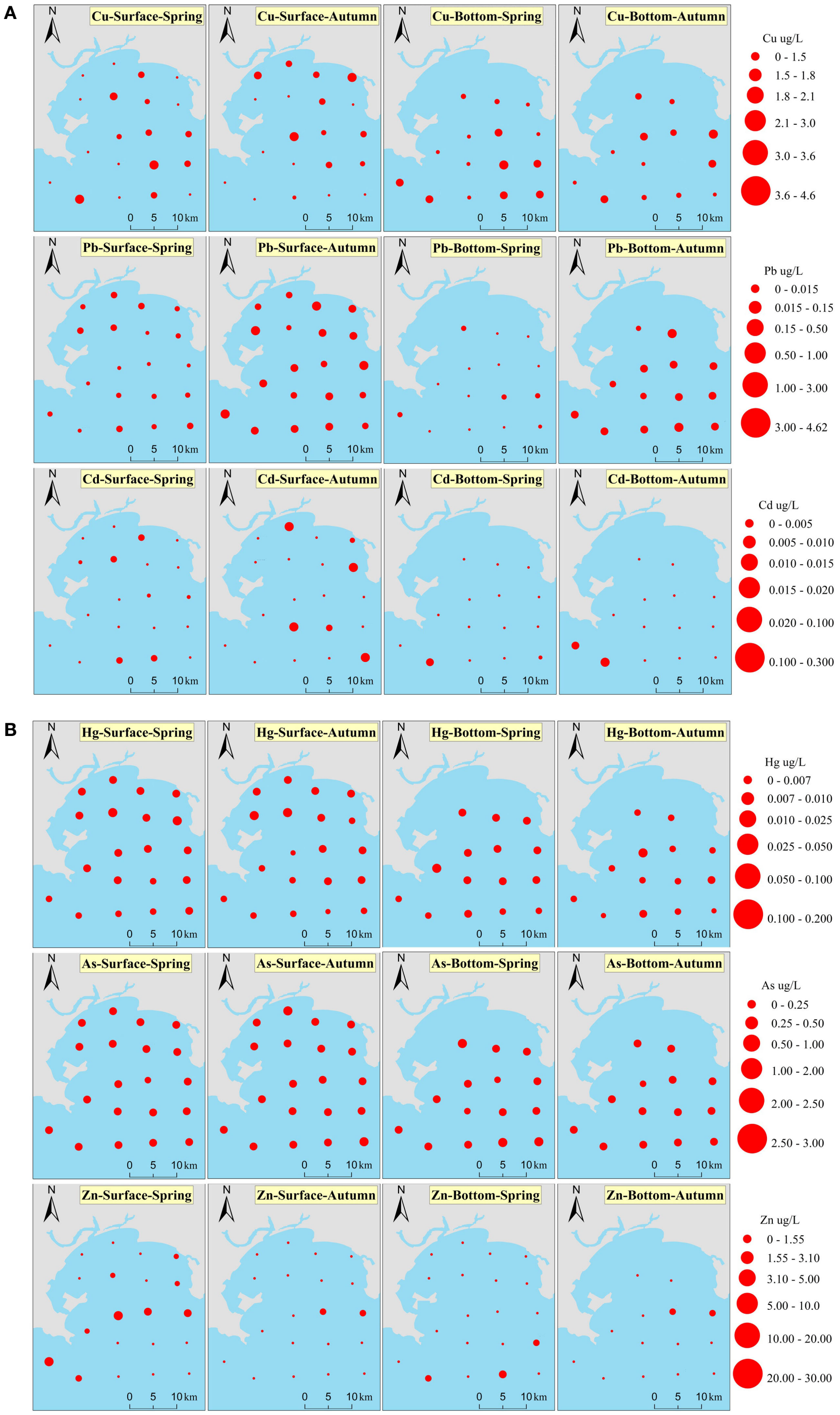

FIGURE 2
(A) The concentrations of HMs in seawater for Cu, Pb, Cd. **(B)** The concentrations of HMs in seawater for Hg, As, Zn.

(Zhang G. et al., 2017; Liu et al., 2018). There is more rain in spring, and the concentration of heavy metals in seawater decreases due to rainfall dilution.

