Spatial distribution and environmental risk assessment of hazardous elements (As, Hg, Pb, Cd, Cu, Cr, Zn, and Se) in wetland soils of Yellow Sea coastal area, Eastern China

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ABSTRACT
The spatial distribution and environmental risk assessment of eight hazardous elements in wetland soils in Yellow Sea coastal area of China were investigated. The concentrations of As, Hg, Pb, Cd, Cu, Cr, Zn, and Se varied between 6.35-46.14, 0.004-0.05, 18.90-62.07, 0.17-3.76, 11.34-76.79, 21.52-80.88, 39.25-136.86, and 0.002-0.16 mg/kg, respectively. The spatial distributions analysis demonstrated that the high levels of As, Pb, Cd, and Zn were in Sheyang area. The results of correlation and PCA indicated that natural and anthropogenic source were major sources for Pb, Cr, Cu, Zn, and Se. As and Cd were from industrial and agricultural sources, and Hg was from natural source. The Cd indicated that 84% of sites experienced moderate and considerable contamination. The PER indices indicated that 68% of sites experienced considerable and high ecological risk. This study suggested that more attention should be paid to hazardous elements contamination of wetland soils in eastern China.

1. Introduction
Hazardous elements attract great attention due to their toxicity and cumulative effect [1, 2, 3, 4]. It is known that with the rapid economic development and industrialization, hazardous elements have been introduced into coastal environments, leading to toxicological effects on ecosystem and human health in the world [5, 6]. Hazardous elements, such as Cr, Pb, Zn, Cd, and Cu, can be detained in soils and sediments, and become a permanent threat to the coastal environment [7, 8]. Furthermore, As and Hg are considered as one of the most potentially hazardous elements in the global scale [9, 10]. Therefore, it is important to survey hazardous elements and evaluate environmental risks in coastal area [11, 12].

The tidal flat wetland in coastal area is regarded as a significant source and sink for hazardous elements and other pollutants [13, 14]. The tidal flat wetland of Yancheng is the largest typical muddy tidal flat wetland in China with the most complete ecological type. It locates in the Jiangsu Province along the coastline of Yellow Sea. The total area of Yancheng coastal beach is 4550 square kilometers. Two national nature reserves, Yancheng Rare Birds National Nature Reserve and Yancheng Milu National Nature Reserve, are in the Yancheng tidal flat, with many rare and endangered wild animals in the area. In recent years, the tidal flat has undergone great economic development including reclamation, fish and shrimp farming, and building industrial park. There are 6,000 tons of As and 30,000 tons of heavy metals discharged through rivers to the Yellow Sea, and 46,000 tons of heavy metal pollutants discharged through the sewage outlet to the Yellow Sea in Jiangsu Province [15]. Dafeng port and Sheyang port located in the Yancheng tidal flat wetland are the important national coastal harbour. The freight transport by mammoth tanker and large cargo boat can also cause hazardous elements pollution [16].

Several published studies have been conducted that the pollution and environmental risk assessment of heavy metals such as Cd, Zn, Pb, and Cu in some specific regions in Yancheng tidal flat wetland [17, 18], however, few studies investigated these eight selected hazardous elements (As, Hg, Pb, Cd, Cu, Cr, Zn, and Se), especially As, Hg, and Se [19]. In contrast, studies on Hg and Se in the Yancheng tidal flat wetland are rare. Furthermore, few studies concentrate on the comprehensive study including spatial distribution, concentration, source, and environmental risk assessment of hazardous elements in tidal flat soils along the coastline of Yancheng tidal flat wetland [20, 21], and the complex interactions among hazardous elements and other geochemical factors, such as Fe, Mn, and total organic carbon (TOC).

Therefore, the objectives of this study are to investigate the concentration and spatial distribution of eight hazardous elements (As, Hg, Pb, Cd, Cu, Cr, Zn, and Se) in tidal flat soil along the coastline of Yancheng...
tidal flat wetland, analyze the source of hazardous elements with the other geochemical factors (Fe, Mn and TOC), and assess the environmental risk of hazardous elements with comprehensive methods.

