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# Spatial distribution and contamination assessment of heavy metal pollution of sediments in coastal reclamation areas: a case study in Shenzhen Bay, China

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#### **Abstract**

**Background:** With the continuous advancement of global urbanisation, humans have begun to overutilise or improperly utilise the natural resources of bay areas, which has led to a series of ecological and environmental problems. To evaluate the spatial distributions and potential ecological risks of heavy metals in sediments of Shenzhen Bay, China, an analysis of As, Cd, Cr, Cu, Pb, and Zn regarding their content, correlation (Pearson coefficient), pollution degree, and potential ecological risks was conducted.

**Results:** The heavy metal contents in the sediments decreased in the order of Zn > Cu > Cr > Pb > As > Cd, with contents of 175.79 mg kg<sup>-1</sup>, 50.75 mg kg<sup>-1</sup>, 40.62 mg kg<sup>-1</sup>, 37.10 mg kg<sup>-1</sup>, 18.27 mg kg<sup>-1</sup>, and 0.20 mg kg<sup>-1</sup>, respectively. The results showed that the overall sediment quality in Shenzhen Bay generally met the China Marine Sediment Quality criteria, and the heavy metal contents were significantly lower than those reported in the same type of bay area worldwide. Furthermore, the order of grade of potential ecological risk of the heavy metals was as follows: As and Cd were found to pose moderate ecological risks, with their potential hazard indices reaching a high level, whereas the potential ecological hazard indices of Cu, Pb, Zn, and Cr were all at relatively low levels.

**Conclusions:** The potential hazard indices of the heavy metals decreased from the inner bay toward the outside. The accumulation and content of the analysed heavy metals in the Shenzhen Bay sediments are mainly controlled by historical land-source pollution and land reclamation projects. This study presents the current state of sediment quality in Shenzhen Bay. The results may assist in the definition of future bay area management measures specifically targeted at monitoring heavy metal contamination.

Keywords: Heavy metal, Sediments, Potential ecological risk, Source analysis, Shenzhen Bay

#### **Background**

Coastal bays adjacent to rapidly developing cities are an important resource for human survival and social development, and are regions of active land—ocean interaction

that are sensitive to anthropogenic coastal occupation and global sea-level rise [29, 32]. Owing to their superior geographical locations and unique natural environments, they tend to have significant economic and natural values [23]. Many coastal bays around the world play an important role in port construction and house large capital cities with rapidly growing populations [2, 3]. However, with economic development, humans have begun to overutilise and improperly use this precious natural

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resource in some highly developed coastal bays, including Bohai Bay [6], Jiaozhou Bay [39], Zhelin Bay [13], and Quanzhou Bay [40] in China, and worldwide [1, 5, 14, 24, 28, 30, 39]. Moreover, due to the weak hydrodynamic conditions of coastal bays, increasing amounts of heavy metal pollutants are discharged into coastal waters but cannot be diluted or degraded, which causes heavy metal pollution [25, 26].

Coastal bays are located in marine-terrestrial interlaced zones. Active land and sea and frequent human activities are resulting in increasingly complex environments, causing these regions to become sensitive to natural diversity and anthropogenic pollution [43]. Pollutants such as heavy metals are transported and accumulated in sediments, rendering them important destinations for pollutants. When heavy metals enter coastal environments, they accumulate in sediments, which act as a sink, or are released from sediments by natural or anthropogenic disturbance (i.e., pH, dissolved oxygen, or electrical conductivity), in which case they act as a source of heavy metals in surface water, presenting another source of water body pollution [25, 26]. Numerous studies have shown that > 90% of heavy metals are absorbed by suspended sediments, resulting in significant accumulation of heavy metals in coastal sediments [35]. Therefore, sediments are recognised as the most significant pollution sink and also a pollution source of heavy metals, and regular monitoring and assessment of heavy metals in sediments is a prerequisite for ecological risk evaluation [44].

Heavy metals in coastal sediments originate from natural processes (i.e., atmospheric input, soil erosion, and rock weathering set the background values of heavy metals) and anthropogenic activities (i.e., industrial discharge, mining, agriculture, transportation, and wastewater) [22, 33]. Anthropogenic inputs are the main source of pollution in the marine environment, and heavy metals are increasingly introduced into estuarine and coastal environments by draining rivers and oceanic dumping [21]. Heavy metals are recognised as a group of pollutants with high ecological risks because they are not removed from water via self-purification [10]. They can accumulate in suspended particles and sediments, be released back into water body under favourable conditions, enter the food web, and cause health problems [10, 27].

