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Distribution and assessment of heavy metals in surface sediments from the Bohai Sea of China

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ABSTRACT

In this study, we analyzed heavy metals in 404 surface sediment samples from the Bohai Sea to measure contamination status and distribution. We found Zn levels to be the highest, whereas Hg concentrations were the lowest of measured heavy metals. We found that the samples containing the most heavy metals were those collected from Fuzhou Bay, Jinzhou Bay, central Bohai Sea mud area, and the Yellow River Delta. Further analyses suggest that these heavy metals in surface sediments in the Jinzhou Bay and Fuzhou Bay pose a serious ecological risk, with substantial Cd and Hg accumulation in the Jinzhou Bay and Yellow River Delta regions being indicative of intense human activities.

Presently, the heavy metal contamination of sediment poses a serious and global environmental challenge that must be urgently addressed (Nriagu, 1996; Pacyna and Pacyna, 2001; De Vleeschouwer et al., 2007; Zhang and Shan, 2008; Islam et al., 2015). Marine sediments serve as a key sink for heavy metals that are released into the sea as a consequence of human actions, regardless of whether it is direct or indirect release (Neto et al., 2000; Yuan et al., 2004). As these heavy metals accumulate, they become increasing enriched in surface and subsurface sediments, thus posing a serious risk to marine life (Park et al., 2011; Liu et al., 2014; Vukosav et al., 2014). It is therefore essential that the levels and sources of heavy metal contamination be clarified in order to better manage marine ecosystems (Hosono et al., 2011; Naser, 2013).

The Bohai Sea has a mean depth of ~18 m and is approximately ~80,000 km^2 in area (Liu et al., 2011). The sea is composed a largely enclosed area, with distinct sections including Liaodong Bay, Bohai Bay, Laizhou Bay, Central Sea Basin, and Bohai Strait (Fig. 1). The Bohai Sea is surrounded by the Shandong Peninsula and the Liaodong Peninsula, with Bohai Strait serving as a path into the Yellow Sea.

As industrialization and urbanization have been progressing at an extremely rapid rate in recent decades in the Bohai Sea region, the increasing quantities of compounds released into this sea because of human activities pose a serious environmental risk to this ecosystem (Wei et al., 2008; Xu et al., 2009; Pan and Wang, 2012; Zhao et al., 2014; Liu et al., 2015). The cities of Dalian, Yingkou, Tianjin, and Dongying in particular are near the Bohai Sea and are sites of extensive chemical production (Duan and Li, 2017). The total combined runoff and sediment flux of the Yellow River, Liao River, Hai River, and Luan River reach up to $637.4 \times 10^8\, m^3$ and 1361×10^6 tons, respectively, thus potentially releasing large quantities of heavy metals into this sea through the transfer of contaminated water and sediment (Table 1). As the Bohai Sea exchanges only limited quantities of water with the open sea, these heavy metals tend to accumulate and have resulted in the Bohai Sea being the most polluted sea in China (Duan and Li, 2017).

Several previous studies have highlighted heavy metal contamination of surface sediments in specific regions within the Bohai Sea (e.g., Wei et al., 2008; Wang et al., 2010; Pan and Wang, 2012; Hu et al., 2013; Liu et al., 2015; Duan and Li, 2017). These reports have indicated that the heavy metal contamination of Bohai Bay and northeastern Liaodong Bay (Jinzhou Bay) is especially pronounced (Wang et al., 2010; Hu et al., 2013), with additional alarming levels of contamination in the intertidal zones of the Yellow and Liao River Deltas because of human activities (Zhang et al., 2016; Yan et al., 2018). However, these past studies have not conducted simultaneous surveys of heavy metal contamination throughout the entire the Bohai Sea. Therefore, in the present study, we provide an overview of heavy metal distributions throughout the Bohai Sea based upon 404 samples of surface sediments that were collected and analyzed by identical approaches. We further explored the ecological risks, geoaccumulation indices, and enrichment factors associated with these heavy metal levels in the Bohai Sea.