2. Materials and methods

2.1 Study area and sample collection

Soil samples were taken from the Yancheng tidal flat wetland in Jiangsu Province along the coastline of Yellow Sea, eastern part of China. The length of the coastline of Yancheng tidal flat wetland is 582 km, accounting for 61% of the coastline length of Jiangsu Province. A total of 38 surface soil samples were collected from the study area in 2016. Soil samples were collected from the upper layer (0 ~ 20 cm). The geographical coordinates of sampling sites were recorded with a hand-held GPS. The sampling locations are shown in Figure 1. The samples were placed in polypropylene bags, stored in coolers, and transferred to the laboratory. The soil samples were air-dried, then sieved through 2 mm nylon sieve. Sub-samples were passed through 0.15 mm sieves for hazardous elements analysis.

2.2 Chemical analysis

Samples were digested and analyzed for elements of As, Hg, Pb, Cd, Cu, Cr, Zn, Se, Fe, and Mn. Soils (about 0.5 g) were digested with HF–HNO$_3$–HClO$_4$ by the microwave (Tank, Hanon, China) [22]. The digested solution was passed through a 0.45 µm syringe filter and diluted to 25 mL. The As, Hg and Se concentrations in digestion solutions were determined by Atomic Fluorescence Spectrometer (AFS-PF52, Beijing Pu-Xi, China). The Pb, Cd, Cu, Cr, Zn, Fe, and Mn analysis were conducted using Atomic Absorption Spectrometer (AAS-TAS990, Beijing Pu-Xi, China). Chinese marine sediment standard (GBW07314) was analyzed to evaluate the recovery rate. The recoveries were between 94% and 109% for all the elements. TOC was analyzed using Potassium Dichromate Oxidation Spectrophotometric Method [23].

2.3 Assessment of hazardous elements contamination

The level of hazardous elements contamination in tidal flat soil was estimated with the geoaccumulation index ($I_{geo}$), contamination degree ($C_d$), and potential ecological risk (PER). The equation used for the calculation of $I_{geo}$ [24] is:

$$I_{geo} = \log 2 \left( \frac{C_n}{1.5B_n} \right)$$

Where $C_n$ is the measured concentration of element ‘$n$’, and $B_n$ is the background concentration of element ‘$n$’. The $B_n$ used in this study was the corresponding element background value in soil of Jiangsu Province (Table 1, Table S1). $I_{geo}$ was divided into seven levels: <0, practically unpolluted; 0–1, slight to moderately polluted; 1–2, moderately polluted; 2–3, moderately to strongly polluted; 3–4, strongly polluted; 4–5, strongly to extremely polluted; and >5, extremely polluted.

Furthermore, the contamination degree ($C_d$) is usually used to describe the hazardous elements contamination in soil [25]. The calculation equation is following:

![Figure 1. Location of sampling stations in the tidal flat of the Yancheng wetland.](image-url)
Table 1. The average concentrations and ranges of hazardous elements compared with data reported in other coastal areas.