Shenzhen area is located in the Pearl River Delta region, and is a densely populated economically developed centre in southern China. As the most successful special economic zone in China, it has experienced rapid industrialisation, urbanisation, and population growth over the past 40 years [36]. Consequently, both Pearl River Estuary and Shenzhen Bay (SZB) have experienced severe contamination from human activities [15]. Since the opening and reform of China, over the

last two decades, heavy metals have accumulated in the sediments of the western coast of Shenzhen and have become a serious pollution concern [17]. Previous studies have shown that the heavy metal content in sediments has increased in the SZB in recent years [20, 31, 36, 38, 45]. However, currently available data on heavy metals in the SZB are insufficient for investigating the present quality and pollution levels of the sediments following the implementation of water pollution control in 2015.

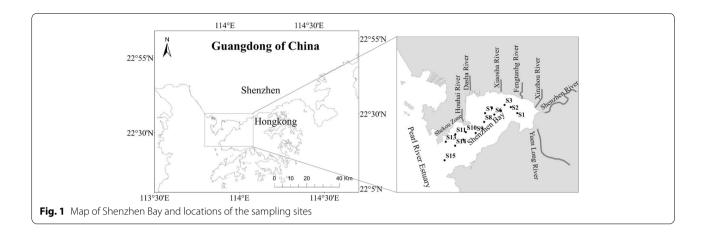
Therefore, the objectives of this study were to: (1) determine the spatial distributions of As, Cd, Cr, Cu, Zn, and Pb in sediments of the SZB; (2) assess the heavy metal contamination status and potential environmental risks of the sediments using the geo-accumulation and potential ecological risk indices; (3) identify possible sources of heavy metals through Pearson correlation and cluster analysis, and (4) analyse of the impact of reclamation on heavy metals in the SZB sediments. This study is helpful for understanding the variability of heavy metals status in the SZB; it provides the detailed information for coastal environmental management.

## Material and methods

#### Study area

This study was conducted in the SZB (22°24′–22°32′E, 113°53′-114°02′N), a semi-enclosed coastal embayment located in the east of the Pearl River Estuary in Guangdong Province, China. The northern half of the SZB belongs to Shenzhen, while the southern half belongs to the Hong Kong Special Administrative Region. It is approximately 17 km long and 4-10 km wide, with a water area of 90 km<sup>2</sup>, and a water depth of less than 5 m [41]. The main rivers flowing into the bay are the Pearl, Shenzhen, Yuen Long, Xiaosha, Dasha, Fengtang, and Xinzhou rivers, with additional inflow from other subrivers. The Shenzhen River flows northeast to southwest into the bay, and its hydrodynamic conditions are mainly affected by the tides and runoff of the Pearl River Estuary; other coastal runoffs are also partially affected. Owing to the relatively closed coastal bay, no obvious spatial differences in the hydrodynamic conditions are apparent. At the time of this study, the average water depth was 2.9 m, the average tidal range was 1.4 m, and the average flow velocity was 0.3 m s<sup>-1</sup> [41]. Pollutants from the coasts of Shenzhen and Hong Kong and along the Shenzhen River enter the bay through rivers, and pollutants from other parts of the Pearl River Delta reach the bay through the runoff of the Pearl River Estuary. The heavy metal data used in this study were obtained from the SZB sediment samples collected in June 2020. Maps of the SZB and sampling sites are shown in Fig. 1.

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#### Sediment collection and analysis

On June 28, 2020, 15 surface sediment samples (0–5 cm) were collected from the intertidal zone of the SZB (Fig. 1). The sediments were placed in clean sealed plastic bags and frozen at  $-20\,^{\circ}\text{C}$  for pre-treatment. In the laboratory, the sediments were thawed and air-dried under dust-free conditions. The semi-dried state, it was crushed with a clean glass bottle, placed in an oven, and dried at a constant temperature of  $35\pm2\,^{\circ}\text{C}$ . Once dry, any residual roots, shells, and other debris were removed, and the clean sample was ground with an agate mortar and pestle and sieved using a 63- $\mu$ m nylon sieve. The sieved samples (<63  $\mu$ m) were stored in sealed plastic containers at 4  $^{\circ}\text{C}$  for further measurements.

Then, 0.2–0.5 g (with an accuracy of 0.001 g) of each sample was taken for microwave digestion (CEM MARS 6, USA). After complete digestion, the sample was diluted with deionised water to 100 mL. For each digested sample batch, the reagent blank and the sediment reference material (GBW07314, 2020), issued by the State Oceanic Administration, were subjected to the same digestion procedure. The samples were analysed using an inductively coupled plasma optical emission spectrometer (ICP-OES, PerkinElmer, Avio<sup>TM</sup>500, USA).