To compare the variability between disparate groups and characteristics, the coefficient of variation was calculated (Table 2) to learn how much the data varied relatively. The result of the coefficient variation showed a relatively high variation in HMs, except As, indicating a diverse distribution of HMs in the study area, which might be from other different sources.

The comprehensive pollution index of HMs in the seawater of Jieshi Bay is shown in Figure 3. It shows that, regarding the bottom and surface seawaters of Jieshi Bay, the comprehensive pollution index of the heavy metals Cu, Pb, Hg, Cd, As, and Zn was less than an average of 1; thus, the pollution in the bottom seawater of Jieshi Bay is slight and could be ignored. The bottom seawater is clean. The mean of the comprehensive pollution index measured at two meteorological stations was greater than 1 only for the surface seawater in autumn, while the pollution comprehensive index for all other timepoints was less than 1. It can be seen that, in autumn, the respective stations on the west side of the bay and the east side of the bay's mouth were slightly affected by pollution. According to the annual wind speed variation statistics from Lufeng Meteorological Station, warm and cold air masses are active and cyclones are frequent. According to a study on the hydrodynamic characteristics of the sea area, the waves of Jieshi Bay are strong: their current

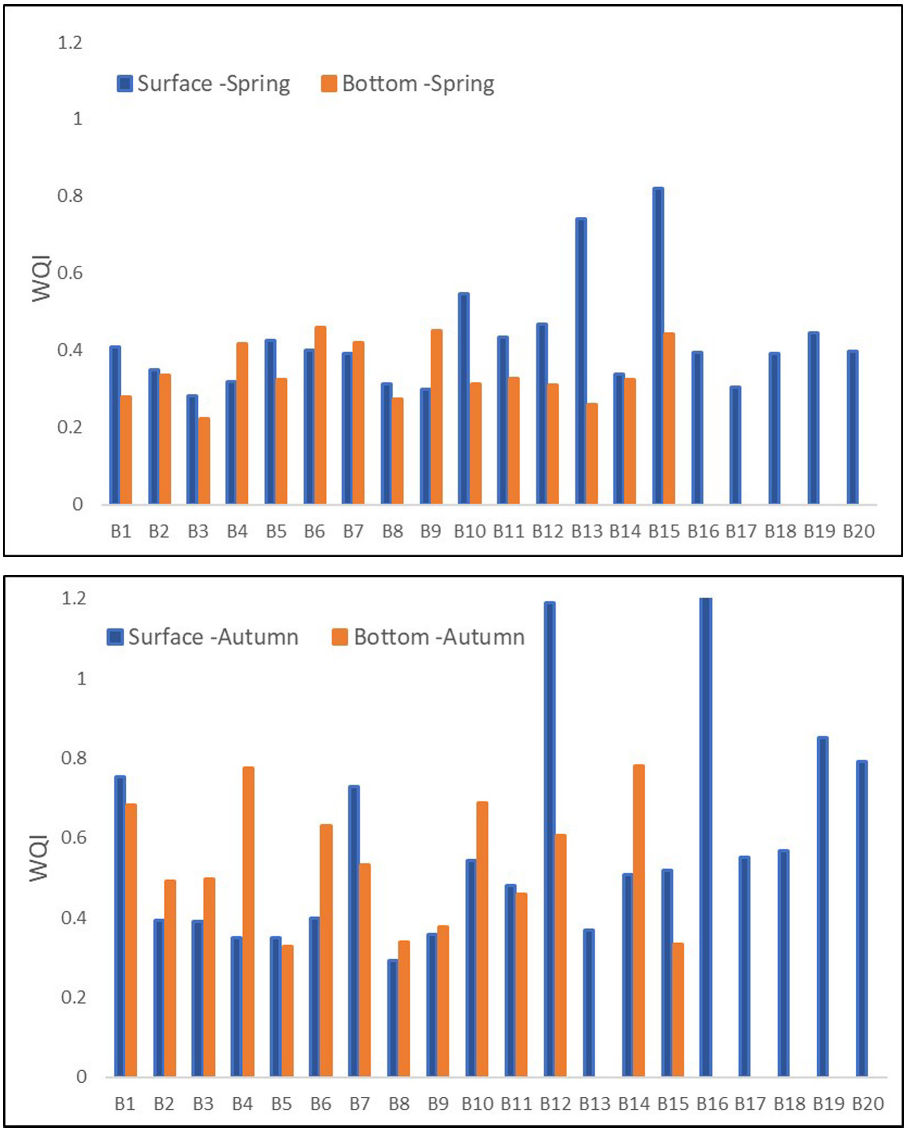

FIGURE 3
WQI of HMs in seawater.