| Element | Tidal flat area, Yellow Sea (n = 38) | Coast zone area, Yellow Sea (n = 30) | Tidal flat area, Yellow Sea (n = 90) | Coastal mud-flat area, Yellow Sea (n = 48, 2014) | Coastal mud-flat area, Yellow Sea (n = 48, 2009) | Bohai Sea (n = 405) | Laizhou Bay, Bohai Sea (n = 18) | Yangtze River Estuary (n = 860) | The Yellow River (n = 13) | Background of Jiangsu Province | Background of China | Chinese soil quality guideline Class I | Chinese soil quality guideline Class II | TEL guideline<sup>a</sup> | PEL guideline<sup>b</sup> |
|---------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|-------------------|--------------------------------|-------------------|-------------------|-------------------------|----------------|------------------|------------------|----------------|------------------|
| As      | 18.16 ± 9.79(6.35–46.14)         | 9.47(5.61–13.33)                 | 7.52(5.35–15.10)                 | 7.07(5.21–9.35)                  | 6.87(5.34–9.25)                  | 9.9(2.8–83.5)      | 7.14(6.65–9.65)                 | 9.23(3.50–19.30)   | 12.8(1.79–28.1)   | 9.4(3.96–21.67)  | 10.6(0.4)       | 1.5(0.15)              | 2.2(0.5)            | 7.24             | 41.6             |
| Hg      | 0.02 ± 0.01(0.00–0.05)           | 0.017(0.007–0.04)                | 0.013(0.006–0.034)               | 0.09(0.08–0.11)                  | 0.03(0.016–0.26)                  | 0.05 ± 0.02(0.01–4.1)| 0.04(0.02–0.05)         | 0.053(0.004–0.19) | 0.04(0.0–0.14)    | 0.03(0.03)     | 0.04(0.0)       | 0.13(0.0)             | 0.13(0.0)            | 0.13             | 0.7              |
| Pb      | 28.61 ± 8.57(18.90–62.07)        | 21.13(4.88–55.73)                | 13.91(8.14–26.40)                | 15.46(10.21–36.39)               | 14.07(10.09–32.48)                | 78.04(35.08–128.35)| 13.79(6.95–17.65)               | 22.72(13.5–48.2)   | 17.18(15.5–24.6)  | 7.27(5.20–16.7)  | 0.04(0.0)       | 0.43(0.01)             | 0.43(0.01)            | 0.13             | 28.2             |
| Cd      | 1.28 ± 0.79(0.17–3.76)           | 1.65(0.37–2.86)                  | 0.09(0.06–0.19)                  | 0.09(0.08–0.11)                  | 0.09(0.08–0.11)                  | 0.09 ± 0.02(0.00–1.3)| 0.09(0.02–0.05)         | 0.15(0.04–0.67)    | 0.12(0.27)        | 0.10(0.08–0.17)  | 0.10(0.0)       | 0.17(0.0)             | 0.17(0.0)            | 0.13             | 2.3              |
| Cu      | 29.35 ± 12.85(11.34–76.79)       | 37.99(5.28–96.80)                | 14.33(9.76–27.00)                | 6.3(0.26–36.39)                  | 18.4(10.09–32.48)                 | 37.95(24.20–51.07)| 10.99(7.57–21.29)               | 21.93(9.51–79.9)   | 17.34(14.1–30.3)  | 34.4(13.7–79.7)  | 37.95(24.20–51.07)| 22.42(19.9–74.6)                          | 18.420(29–29.8)                 | 77.8            | 31.7             |
| Cr      | 58.29 ± 15.12(21.52–80.88)       | 165.13(69.64–247.17)             | 58.30(40.70–88.00)               | 61.16(48.05–73.80)               | 59.17(53.92–71.07)                | 107.17(70.10–129.18)| 32.69(25.85–42.75)               | 76.06(42.89–301.0) | 81.3–139.5       | 18.4(10.09–32.48)| 107.17(70.10–129.18)| 22.42(19.9–74.6)                          | 59.75(29.17–73.80)               | 62.6            | 31.7             |
| Zn      | 75.7 ± 22.3(39.25–136.86)        | 96.13(69.64–247.17)              | 50.73(38.40–81.20)               | 76.06(42.89–73.80)               | 59.75(29.17–73.80)                | 313.74(179.34–472.75)| 32.69(25.85–42.75)               | 76.06(42.89–301.0) | 81.3–139.5       | 18.4(10.09–32.48)| 313.74(179.34–472.75)| 22.42(19.9–74.6)                          | 96.13(69.64–247.17)              | 77.8            | 31.7             |
| Se      | 0.08 ± 0.04(0.002–0.16)          | 0.06(0.03–0.12)                  | 0.03(0.016–0.26)                 | 0.09(0.08–0.11)                  | 0.09(0.08–0.11)                  | 0.09 ± 0.02(0.00–1.3)| 0.09(0.02–0.05)         | 0.15(0.04–0.67)    | 0.12(0.27)        | 0.10(0.08–0.17)  | 0.09 ± 0.02(0.00–1.3)| 0.12(0.27)                      | 0.09 ± 0.02(0.00–1.3)               | 0.09(2.3)      | 34.9             |
| Reference | 19                              | 29                              | 17                               | 18                               | 18, 41                           | 0.16(0.02–1.3)      | 0.2(0.02–0.05)         | 0.5(0.004–0.19)    | 0.5(0.0–0.14)     | 0.34(0.01–0.15)  | 0.16(0.02–1.3)  | 10                  | 42                  | 43                  | 44                  | 30                  |

<sup>a</sup>TEL guideline: threshold effect level (metal concentration in marine sediments below which adverse effects on biota are rarely observed).