#### Pollution assessment methods

#### Geo-accumulation index

The geo-accumulation index  $(I_{geo})$  was used to evaluate the degree of heavy metal pollution in the sediments [9], and was calculated using the following formula:

$$I_{geo} = log_2 \left(\frac{C_n}{1.5B_n}\right), \tag{1}$$

where  $C_n$  is the heavy metal content;  $B_n$  is the geochemical background value of the heavy metals, and 1.5 is the coefficient of possible variations of the background value

of the metals [16]. The  $I_{geo}$  values were grouped into classes representing different pollution degrees, which are listed in Additional file 1: Table S1.

#### Potential ecological risk index (PERI)

The PERI is a quantitative index used to evaluate the pollution and potential ecological risks linked to the accumulation of one or more heavy metals. The calculation methods for the ecological risk of single heavy metals and the comprehensive ecological risk of multiple heavy metals are as follows [18]:

$$E_r^i = T_r^i \times \frac{C_i}{C_r^i},\tag{2}$$

$$RI = \sum_{i}^{n} E_{r}^{i} = \sum_{i}^{n} T_{r}^{i} \times \frac{C_{i}}{C_{r}^{i}}, \qquad (3)$$

where  $T_r^i$  is the toxic response factor of heavy metal i,  $C_r^i$  is the background content of heavy metal i, and  $C_i$  is the content of heavy metal i. The classification of the PERI is listed in Additional file 1: Table S2.

#### Pearson correlation analysis

Pearson correlation analysis was used to evaluate the correlation of different heavy metals in the SZB sediments, and the spatial correlation between various heavy metals was compared. Correlation was distinguished according to the magnitude of the Pearson coefficient, with values less than 0.3 indicating that the correlation can be regarded as irrelevant, values of 0.3–0.5 indicating a low correlation, values of 0.5 to 0.8 indicating a moderate correlation, and values of 0.8–1 indicating highly correlated heavy metals.

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#### Cluster analysis (CA)

To determine the heavy metal source, CA was carried out using hierarchical clustering, which connects data into clusters according to their distance. The results are illustrated in dendrograms representing the steps in the hierarchical clustering solution and the values of the distances between the clusters, which reflect the proximity and interrelationship among the heavy metals.

#### Statistical analysis

Variance analysis of the heavy metal contents was carried out using Microsoft Excel 2019. Pearson correlation analysis and CA were conducted using SPSS 25.0 (SPSS Inc., Chicago, USA). All figures were prepared using Origin 2021b software for Windows.

#### **Results and discussion**

## Contents and spatial distribution of heavy metals in sediments

The contents of As, Cd, Cr, Cu, Pb, and Zn, specified in the China Marine Sediment Quality Standard (GB18668-2002), are listed in Table 1. In the sediments, their contents ranged from 4.02 to 41.34 mg kg $^{-1}$ , 0.05 to 0.81 mg kg $^{-1}$ , 5.06 to 81.87 mg kg $^{-1}$ , 16.41 to 81.48 mg kg $^{-1}$ , 5.41 to 67.21 mg kg $^{-1}$ , and 83.22 to 259.57 mg kg $^{-1}$  for As, Cd, Cr, Cu, Pb, and Zn, respectively, in the order of Zn>Cu>Cr>Pb>As>Cd. The

metals with the highest contents, Zn and Cu, had average contents of 175.80 mg kg<sup>-1</sup> and 50.74 mg kg<sup>-1</sup>, respectively (Fig. 2 and Table 1). The heavy metal contents at all sampling sites met the Class-2 standard of marine sediments. The contents of As and Pb at S2, S4, S5, S8, S9, and S10 were higher than the values of the Class-1 standard, but lower than the values of the Class-2 standard. The Cd content at S1 and S2 was higher than that in the Class-1 standard and lower than that in the Class-2 standard. Only the Cr values at S9 were slightly higher than that in the Class-1 quality standard, which was 81.87 mg kg<sup>-1</sup>. The contents of Cu at S11, S14, and S15 were lower than that in the Class-1 standard. The Zn contents at S1, S2, S3, S4, S5, S6, S8, S9, and S10 were higher than the Class-1 standard value but lower than the Class-2 standard value.