velocity is large, their reflow was dominant in the past, their flow direction is perpendicular to the shoreline, strong waves are mainly in the E direction, and their occurrence frequency is about 27.3%. It can be speculated that the high comprehensive index of seawater pollution recorded in autumn at the stations on the west side of the Bay and the northeast side of the bay's mouth was mainly due to a seasonal phenomenon caused by the diffusion of high concentrations of heavy metals in the near-shore surface caused by high wind speeds and tidal current. The west and east sides of the bay's mouth appear to have larger pollution indices. In the new Shanwei port area and the west side of the Wukan waterway, particularly the mouth on the east side of the channel, the combined Pb and Hg ion concentration was higher, and it may be that the meteorological station was affected by the port wastewater, such as emissions from ships (Zhan et al., 2010).

Based on the water quality monitoring results taken over the years, it can be seen that Pb in Jieshi Bay has experienced an obvious upward trend, while other elements have experienced a steady downward trend (Gao and Chen, 2012; Pan et al., 2014; Gu, 2018; Zhao et al., 2018). Compared with other studies on heavy metals in seawater, the content of toxic metals, such as Cu, Pb, Cd, As, and Zn, in the seawater of Jieshi Bay was low, but Hg was slightly higher than in other research areas (Gu et al., 2012; Xiao et al., 2013; Li et al., 2017; Tang et al., 2018; Zhao et al., 2018; Lao et al., 2019; Nour, 2019; Zhang C. et al., 2020). On the whole, the seawater of Jieshi Bay is not polluted by heavy metals at present, but it should be noted that, with the expansion of the port, potential heavy metal pollution may be brought on by improvements in ship throughput and the rapid development of industry.

3.2 Distribution and risk assessment of HMs in sediment

The content of heavy metals in sediments can reflect the degree of heavy metal pollution in an area, which can provide a relevant basis to further understand the migration and transformation of various heavy metals in sediments as well as their possible sources. It can be seen from Table 3 that the sediment quality in the study area was good. From highest to lowest, the mean concentrations of all the heavy metal elements in the sediment were Zn (96.10mg/kg) > Pb (18.55mg/kg) > Cu (12.45mg/k.mg/kg) > As (6.13mg/k.mg/kg) > Hg (0.06mg/k.mg/kg) > Cd (0.02mg/kg). The coefficient of variation for Hg and As in spring, as well as for Cd in autumn, was greater than 50%, which is highly variable (CV > 36%). It can be inferred that Hg, As, and Cd in the sediments of Jieshi Bay may have been influenced by man-made sources, resulting in a high coefficient of variation (Wilding, 1985; Ma and Zhao, 2010; Liu et al., 2016). The anthropogenic source of Cd in sediments is mainly from industrial and agricultural activities, the anthropogenic source of Hg is mainly atmospheric deposition from human activities (Li et al., 2006), and the anthropogenic source of As in sediments is mainly from industrial activities (Mohammadi et al., 2015; Huang et al., 2018). This conclusion is also consistent with the large spatial dispersion of heavy metals in seawater found in this study.

It can be seen from the survey data on sediment heavy metals taken in this area for the last 15 years that heavy metal pollution in Jieshi Bay generally presents a downward trend (Qiu et al., 2011; Ye et al., 2018; Zhang L. et al., 2020), with Pb and As significantly declining, Cu and Zn remaining basically stable, and Hg and Cd slightly increasing. Combined with the coefficient of variation, it can be inferred that human activities have a great influence on the presence of Pb, As, Hg, and Cd in the sediments of this sea area.

The potential ecological hazard index method was used to evaluate the content of heavy metals in the sediments in the study area. The results in Table 4 showed that the potential ecological hazard index of six heavy metals, in descending order, was Hg > As > Pb > Cu > Cd > Zn, all of which are less than 40 and belong to the low hazard degree, among which, Hg, As, and Pb have a relatively high hazard degree. Hg and As were affected

TABLE 3 Heavy metal concentrations in sediments.