<sup>b</sup>PEL guideline: probable effect level (metal concentration in marine sediments above which adverse effects on biota are frequently observed).
where \( C_d \) represents the sum of all contamination factors (\( C^i \)) in soil; \( C' \) represents the contamination factor of a single element; \( C_{\text{sample}} \) represents the element concentration of the sample, and \( C_i \) represents the background concentration of the element (Table S1). The contamination degree (\( C_d \)) can be divided into four levels: \( C_d < 8 \), low degree of contamination; \( 8 \leq C_d < 16 \), moderate degree of contamination; \( 16 \leq C_d < 32 \), considerable degree of contamination; \( C_d \geq 32 \), very high degree of contamination indicating serious anthropogenic pollution.

The potential ecological risk (PER) is used to assess the potential ecological risks caused by the hazardous elements in the ecosystem [25–27]. The RI represents the ecological risk index for various hazardous elements in soils. The calculation equation is following:

\[
RI = \sum_{i=1}^{m} E_r^i \\
E_r^i = T_r^i \cdot C_i^i \\
C_i^i = \frac{C_i}{C_n}
\]

Where \( T_r^i \) and \( C_i^i \) are the toxic-response factor and the contamination factor of hazardous element \( i \), respectively. \( E_r^i \) is the potential ecological risk coefficient of hazardous element \( i \). The toxic-response factors (\( T_r^i \)) of hazardous elements are shown in Table S1. The degree of ecological risk of hazardous elements in soil can be divided into the following levels [3b; 28]: \( E_r^i < 40 \), RI < 150, Low risk; \( 40 \leq E_r^i < 80 \), 150 < RI < 300, Moderate risk; \( 80 \leq E_r^i < 160 \), 300 < RI < 600, Considerable risk; \( 160 \leq E_r^i < 320 \), High risk; \( E_r^i \geq 320 \), RI > 600, Very high risk.

3. Results and discussion

3.1 Hazardous element concentrations in tidal flat soil

The statistics of hazardous elements concentrations of tidal flat soil are presented in Table 1. The average concentrations of As, Hg, Pb, Cd, Cu, Cr, Zn, and Se were 18.16, 0.02, 28.61, 1.28, 29.35, 58.29, 75.75, and 0.08 mg/kg, respectively. Compared with the background values, the average concentrations of As and Cd were greatly higher than the environmental background values of Jiangsu Province, and the average concentrations of Pb, Cu, and Zn were slightly above the environmental background values of Jiangsu Province, which indicated that the study area might be polluted considerably. The average concentrations of Hg, Cr, and Se were below the environmental background values of Jiangsu Province, indicating that these elements originated from natural sources and were affected by human activity negligibly. The average concentrations of these hazardous elements were below the Chinese soil quality guideline Class I, except for As and Cd. The average concentrations of As, Cd, and Cu were above the values of threshold effects level (TEL) and below the values of probable effects level (PEL), indicating that they were in a relatively contaminated status and might cause potential toxicity risks.

The concentrations of eight hazardous elements in this study and the previous studies of tidal flat area of Yellow Sea, and other coastal areas in the world are presented in Table 1. The concentrations of eight hazardous elements (As, Hg, Pb, Cd, Cu, Cr, Zn, and Se) varied between 6.35–46.14, 0.004–0.05, 18.90–62.07, 0.17–3.76, 11.34–76.79, 21.52–80.88, 39.25–136.86, 0.002–0.16 mg/kg, respectively. The average concentration of As was higher in this study than those reported from previous study in tidal flat area [17,19,29] (Figure S1), and was comparable to that data of Jobos Bay soil, Puerto Rico [30]. The average concentration of Cd was higher compared with the previous results from coastal mud-flat area of Yancheng [17,18], but was similar with the values reported by 29 (Figure S1), and was comparable to the data from estuarine wetland of Haizhou Bay which was polluted seriously by heavy metals, South Yellow Sea [31]. The average concentration of Pb and Zn in this study was found to be higher than the data reported in the previous study [17,18,29], but lower than that of Haizhou Bay [31]. Moreover, the average concentration of Hg and Se was comparable to that in tidal flat soils of coastal area in Yancheng [19]. The average concentration of Cr was similar with the previous studies [17,18], but was lower than the values reported by 29, from Yancheng tidal flat.