Dai et al. [8] continuously monitored the SZB sediments from 2000 to 2007 and their results showed average contents of As, Cd, Cr, Cu, Pb, and Zn in the middle and at the mouth of the bay that were slightly higher than the results of this study. This is believed to be mainly due to the accelerated transfer and shutdown of polluting enterprises in the surrounding areas of the SZB in recent years. The high-tech industry is continuously developing, and the level of heavy metal pollution is continuously decreasing. Analysing the heavy metal content in sediments from the inside to

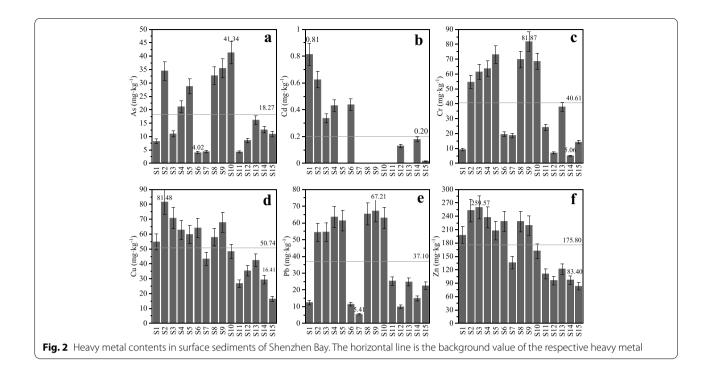
**Table 1** Heavy metal contents in surface sediments of the Shenzhen Bay compared with the average heavy metal contents in sediments of other bays/harbours around the world (mg·kg<sup>-1</sup>, dry weight)

	As	Cd	Cr	Cu	Pb	Zn	No. of samples	References
Shenzhen Bay, China	18.27	0.2	40.61	50.74	37.09	175.80	15	This study (average)
	4.02-41.34	0.05-0.81	5.06-81.87	16.41-81.48	5.41-67.21	83.22-259.57	15	This study (range)
Yellow River Estuary, China	NA	NA	44.09	22.85	43.05	54.12	96	[33]
Pearl River Estuary, China	NA	NA	106	45.7	57.9	177	23	[42]
Yangtze River Estuary, China	9.1	0.19	79.1	24.7	23.8	82.9	30	[37]
Jiaozhou Bay, China	NA	0.42	69.9	38.8	55.2	107.4	29	[39]
Zhelin Bay, China	5.47	0.051	69.9	88.0	50.9	204.5	16	[13]
Oyster Bay, Australia	15	NA	NA	35	98	204	55	[1]
Florida Bay, USA	NA	NA	162	15	8.4	31	40	[5]
Izmit Bay, Turkey	21.8	4.9	74.3	67.6	102	930	8	(Pekey, H., 2006)
Masan Bay, Korea	NA	1.24	67.07	43.4	43.97	206.26	56	[14]
Gironde Estuary, France	18.7	0.48	78.4	24.5	46.8	168	323	[24]
Andaman Island, India	NA	0.8-1	13-20	81–88	4–5	12-23	15	[28]
Class-1	20	0.5	80	35	60	150		CSBTS (2002)
Class-2	65	1.5	150	100	130	350		CSBTS (2002)
Class-3	93	5.0	270	200	250	600		CSBTS (2002)
BG	10	0.5	60	25	30	80		

NA not available

Class-1, Class-2, and Class-3 are the Marine Sediment Quality Standards (GB 18668–2002) issued by the China State Bureau of Quality and Technical Supervision (CSBTS, 2002). The background (BG) value was taken from the Chinese National Marine Sediment Category Standard (State Oceanic Administration, 2004)

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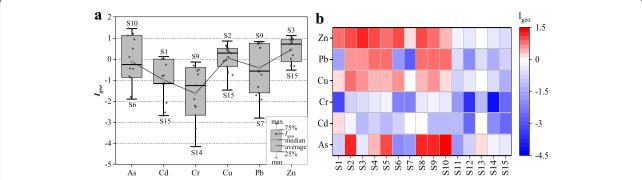
the outside of the bay, it was found that the contents of As, Cd, Cr, Pb, and Zn gradually decreased from the inside to the outside of the bay, while Cu remained at the same level and showed no obvious downward trend. Differences in the spatial distribution of heavy metals are largely related to their different sources or the sediment origin (texture, TOC, etc.). Different pollutants often contain different heavy metals. The possible sources of As, Cd, Cr, Cu, Pb, and Zn in the SZB sediments are the discharge of domestic sewage and industrial wastewater from Shenzhen and Hong Kong into the bay.