Season	Index	Concentration: mg/kg					
		Cu	Pb	Cd	Hg	As	Zn
Spring	Range	8.3–15.6	17.5–29.3	0.02	0.014–0.184	0.37–8.89	65.4–110.7
	Average	12.0	22.4	0.02	0.059	5.41	92.5
	Standard deviation	2.72	4.30	0.00	0.05	2.76	17.79
	Variable coefficient	22.68%	19.23%	0.00%	79.36%	51.15%	19.23%
Autumn	Range	9.1–16.0	5.8–22.8	0.04–0.19	0.055–0.118	5.32–11.80	77.0–116.7
	Average	12.9	14.7	0.02	0.069	6.84	99.7
	Standard deviation	2.44	4.55	0.05	0.02	1.82	13.20
	Variable coefficient	18.82%	30.92%	229.73%	26.14%	26.55%	13.24%
	Average	12.45	18.55	0.02	0.064	6.13	96.10

TABLE 4 Potential hazard index of heavy metals in sediments.

Season	Index	E_f^i						E_{RI}
		Cu	Pb	Cd	Hg	As	Zn	
Spring	Range	0.83–1.56	1.25–2.09	0.60	2.24–29.44	0.25–5.93	0.37–0.63	5.54–39.65
	Average	1.20	1.60	0.60	9.44	3.61	0.53	16.98
Autumn	Range	0.91–1.60	0.41–1.63	1.20–5.70	8.80–18.88	3.55–7.87	0.44–0.67	15.31–36.34
	Average	1.29	1.05	0.60	11.04	4.56	0.57	19.11
	Average	1.25	1.33	0.60	10.24	4.08	0.55	18.04

by man-made sources in the coefficient of variation. Therefore, the potential ecological hazard index of sediments in the study area was mainly affected by coastal industrial and agricultural activities and anthropogenic emissions. The comprehensive potential ecological hazard index was higher in autumn, but the overall hazard degree was low.

Compared with the coastal waters of the surrounding South China Sea, the overall quality of Jieshi Bay sea sediment is good (Liu et al., 2012; Wang et al., 2013; Zhang et al., 2015; Zhang et al., 2016; Wang et al., 2022; Zhang et al., 2022). We found that most of the heavy metal contents in the surface sediment was at the medium level, though the Hg and Zn contents were comparatively high in some sea areas. Although there was no obvious heavy metal pollution, we should still pay attention to human activities, which can, in part, lead to potential heavy metal pollution.

3.3 Concentrations and human health risk of HMs in organisms

The concentrations of Cu, Zn, As, Pb, Cd, and Hg in fish and crustaceans in summer and autumn are shown in Table 5. No shellfish were collected at the survey station; only fish and crustaceans were collected. The average contents of various metals in crustaceans captured in spring and autumn were Cu—13.8 and 12.6 mg/kg, Pb—0.20 and 0.59 mg/kg, Cd—0.140 and 0.212 mg/kg, Hg—0.017 and 0.009 mg/kg, As—1.50 and 1.20 mg/kg, and Zn—8.50 and 18.50 mg/kg, respectively. The average heavy metal contents in spring and autumn were Cu—1.3 and 0.8mg/kg, Pb—1.12 and 0.05mg/kg, Cd—0.044 and 0.005 mg/kg, Hg—0.022 and 0.007 mg/kg, As—0.40 and 0.40 mg/kg, and Zn—3.55 and 12.6 mg/kg, respectively. The differences between the same elements in different species may be related to the living habits, physiological characteristics, and specific physiological methods of metal accumulation in various marine organisms (Wenxiong and Jinfen, 2004). Crustaceans generally live in an environment close to the bottom of the sea, with polychaetes and other zooplanktons in the sediment serving as their main feeding source, causing them to absorb more heavy metals. The HM content in fish was low, which is closely related to their water layer habitat and feeding habits (Yi et al., 2014).