2.4 Data analysis

The principal component analysis (PCA) and Spearman’s rank correlation analysis were performed using SPSS 25.0 software. The redundancy analysis (RDA) was performed using CANOCO 5.0 software. Hierarchical clustering was performed with Origin 2017. Spatial distribution maps of hazardous elements were drew using geostatistical method of geographic information system (GIS) software (ArcGIS, version 9.3).
3.2 Spatial distribution of hazardous elements

The spatial distribution of hazardous elements is shown in Figure 2. As, Pb, Cd, Zn, Cu, Cr, and Se displayed similar distribution pattern: all of the elements presented high levels in Sheyang county area. Moreover, Zn, Cu, Cr, and Se also presented elevated levels in Xiangshui county area. The distribution of Hg was very different with other elements, and presented high levels in Dafeng county area.

The Fe, Mn, and TOC also play an important role on geochemical behavior of hazardous elements in soil and sediments [10,32]. The spatial distribution of Fe, Mn, and TOC is shown in Figure S2. The TOC values were in the range of 0.4–2.2% in the study area (Table S2). The spatial distribution of TOC presented elevated levels in Xiangshui and Sheyang area. Specifically, the distributions of Zn, Cu, and Se were similar with TOC, and the high concentrations of Zn, Cu, and Se were associated with higher TOC content. The spatial
distribution of Fe shared greater similarity with Zn, Cu, Cr, and Se, and presented high levels in Xiangshui and Sheyang area. The spatial distribution of Mn presented high levels in Xiangshui and Dafeng area.

Generally, the high levels of As, Pb, Cd, and Zn were observed in Sheyang area. The hotspots were observed in the papermaking industrial area of Sheyang (site 32, 33, 34, and 35). The similar spatial distribution trends of As, Pb, Cd, and Zn indicated that these hazardous metals might originate from the same source, causing by the discharge of papermaking industry. The distribution of Cu, Cr, and Se was similar with TOC, Fe, or Mn in Sheyang and Xiangshui area, indicating that the source might be the natural source or the accumulation of these hazardous elements in soils were regulated by similar mechanisms, such as complexation with organic content [10] and adsorption on Fe/Mn oxides in soils [32].

### 3.3 Correlation and principal component analysis

The correlation coefficient matrix including hazardous elements (As, Hg, Pb, Cd, Cu, Cr, Zn, and Se), major elements (Fe and Mn), and TOC is shown in Table 2. Pb, Cu, Cr, Zn, and Se had positive correlation with Fe (P < 0.05). Cr, Zn and Cu had positive correlation with Mn (P < 0.05). As, Pb, Cu, Zn, Se, and Fe had strong positive correlation with TOC (P < 0.05). Fe, Cu, and Zn was significantly correlated with each other (P < 0.05). Additionally, there was no elements which had correlation with Hg. The property of Hg is different from other hazardous metals. It is a liquid element at normal temperature and has strong mobility in the atmosphere, which usually shows a lower correlation with other elements [33].

Principal component analysis (PCA) can be used to identify possible sources for these hazardous elements. Three PCs were extracted, and accounted for 69.5% of the total variance (Table 3, Figure 3). PC1 accounted for 40.28% of the total variance, which was contributed by elements (Pb, Cr, Cu, Zn, Se, Fe, and Mn), and TOC (Figure 3a). The results indicated that PC1 mainly represented these elements generally had similar origin or accumulated by similar mechanisms. Compared with background values of Pb, Cr, Cu, Zn, and Se (Table 1), the results showed that Cr and Se had lower values than background values of Jiangsu Province, and Pb, Cu and Zn had higher values than background values of Jiangsu Province. In the spatial distribution maps (Figure 2), the high levels of these elements predominately distributed in Sheyang and Xiangshui area. In this study area, there are many industrial parks with economic development, including papermaking industry, aquatic product processing, biomass energy factory, and several important harbours, such as Sheyang ports. The discharged waste water and gas