The heavy metal contents in the sediments of the SZB were also compared with those of other bay areas and coastal regions. In Table 1, which lists the heavy metal contents obtained in the study area along with those of other regions, it can be seen that the heavy metal contents in the SZB are slightly higher than those reported for the Yellow River, Yangtze River, and Pearl River Estuary. Furthermore, the levels obtained for Zhelin Bay and Jiaozhou Bay sediments were equal or lower. In addition, the heavy metal contents obtained in this study are significantly lower than those of the world's largest industrialised/urban ports and estuaries (Table 1), such as the Masan Bay in South Korea, Oyster Bay in Australia, Florida Bay in the United States, Izmit Bay in Turkey, and Gironde Estuary in France.

#### Assessment of heavy metal pollution in sediments

The I<sub>geo</sub> index can be used to confirm the pollution level of heavy metals in sediments. As shown in Fig. 3, the I<sub>geo</sub> index of heavy metals in the SZB sediments decreased in the order Zn>Cu>As>Pb>Cd>Cr. The average  $I_{geo}$ indices of Zn and Cu were 0.451 and 0.062, respectively, which indicate light pollution levels. The average  $I_{geo}$  indices of the other heavy metals were less than 0, indicating no pollution. For Cr, the  $I_{\rm geo}$  index was less than 0 in the sediments at all sampling points, indicating no pollution. As, Cd, Cu, and Pb showed  $I_{\text{geo}}$  indices greater than 0 at some sampling points. For example, the values for As at S10, S9, S8, and S2 indicated medium pollution, and those at S5, S13, and S4 light pollution; Cd exhibited only slight pollution at sampling point S1, with an  $I_{\rm geo}$  index of 0.12. The I<sub>geo</sub> of Cu at all sampling points indicated slight pollution, with a maximum value at S9 (0.59), and Pb also showed slight pollution at sampling points S3 (0.54), S5 (0.71), S10 (0.75), S9 (0.84), S8 (0.80), S4 (0.76), and S2 (0.54). Zn showed pollution conditions at all sampling points except S11, S12, S14, and S15, but all were below the moderate pollution level ( $1 \le I_{geo} < 2$ ). It is worth noting that the sampling points S10, S9, and S8 were polluted by all analysed heavy metals, except for Cd and Cr. Based on the I<sub>geo</sub> index, the SZB sediments were mostly polluted by Zn and Cu. Dai et al. [8] studied the Igeo indices of Pb, Cu, and Zn in the same sediments from 2000 to 2007, and found that the values all ranged between 0

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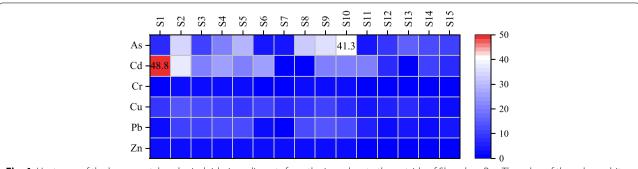


**Fig. 3** a Box and whisker plot of the  $I_{geo}$  indices of the six heavy metals analysed in the sediments of Shenzhen Bay (n = 15, a). The box represents the 25th to 75th percentiles,  $\square$  represents the average values, and the top and bottom horizontal lines represent the maximum and minimum values, respectively. **b**  $I_{geo}$  values in sediments of different sites from the inner to the outer of Shenzhen Bay

and 1, indicating slight pollution. Moreover, the level of pollution and heavy metal contents were lower at the bay mouth than inner bay, and both decreased gradually from the inside to the outside of the bay.

Heat maps have been widely used in studies of potential ecological risks of heavy metals. The colour scale ranges from blue to red and indicates increasing ecological risks [11]. The toxicity values in the environment are 30, 10, 5, 5, 2, and 1 for Cd, As, Cu, Pb, Cr, and Zn, respectively. The ecological risk of heavy metals in the SZB sediments is shown in Fig. 4. Except for As at S10 (41.3) and Cd at S1 (48.8), the potential ecological risks of the six heavy metals were all lower than 40, indicating low potential ecological risks. The comprehensive potential ecological hazards were found in the following order: RI  $_{As} = 274.05 > RI _{Cd} = 265.07 > RI _{Cu} = 126.87 > RI$  $_{Pb}$ =111.29>RI  $_{Zn}$ =32.96>RI  $_{Cr}$ =20.31. The results showed that As and Cd in the SZB sediments pose moderate potential ecological risks, implying that their pollution cannot be ignored; however, the overall potential ecological risks of the other heavy metals were relatively low. The moderate ecological risks of As and Cd are mainly due to their high ecological toxicity values; although their contents are relatively low, the ecological risk value is high. Therefore, special attention should be paid to the monitoring and management of As and Cd. In general, although the Zn contents in the sediments of the entire study area exceeded the background value, its potential ecological risk was only 32.96, which corresponds to a slight pollution level. These results are consistent with those of Zuo et al. [45], who used the ecological model evaluation method to analyse the heavy metals in SZB sediments in 2006 and showed that Cd had the largest potential ecological hazard coefficient. Comparing different sampling points, the comprehensive and individual ecological risks of heavy metals in the SZB sediments corresponded to the changes in the heavy metal content, and also showed a gradual decrease from inner bay to the outside of the bay.