In spring, the average content of each pollutant in organisms was (in specific order) Cu > Zn > As > Pb > Cd > Hg, and in autumn, the average content of each pollutant in organisms was Zn > Cu > As > Pb > Cd > Hg. Cu and Zn are essential trace elements in living organisms, so their content in living organisms was higher than other metals. Pb and Hg are inimical to living organisms, so their contents were relatively low. Only in spring did the presence of Pb in fish exceed the relevant standards. As Pb ions in organisms may cause protein inactivation (Pourrut et al., 2008), higher Pb content in environmental media may affect the biological production of testosterone and estrogen; therefore, it can cause reproductive toxicity in human beings (Yang et al.,

TABLE 5 Biological quality monitoring results for Jieshi Bay.

Season	Species	Value	Concentration: mg/kg					
			Cu	Pb	Cd	Hg	As	Zn
Spring	Crustaceans	Range	9.4–22.2	0.02–0.51	0.041–0.298	0.009–0.023	0.50–2.80	0.20–18.8
		Average	13.8	0.20	0.140	0.017	1.50	8.50
	Fish	Range	1.0–2.2	0.02–3.92	0.003–0.061	0.008–0.055	0.30–0.50	0.2–7.2
		Average	**1.3**	**1.12**	**0.044**	**0.022**	**0.40**	**3.55**
Autumn	Crustaceans	Range	6.8–20.7	0.20–1.86	0.050–0.416	0.006–0.012	0.70–1.80	10.6–24.5
		Average	12.6	0.59	0.212	0.009	1.20	18.5
	Fish	Range	0.7–0.9	0.05–0.07	0.004–0.006	0.006–0.010	0.37–0.45	11.0–13.2
		Average	**0.8**	**0.05**	**0.005**	**0.007**	**0.40**	**12.6**

2019; Liu et al., 2022). Given the above-mentioned phenomenon of excessive Pb in seawater in autumn, in the future, the control of Pb emissions should be prioritized with respect to water pollution control in this region.

As can be seen from Table 6, the weekly assessed intake (EWI) of heavy metals in seafood in this region was lower than the PTWI for human beings as established by WHO. The harm quotient values of Pb, Cd, Hg, and As in crustaceans were all less than 1, indicating that there was no potential risk from Pb, Cd, Hg, and As to human health when eating this kind of seafood. The harm quotient values of the heavy metals Cd and Hg in fish were less than 1, indicating that there is no potential risk from Cd and Hg to human health when eating this kind of seafood. However, the hazard quotients of Pb and As in fish were greater than 1, indicating that there is a certain health risk in eating fish. The anthropogenic sources of Pb are mainly lead mining, metallurgy, and waste gas. Lead is a toxic heavy metal that can cause great harm to the human body. Lead and its compounds can cause harm to the nervous system, hematopoietic system, digestive system, kidney system, cardiovascular system, and endocrine system after entering the body. If the content is too high, lead poisoning will occur. As exists widely in the natural environment, and its anthropogenic sources are mainly smelting activities and chemical synthesis agents. As is divided into two forms: an organic form without toxicity and an inorganic form with high toxicity. The total amount of As was measured in laboratory tests and analyses, and the organic form of As was about 90%. As poisoning can cause abdominal pain, vomiting, and muscle weakness and can persist in the body for a long time.

3.4 Interrelationship and source analysis of HMs

In this study, the coefficient of variation was selected to quantitatively reflect the difference in heavy metal fluctuation degrees on a spatial scale among the monitoring stations. Excel statistical software was used to conduct a statistical analysis of the survey data, and the results are shown in Table 2. Except for As, the coefficient of variation for the elements was relatively large. This indicates that the spatial dispersion of heavy metal content in the water between the survey stations was large; thus, the heavy metal pollutants in the study sea area may have come from different pollution sources.

Since the HMs were hardly absorbed in the seawater, they were not stable. They were easily affected and transferred by hydrodynamic and environmental conditions. Thus, the sources of HMs in seawater were difficult to distinguish. The coefficient of variation for the HMs presented strong correlations, which means that they may originate from the same source; thus, the homology of the source must be determined by correlating the HMs (Li et al., 2009). Pearson's correlation coefficient was used to measure the statistical association between the continuous variables of interest. The results of Pearson's correlation coefficient matrix for the HMs in Jieshi Bay (Figure 4) showed a moderate degree of correlation between Cd and As in the surface water. For the bottom water, the HMs had weak correlations. In conclusion, Cd and As in surface water may share common sources, and the other HMs might not have a strong relationship.