### Table 2. Spearman rank correlation analysis for tidal flat soil components.

| Variables | As  | Hg  | Pb  | Cu  | Cr  | Cd  | Zn  | Se  | Fe  | Mn  | TOC |
|-----------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| As        | 1.00 |     |     |     |     |     |     |     |     |     |     |
| Hg        | −0.145 | 1.00 |     |     |     |     |     |     |     |     |     |
| Pb        | 0.234 | 0.096 | 1.00 |     |     |     |     |     |     |     |     |
| Cu        | 0.135 | 0.014 | 0.751* | 1.00 |     |     |     |     |     |     |     |
| Cr        | −0.534* | −0.009 | 0.221 | 0.347* | 1.00 |     |     |     |     |     |     |
| Cd        | 0.119 | −0.047 | 0.143 | 0.250 | −0.191 | 1.00 |     |     |     |     |     |
| Zn        | 0.280 | 0.094 | 0.703* | 0.910* | 0.398* | 0.348* | 1.00 |     |     |     |     |
| Se        | 0.000 | −0.111 | 0.158 | 0.347* | 0.210 | 0.152 | 0.376* | 1.00 |     |     |     |
| Fe        | 0.273 | 0.147 | 0.628* | 0.858* | 0.575* | 0.174 | 0.887* | 0.325* | 1.00 |     |     |
| Mn        | −0.034 | 0.054 | 0.279 | 0.447** | 0.350* | −0.237 | 0.432* | 0.161 | 0.565* | 1.00 |     |
| TOC       | −0.350* | −0.197 | 0.416* | 0.602* | 0.230 | 0.040 | 0.434* | 0.548* | 0.372* | 0.233 | 1.00 |

*Correlation is significant at the 0.05 level.

### Table 3. Loadings for principal component analysis of tidal flat soils.

| Variables | PC1 | PC2 | PC3 |
|-----------|-----|-----|-----|
| Pb        | 0.698 | −0.104 | −0.429 |
| Cr        | 0.614 | −0.555 | 0.268 |
| Cd        | 0.008 | 0.679 | 0.213 |
| Cu        | 0.871 | 0.209 | −0.034 |
| Zn        | 0.935 | 0.072 | −0.078 |
| As        | 0.039 | −0.803 | 0.337 |
| Hg        | 0.042 | 0.202 | −0.702 |
| Se        | 0.537 | 0.379 | 0.469 |
| Fe        | 0.911 | −0.067 | −0.088 |
| Mn        | 0.622 | −0.312 | −0.228 |
| TOC       | 0.651 | 0.376 | 0.346 |
| Eigenvalues | 4.430 | 1.900 | 1.314 |
| % Variance  | 40.28 | 17.28 | 11.94 |
| % Cumulative | 40.28 | 57.56 | 69.50 |
can cause Pb, Cu and Zn contamination [18]. In addition, the fishery industry including snail, fish and shrimp culture was developed in the study area, which also can lead to Pb and Zn pollution [16]. In addition, the RDA analysis indicated (Figure 3c), the TOC had positive correlation with Se and Cu, and Fe had positive correlation with Cr, Pb, Cu, and Zn. TOC, Fe, and Mn in this component might display a function in the accumulation of hazardous metals in the soils. Therefore, natural source and anthropogenic source might be a major source for PC1, and the anthropogenic source was primarily industry activity. PC2 accounted for 17.28% of the total variance, which was contributed by As and Cd. The average concentrations of As and Cd were considerably higher than the background values of Jiangsu Province (Table 1).

Figure 3. Principal component analysis (PCA) of hazardous elements, Fe, Mn and TOC in soils (a and b). The relationships between the geochemical factors (Fe, Mn and TOC) and hazardous elements were revealed by RDA (c).
Considering the spatial distribution of As and Cd (Figure 2), they had elevated levels in Sheyang area. Therefore, anthropogenic source was the major source for As and Cd. There was a hotspot in papermaking industrial area of Sheyang with the highest levels of As and Cd. Moreover, the agricultural activity, such as fertilization, can also lead to Cd and As contamination [34]. The inning and land use have been developed well in the tidal flat of Yellow Sea, therefore the phosphate fertilizer is common in the farmland. Phosphorus ore associated with Cd is the raw material for phosphate fertilizer production [35]. Moreover, the phosphate has competitive adsorption activity with As. It has been reported previously at Kandal in Cambodia that the extensive phosphate fertilizer had been applied in agricultural lands [36]. Thus, the industrial and agricultural activity might be the major source for PC2. PC3 with 11.94% of the total variance had significant loading on Hg (Figure 3c). Hg had no correlation with other elements, and the distribution was different from others (Table 2, Figure 2). The average concentration of Hg was comparable to that in tidal flat soil of coastal area in Yancheng (Table 1). Therefore, Hg was attributed to the natural source [37–39].

### 3.4 Assessment of the hazardous elements contamination

In order to estimate heavy metal contamination in the study region accurately, three evaluation methods were used. Then, the three evaluation results can be compared.