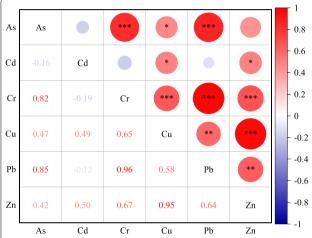
The distribution of the potential ecological risks showed that: (1) the risk of Shenzhen River Estuary was the lowest, and at a slight level. This is mainly attributed to the fact that the surface sediments in the estuary area are mainly silt and sandy silt with coarse particle sizes



**Fig. 4** Heat map of the heavy metal ecological risks in sediments from the inner bay to the outside of Shenzhen Bay. The value of the colour white is 40. Values less than 40 indicate that the heavy metal has a low potential ecological risk

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that have a small adsorption capacity for heavy metals, which resulted in a low degree of pollution. The concentrations of Cr, Pb, and Cu indicate no pollution. (2) The potential risk to the Shekou zone was at a medium level. Because this area is located in Shenzhen Bay Estuary, the hydrodynamic force was strong, and the surface sediments were easily disturbed; at the same time, this area was far away from Shenzhen city and there was no large river to flow into. Furthermore, the heavy metals that were transported in the water were deposited in the inner



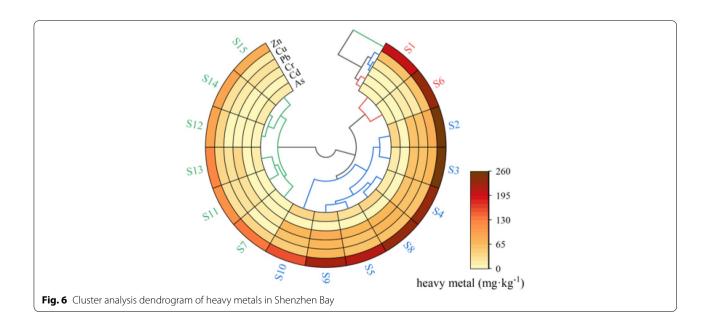
**Fig. 5** Correlation coefficient map of heavy metals in SZB sediments. Among them, red represents a positive correlation, blue represents a negative correlation, the number in the lower triangle represents the correlation coefficient, and the size of the circle in the upper triangle represents the size of the correlation coefficient

bay where the water exchange is weak. (3) The potential risks of the sampling points except Shenzhen River Estuary and Shekou area were all at a high level. These sampling points were all located inside the SZB where the Dasha, Xinzhou, and Fengtang rivers bring a large amount of heavy metals into the bay.

#### Source apportionment of heavy metals in sediments

The correlation between heavy metal contents can be used to evaluate the potential common source and migration processes of these elements. A high correlation between two elements indicates that they have a common source and similar migration process. The correlation coefficients between the heavy metals in the SZB sediments are shown in Fig. 5. Cr–Pb (0.96), Cu–Zn (0.95), As–Pb (0.85), As–Cr (0.82), Cr–Cu (0.65), and Cr–Zn (0.67) showed a strong significant correlation (p<0.01), while Pb–Cu (0.58) and Pb–Zn (0.64) showed a significant correlation (p<0.05). These results suggest that these pairs of metals may have the same pollution sources and migration processes.

CA is widely used to identify pollutant sources of heavy metals in sediments to distinguish natural from anthropogenic contributions (Loska and Wiechula, 2003; [19]. The clusters of six heavy metals obtained from the cluster analysis are presented by the relationship between groups of variables, as shown in Fig. 6, with a lower value on the horizontal axis showing a more significant association. There are three statistically significant clusters, among which cluster 1 only includes the heavy metals Zn that has been identified to have a light pollution degree from the  $\rm I_{\rm geo}$  index.