PCA is a mathematical tool that represents variations present in a dataset using a small number of factors (Granato et al., 2018). For visual analysis, a two- or three-dimensional projection of samples is usually constructed with axes (principal components, PCs) as factors. PCA was applied to explore similarities and hidden patterns among the HMs to find potential sources for the principal components (Table 7). PC1, accounting for 30.36% of the total data variance, had high positive loading for Cd and As. PC2, accounting for 19.97% of the total data variance, had negative loading for Pb and Zn. PC3, accounting for 17.22% of the total data variance, had negative loading for Cu and Hg. The accumulated percentage of variance in the three PCs was 67.54%, which means that these three PCs could represent the principal component of the samples. The characteristic performance factor variable of principal component 1 had a high load of Cd and As elements, indicating that Cd and As may have the same source (Figure 5). The results of the principal component analysis are similar to the Pearson correlation analysis. The waters around Jieshi Bay are surrounded by several Shanwei towns (Lufeng City, Jieshi Town, Taihu Town, Jinxiang Town, and Jiesheng Town). The industrial park is distributed around the continental area of Jieshi Bay, and it is reasonable to assume that a large amount of Cd and

TABLE 6 Weekly assessed intake and hazard quotients of heavy metals in marine organisms.

Season	Species	TTHQ	THQ				EWI			
			Pb	Cd	Hg	As	Pb	Cd	Hg	As
Spring	Crustaceans	0.47	0.005	0.013	0.003	0.452	0.126	0.089	0.011	0.949
	Fish	1.02	0.168	0.026	0.026	0.8	4.704	0.185	0.092	1.68
Autumn	Crustaceans	0.40	0.013	0.019	0.002	0.361	0.373	0.134	0.006	0.759
	Fish	0.82	0.008	0.003	0.008	0.8	0.21	0.021	0.029	1.68

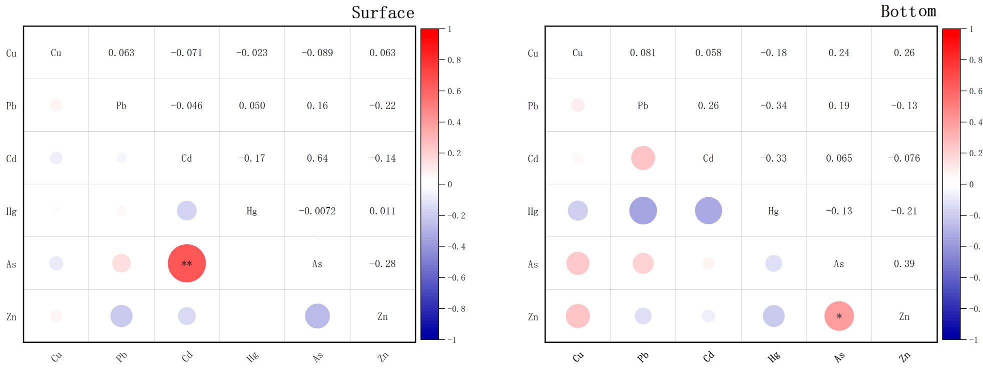

FIGURE 4
The correlation analysis of HMs in the seawater of Jieshi bay, respectively (P<0.05, P<0.1).

TABLE 7 Principal component analysis results for heavy metals in the seawater of Jieshi Bay.

Principal component	Initial eigenvalue			HMs	Load of each item		
	Eigenvalue	Percentage of variance (%)	Cumulative (%)		PC1	PC2	PC3
PC1	1.821	30.358	30.358	Cu	-0.136	0.162	0.798
PC2	1.198	19.973	50.331	Pb	0.173	0.701	0.206
PC3	1.033	17.216	67.547	Cd	0.605	-0.334	0.065
PC4	0.911	15.188	82.735	Hg	-0.125	0.427	-0.560
PC5	0.731	12.186	94.921	As	0.651	-0.015	-0.038
PC6	0.305	5.079	100.000	Zn	-0.382	-0.434	0.031

As in this area comes from surface runoff and industrial wastewater discharge. The characteristic performance factor variable of principal component 2 had a high load of Pb and Zn elements, but the Zn load in principal component 2 was negative, so it is difficult to judge whether the source is the same. The results of the principal component analysis are similar to the Pearson correlation analysis. The characteristic performance factor variables of principal component 3 had a high load of Cu and Hg elements, but the Hg load on principal component 3 was negative, so it is difficult to judge whether the source is the same. The results of the principal component analysis are similar to the Pearson correlation analysis. Based on the analysis of the correlation coefficient relationship matrix of heavy metals in seawater, except for Cd and As, elements such as Cu, Pb, Zn, and Hg had little correlation with each other, and the probability of a common source is low, which is consistent with the conclusion of the Pearson correlation analysis.