#### 3.4.1 Geoaccumulation index

The calculated $I_{geo}$ values of hazardous elements in soils are shown in Figure 4 and Table S3. The percentage of soil samples that had $0 < I_{geo} < 1$ was 53%, 16%, 8%, 3%, 32%, 0%, 18%, and 5% for As, Hg, Pb, Cd, Cu, Cr, Zn, and Se, respectively, which indicated slight to moderate contamination (Table S3). The percentage of soil samples that had $1 < I_{geo} < 2$ was 10%, 15%, and 3% for As, Cd, and Cu, respectively, which indicated moderate contamination. Only some samples for Cd had $2 < I_{geo} < 3$, $3 < I_{geo} < 4$, and $4 < I_{geo} < 5$, and the percentages were 63%, 8%, and 8%, respectively. Moreover, the $I_{geo}$ ranged from −1.15 to 1.71 (average 0.20) for As, −7.97 to 0.31 (average −1.22) for Hg, to 1.06 to 0.66 (average −0.51) for Pb, to −0.16 to 4.32 (average 2.53) for Cd, −1.56 to 1.20 (average −0.31) for Cu, to −2.44 to −0.53 (average −1.06) for Cr, to −1.26 to 0.54 (average −0.36) for Zn, and to −5.70 to 0.18 (average −1.18) for Se (Figure 4). The average values of $I_{geo}$ decreased in the order of Cd > As > Cu > Zn > Pb > Cr > Se > Hg. The average $I_{geo}$ values of Cd pointed to moderately to strongly polluted, while the average $I_{geo}$ values of As indicated slight to moderately polluted. The average $I_{geo}$ values obtained for Hg, Pb, Cu, Cr, Zn, and Se showed that the study soils were unpolluted. Overall, based on the $I_{geo}$ values, the study area was polluted with Cd and As.

#### 3.4.2 Contamination degree

The spatial distribution of contamination degree ($C_d$) is presented in Figure 5. The $C_d$ values indicate a comprehensive pollution condition of the eight hazardous elements in soils. In Figure 5, Sheyang was the most contaminated area, and Xiangshui and Dafeng area was lesser contaminated. The results of $C_d$ values showed that 5% of soil samples had values of $C_d < 8$, indicating low degree of contamination, 45% of soil samples had values of $8 \leq C_d < 16$, indicating moderate degree of contamination, 39% of soil samples had values of $16 \leq C_d < 32$, indicating considerable degree of contamination, and 11% of soil samples had values of $C_d > 32$.

![Figure 4](image-url)  
Figure 4. Box-plots of geoaccumulation index ($I_{geo}$) for hazardous elements in tidal flat soil.
≥32, indicating a serious anthropogenic pollution in the area. Four sites in Sheyang industry area had high $C_d$ values (34, 42, 41, and 38), which indicated severe pollution condition.

### 3.4.3 Potential ecological risk

The single potential ecological risk index ($E_i^r$) of hazardous elements in tidal flat is shown in Table 4 and Figure 6. The $E_i^r$ values of Hg, Pb, Cu, Cr, and Zn in all

|        | As  | Hg  | Pb  | Cd  | Cu  | Cr  | Zn  |
|--------|-----|-----|-----|-----|-----|-----|-----|
| Mean   | 19.32 | 27.00 | 5.46 | 305.16 | 6.58 | 1.50 | 1.21 |
| Range  | 6.76–49.09 | 0.24–38.56 | 0.72–2.37 | 40.40–895.89 | 2.54–17.22 | 0.55–2.08 | 0.63–2.19 |
| <40    | 34  | 38  | 38  | 0   | 38  | 38  | 38  |
| 40–80  | 4   | 0   | 0   | 2   | 0   | 0   | 0   |
| 80–160 | 0   | 0   | 0   | 4   | 0   | 0   | 0   |
| 160–320| 0   | 0   | 0   | 23  | 0   | 0   | 0   |
| >320   | 0   | 0   | 0   | 9   | 0   | 0   | 0   |