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Cluster 2 includes Cu, Pb, and Cr, and can be further divided into two sub-clusters because the distance between Cu and Pb–Cr was relatively long. Sub-cluster 1 includes Cr and Pb, which were shown to have no pollution and a slight pollution degree according to the  $I_{\rm geo}$  index, respectively, Sub-cluster 2 includes Cu, which was shown to have a slight pollution degree according to the  $I_{\rm geo}$  index. Cluster 3 includes Cd and As, which posed moderate potential ecological risks based on the PERI. The CA results are consistent well with the results from the Pearson correlation analysis.

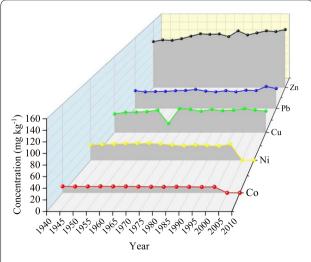
The correlation coefficients among Zn, Pb, Cr, and Cu were all higher than 0.5, indicating that these four elements may have similar sources and migration processes. The correlation coefficients between Zn and Cu, and Pb and Cr in particular were very high, 0.95 and 0.96, respectively, representing an extremely strong correlation. This may be because these elements are all sulfophilic elements; that is, these elements often form less soluble sulfides, which accumulate in water sediments. The correlations between As and Cd and other heavy metals were relatively low, indicating that there may be differences between their sources and those of the other elements in the sediment. For example, the source of As may be related to the transportation of fossil fuels, such as coal, in the SZB [4].

Heavy metals in sediments can originate from natural pollution sources or from human activities. Using the correlation between Fe and heavy metals, it was possible to distinguish between the natural and anthropogenic sources of the heavy metals. Naturally sourced heavy metals are often significantly correlated with Fe. Huang et al. [15] found that Pb, Cu, Zn, and Fe were relatively poorly correlated (Pb had a certain correlation with Fe at low concentrations), indicating that these heavy metals are related to anthropogenic pollution. The accumulation of historical heavy metals is an important cause of changes in the sedimentary environment of the SZB. During the rapid economic development of Hong Kong in the 1970s, industries in the northern New Territories developed rapidly, and a large amount of wastewater was discharged directly into the SZB or transported there via Shenzhen River. When entering the bay, the heavy metals were quickly incorporated into the sediments. Furthermore, the period from 1985 to 2000 was an era of economic rise in Shenzhen and other parts of the Pearl River Delta. A large amount of industrial wastewater, especially wastewater from electronics and electrical appliances, was polluted with heavy metals, which were directly discharged into the SZB. Heavy metals were further transported to the sea through the mouth of the SZB via the runoff and tidal currents of the Pearl River Estuary [25, 26].

#### Effect of land reclamation on heavy metals in sediments

Coastlines are formed through long-term evolution under various dynamic factors and are in a state of relatively dynamic equilibrium. Land reclamation activities are processes of creating new, dry land on the seabed, which changes the natural coastal pattern in a short time and on a small scale, and have a strong impact on the marine environment, causing an imbalance. Furthermore, the physical and chemical properties of the reclamation material itself may have a long-term impact on the heavy metal content in bay sediments.

Massive changes have occurred both on land and in the seawater in the SZB area since 1980. By 2005, the reclaimed land area had reached 1963 ha (Additional file 1: Figure S3). After 2005, however, no large-scale reclamation activities were carried out [7]. Nevertheless, land reclamation disrupted the dynamic balance of the original coastline, reduced the SZB area, led to a rapid decline in the tidal volume and tidal cycle of the bay, and created favourable conditions for sedimentation. The exchange capacity of the seawater was greatly reduced, resulting in pollutants remaining in the bay, especially the inner bay, for a long time. Some studies have shown that the average residence time of pollutants in the inner bay is approximately 10-14 days in the dry season, 8-9 days in the rainy season, 7-8 days near the Shenzhen-Hong Kong Western Corridor, and only 3–4 days in the outer bay [12]. In this case, the ability of seawater to dilute pollutants in upstream or coastal areas was greatly reduced, causing water pollution. Because the rivers entering the bay carry



**Fig. 7** Time series of the average heavy metal contents in the Shenzhen Bay surface sediments from 1940 to 2010. The data were taken from [20] and [34]