The ability of marine organisms to accumulate heavy metals was quantified with BAF: $BAF_{sediment} > 1$ indicates that the organism can accumulate heavy metals. $BAF_{seawater} > 1$ indicates that the organism has a strong enrichment capacity for heavy metals (enrichment capacity is proportional to the value), and $BAF_{seawater} > 100$ indicates that the organism has a strong enrichment capacity for heavy metals. The data in Table 8 show that, in addition to the Pb data from different stations, the $BAF_{seawater}$ of Cu, Cd, Hg, As, and Zn was greater than 100, indicating that the organisms in this sea area have an enrichment capacity for these six heavy metals, especially Cu, Cd, and Zn, which had the highest $BAF_{seawater}$ values. However, as essential elements for biological growth, Cu and Zn levels are relatively high in fish and crustaceans, indicating that the organisms in this sea area have a stronger enrichment capacity for Cd. There were excessive Zn and Hg levels in the seawater of the study area, and the organisms also showed a strong enrichment ability, especially for Zn. In the spring sample data, it was found that the $BAF_{sediment}$ value of Cd for organisms was greater than 1, and the $BAF_{sediment}$ values of Cu and Hg in some sites were within the critical value. In the autumn sample data, it was found that the $BAF_{sediment}$ value of Cd for organisms in most sites was greater than 1. The $BAF_{sediment}$ value of Cu at some sites was within the critical value, and the $BAF_{sediment}$ value of other heavy metals was low.

$BAF_{seawater}$ and $BAF_{sediment}$ were used to evaluate the accumulation of heavy metals in the medium. Cd was the most accumulated element. According to relevant studies,

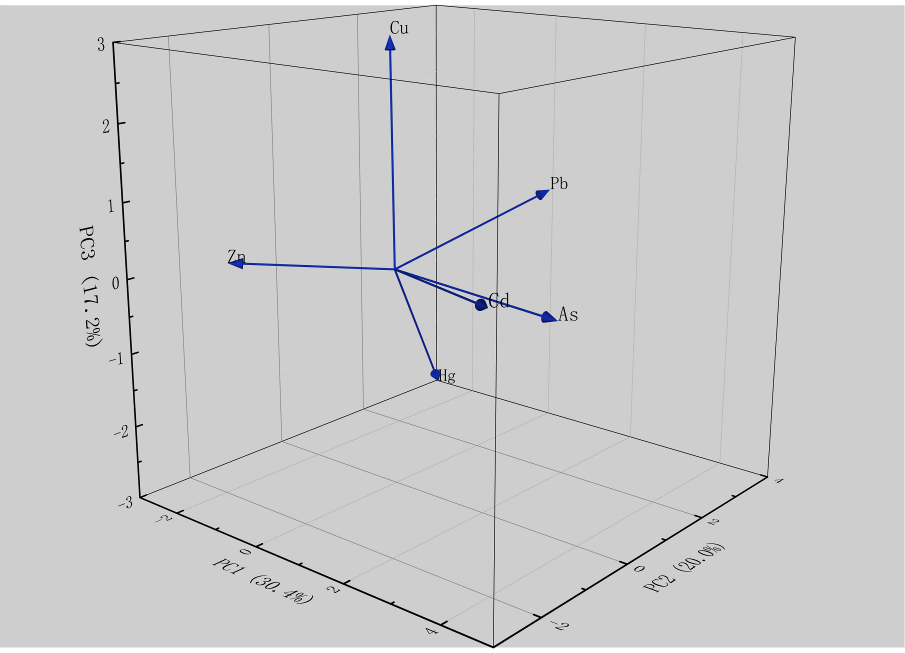

FIGURE 5
PCA analysis of HMs in seawater.