Figure 6. Hierarchical cluster analysis of the potential ecological risk index ($E_i^r$) of hazardous elements in soil samples.
soil samples were less than 40, suggesting that the risk levels for these elements are low. Most $E_i^\text{v}$ values of As were less than 40, but there were four soil samples of As ranged from 40 to 80. The results suggested that As contamination in most areas in tidal flat of Yellow Sea was not very serious. However, at four sample sites (site 32, 33, 34, 35), As contamination reached moderate ecological risk. In contrast, Cd had the highest $E_i^\text{v}$ average value (305.16), which exhibited high ecological risk. All of the $E_i^\text{v}$ values for Cd were more than 40 in the study region, and there were 60% of samples in the range of high ecological risk levels. Even 23% of samples reached very high ecological risk. Meanwhile, hierarchical clustering analyses separated the $E_i^\text{v}$ of 38 soil samples into two groups (Figure 6). The first group contained four samples [site 32–35], which were characterized by relatively high $E_i^\text{v}$ values of As and Cd. The remaining 34 soil samples were grouped together, and were characterized by relatively low $E_i^\text{v}$ values of As and Cd. Concurrent with the present study, the previous studies observed similar findings. Studied the heavy metal pollution of soils in Yancheng coastal zone, and found that there was 66.3% of $E_i^\text{v}$ values for Cd reached high and very high ecological risk [40].

The RI values were calculated to assess the contamination of hazardous elements in tidal flat soils (Figure 7). RI values of soil samples (32% of the soil samples) were lower than 300, indicating low ecological risk. However, RI values for most soil samples (68% of the soil samples) were more than 300, which suggested that most sample sites experience considerable and high ecological risk of hazardous elements. RI values of sampling sites (site 32, 33, 34, and 35) were higher than 600, accounting for 11% of the soil samples, which indicated the very high ecological risk. Therefore, frequent anthropogenic behavior was considered to be responsible for the highest ecological risk from hazardous elements contamination.

Compared with three evaluation methods, the contamination level of hazardous elements and pollution degree of samples were consistent in general. The $I_{geo}$ assessment for single elements showed that there were moderate to strong Cd pollution and slight to moderate As pollution. The comprehensive assessment of $C_d$ and RI for the eight hazardous elements in tidal flat soils of Yancheng indicated that 84% of sample sites had a moderate and considerable degree of contamination, and 68% of sample sites experienced considerable and high ecological risk.

4. Conclusions

The spatial distribution and environmental risk assessment of As, Hg, Pb, Cd, Cu, Cr, Zn, and Se in tidal flat soils of the Yellow Sea coastal area have been investigated. The average concentration of As, Cd, Pb, Cu, and Zn was higher than the environment background values of Jiangsu Province. The average values of As, Cd, and Cu concentration were above their corresponding values of TEL, indicating they were in a relatively contaminated status and might cause potential toxicity risks. The spatial distribution of As, Hg, Pb, Cd, Cu, Cr, Zn, and Se was higher in Sheyang and Xiangshui county of Yancheng City. PCA results

![Figure 7. Spatial variation in the ecological risk indices (RI) of hazardous elements in tidal flat soil. The dotted lines represented the RI values were 150 (limit value for low risk), 300 (limit value for moderate risk) and 600 (limit value for very high risk).](image-url)
showed that natural source and anthropogenic source may be a major source for Pb, Cr, Cu, Zn, and Se, and the anthropogenic source was industry activity primarily. As and Cd was from industrial and agricultural source, and Hg was from natural source.

The environment risk assessment approaches including $I_{geo}$, $C_d$, and $RI$ were used in this study. The $I_{geo}$ values revealed that the order of $I_{geo}$ was Cd $>$ As $>$ Cu $>$ Zn $>$ Pb $>$ Cr $>$ Se $>$ Hg. Higher $I_{geo}$ values for Cd and As in study soils indicated that there were moderate to strong Cd pollution and slight to moderate As pollution. The calculated $C_d$ showed that 45% of sample sites had a moderate degree of contamination, 39% of sample sites had a considerable degree of contamination, and 11% of sample sites had serious anthropogenic pollution in the area. $RI$ values for 68% of soil samples experienced considerable and high ecological risk. Among these eight hazardous elements, Cd and As are most concerning in terms of their contamination degree. Recently, the heavy metal contamination in coastal soil is more serious, and the cause and spatial distribution should be further revealed. The study area is the largest typical muddy tidal flat wetland in China. The results in this study indicate that more attention should be paid to hazardous elements contamination of tidal flat soil along the coastal area. Scientific management measures should be performed to protect the wetland ecosystem for the local government.

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