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a large amount of sediment, the longer the pollutants remain in the water, the greater the amount of pollutants deposited, which causes the pollution of sediments to increase. From 1940 to 2010, heavy metal concentrations were not significantly affected by land reclamation activities and remained stable (Fig. 7). If land reclamation activities created favourable conditions for sedimentation still needs to be answered, however, the heavy metal content has remained stable. In fact, the amount of heavy metal pollution entering into the SZB gradually decreased since the 1980s, and if there were no land reclamation activities, the heavy metal contents in the sediments would have decreased. Thus, the proportion of heavy metals entering the sediments actually increased due to the weak hydrodynamic conditions in the bay and, eventually, the heavy metal contents become relatively stable. Reports indicate that harmful substances (heavy metals and organic toxins) in the form of pollutants accumulate in water bodies or sediments, causing environmental disasters in the mangrove ecosystem along the SZB. Wang et al. (2010) reported that the release of sediment in the inner SZB had an impact of 25% on the water quality; the release of sediment pollution in the estuary bay also accounted for 8%. The pollution load in the sediments of the inner bay exceeded the capacity of the inner bay. With the reduction of land-based pollution from Shenzhen and Hong Kong, this proportion will continue to rise.

It is generally recognised that the filling materials used in the process of land reclamation may affect the heavy metal content of sediments. Chen and Jiao [7] conducted a chemical analysis of the reclamation materials used in a certain reclamation project in the SZB and showed that their heavy metal contents were much lower than those in the sediments (Table 2). Accordingly, it is reasonable to assume that the heavy metal contribution of the filler material to the SZB sediments is negligible. Therefore, it can be concluded that the heavy metals contents in the SZB sediments are probably related to historical land-source pollution from both sides of the bay, i.e., Shenzhen and Hong Kong.

#### **Conclusions**

In this study, six elements (As, Cd, Cr, Cu, Pb, and Zn) in SZB surface sediments were investigated. Based on this data, the main findings of this study are as follows:

- (1) The contents of major heavy metals in sediment were generally higher than the average background value in the offshore sediments of China; however, they were lower than the heavy metal contents in the coastal sediments of similar bay areas around the world, and the heavy metal content has gradually decreased over the last 20 years.
- (2) The pollution levels of the six heavy metals were found to be in the order of As>Cd>Cu>Pb>Zn>Cr. As and Cd were found to pose moderate ecological risks, and their potential hazard indices reached a high level, while the potential ecological risks of Cu, Pb, Zn, and Cr were found to be low.
- (3) The heavy metals Zn, Cu, Pb, and Cr were strongly correlated, which may be due to similar sources and migration processes. The accumulation of historical heavy metals was an important cause of changes in the sediment environment of the SZB, mainly owing to land-based pollution (such as industrial wastewater) from the 1970s to the end of the twentieth century.
- (4) Human activities, such as land reclamation, have led to a decline in the exchange capacity of water bodies in the SZB, resulting in heavy metals deposited in the surface sediments.

There is currently a lack of research regarding the biotransformation processes and carbon cycling effect the heavy metal contents and the distribution mechanism in Shenzhen Bay. Further studies should also explore this potential work.

Table 2 Comparison of heavy metal contents in filler materials and marine sediments of Shenzhen Bay (unit: mg·kg<sup>-1</sup>)

		As	Cd	Cr	Cu	Pb	Zn
Fill material (n = 11)	Mean	NA	0.20	8.08	8.30	39.61	32.32
	Average	NA	0.12-0.50	2.01-24.27	2.82-27.73	11.61-68.81	20.27-44.85
Marine mud	Mean	NA	0.40	75.96	34.63	49.45	119.25
(n = 22)	Average	NA	0.28-0.68	56.86-96.38	21.37-74.21	34.46-67.77	77.36-192.39

Data from the reference Chen and Jiao [7]

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#### **Abbreviations**

SZB: Shenzhen Bay; CA: Cluster analysis; ICP-OES: Inductively coupled plasma optical emission spectrometer;  $I_{geo}$ : Geo-accumulation index; PERI: Potential Ecological Risk Index.

#### **Supplementary Information**

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**Additional file 1: Table S1.** Geo-accumulation index (*lgeo*) classification. **Table S2.** Classification of the ecological hazard coefficient of heavy metal pollution. **Figure S3.** Statistics of land reclamation in the Shenzhen Bay in various periods.

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#### Authors' contributions

QZ and FR: conceptualisation, methodology, software, formal analysis, investigation, data curation, writing original draft, writing—review and editing; conceptualisation, formal analysis, investigation, supervision, project administration, funding acquisition; XX and HG: methodology, formal analysis, supervision, writing—review and editing; YW and WS: investigation, data curation, review; PL, ZL, and YB: investigation, data curation. All authors read and approved the final manuscript.

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#### Availability of data and materials

The datasets supporting the conclusions of this article are included within the article and its Additional file 1.

#### **Declarations**

#### Ethics approval and consent to participate

Not applicable.

#### Consent for publication

Not applicable.

#### Competing interests

The authors declare no competing interests.

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