TABLE 8 Biological enrichment index of heavy metals in each phase of Jieshi Bay.

Parameter	Time	Spring						Autumn					
	location	Cu	Pb	Cd	Hg	As	Zn	Cu	Cu	Cu	Cu	Cu	Cu
BAF$_{sediment}$	3	0.77	0.01	2.38	0.24	0.1	0.09	0.88	0.88	0.88	0.88	0.88	0.88
	5	0.06	0	2.7	1.2	0.09	0	0.05	0.05	0.05	0.05	0.05	0.05
	8	1.04	0.01	8.8	0.19	0.24	0.11	0.9	0.9	0.9	0.9	0.9	0.9
	11	0.17	0	2.43	1.34	0.44	0.03	1.32	1.32	1.32	1.32	1.32	1.32
	15	1.22	0.1	2.88	0.68	0.06	0.04	1.06	1.06	1.06	1.06	1.06	1.06
	19	1.4	0.08	2.98	0.18	0.05	0.04	0.98	0.98	0.98	0.98	0.98	0.98
BAF$_{seawater}$	3	24,750	425	2,375	375	282.61	5,838.71	8,843.75	8,843.75	8,843.75	8,843.75	8,843.75	8,843.75
	5	3,333.33	22.99	10,800	763.89	227.27	129.03	500	500	500	500	500	500
	8	21,500	428.57	35,200	185.48	750	5,806.45	15,166.67	15,166.67	15,166.67	15,166.67	15,166.67	15,166.67
	11	592.59	3,000	4,850	515.38	375	176.92	6,125	6,125	6,125	6,125	6,125	6,125
	15	4,528.57	2,736.11	2,875	199.37	180	902.44	8,233.33	8,233.33	8,233.33	8,233.33	8,233.33	8,233.33
	19	7,000	3,362.9	2,975	146.15	173.91	2,387.1	4,540	4,540	4,540	4,540	4,540	4,540

the metabolic period of Cd in organisms is longer than that of other metals, which is also the reason why the Cd content in organisms exceeds that of other metals (Lin et al., 2017). Moreover, Cd and Ca have similar geochemical properties (Zhang, 1991), which also makes it easier for Cd to accumulate in organisms. The chemical form of heavy metals is also an important factor affecting the content of heavy metals in marine organisms. Cd in the sediments of East Guangdong Sea, where Jieshi Bay is located, mainly exists in an acid-soluble state, which is easier to absorb by organisms. On the whole, organisms in this sea area have a stronger ability to enrich heavy metals in seawater, and the quality of seawater is more important for people who mainly eat seafood.

4 Conclusion

In summary, this study analyzed the distribution characteristics of six heavy metals in the seawater, sediments, and organisms in Jieshi Bay. Generally, the level of heavy metal content was relatively low, and the main sources for these heavy metals could be surface runoff and industrial wastewater discharge in the bay. For seawater, the concentration of the heavy metals in autumn was higher than those in spring, and the surface water has higher heavy metal content than the bottom water. Hg is the main limiting factor according to the statistics. For sediments, the potential ecological risks of Hg, As, and Pb in sediments were relatively high compared with the rest of the contents. For the organisms, the content of heavy metals in fish was much lower than that in crustaceans, and lead in fish had exceeded the standard in spring. The seawater contributes more to bioaccumulation in fish and crustaceans than sediments, indicating that it is important to protect the seawater quality because of the bioaccumulation from seafood product consumption. Even if the heavy metals have not threatened the environment in the bay, it remains necessary to enhance pollutant management while especially developing the discharge of wastewater.

Data availability statement

The original contributions presented in the study are included in the article/supplementary material. Further inquiries can be directed to the corresponding author.

Ethics statement

The study involving animals was reviewed and approved by the Ethics Committee of South China Institute of Environment Sciences.

Author contributions

WZ: main contributor to conception, design of the work, and result analysis. QC: drafting of the article, revising it critically for important intellectual content, and ensuring the discussions related to the work. MH: data analysis and interpretation. YL: data collection and contributes to conception design. DW: data collection. DZ: using PCA analysis to determine the correlationship of heavy metals. All authors contributed to the article and approved the submitted version.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher's note

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