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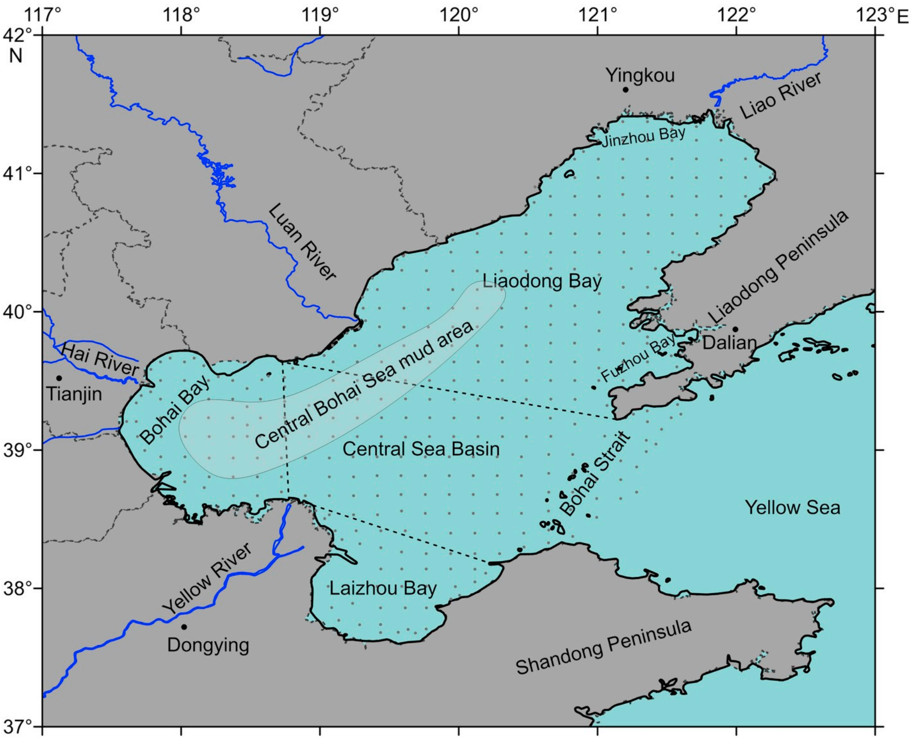

Fig. 1. Surface sediment sample sites (gray circles) in the Bohai Sea.

Table 1
Hydrology of rivers draining into the Bohai Sea.

River	Length (km)	Drainage area (10^4 km^2)	Runoff (10^8 m^3)	Sediment flux (10^6 t)	References
Yellow River	5464	75.22	464	1322	(Chen, 1996)
Liao River	1345	19.95	27.5	8.89	(Wang and He, 1993)
Hai River	–	21.11	98.7	8.03	(Wen and Xing, 2004)
Luan River	877	4.45	47.2	22.19	(Feng and Zhang, 1998)

We used a box sampler to collect 404 distinct surface sediment samples collected across different regions of the Bohai Sea. Following collection, samples were marked with a microchip, sealed, frozen at −20 °C, and transferred to a laboratory for the analysis of surface sediments. 30% H_2O_2 and 10% HCl had been used to remove organic and calcareous matters in each sample. Afterwards, the sample has been centrifugated for three times to remove the salt. A laser particle size analyzer (Malvern-2000, UK) was used for measuring sediment grain sizes in duplicate, with differences between measurements at < 3%. We next conducted grain size parameter calculations according to formulas published by Folk and Ward (1957).

Al_2O_3 and Fe_2O_3 were measured by inductively coupled plasma optical emission spectrometry (ICP-OES, Thermo iCAP-6300, US). Copper, lead, zinc, cadmium, chromium, and nickel were measured by inductively coupled plasma mass spectrometry (ICP-MS, Thermo X-II series, US). An atomic fluorescence spectrometer (AFS-920) was used for As and Hg measurements. For quality assurance and control purposes, we used China Stream Sediment Standards (GBW07309, GBW07313, GBW07314, GBW07316) and appropriate blanks. In addition, we selected 10% of analyzed samples at random and repeated the analyses thereof until variations between measurements were < 5%.

We found that surface sediments had a mean grain size (Mz) from −0.42 to 7.69 Φ (mean: 5.34 Φ; Table 2). The mean surface sediment heavy metal concentrations detected were, in descending order: Zn > Cr > Ni > Pb > Cu > As > Cd > Hg. Heavy metal concentration ranges, in μg/g, were as follows: 5.61 to 110.60 (mean: 57.95) for Cr, 6.06 to 129.60 (mean: 66.15) for Zn, 2.89 to 48.25 (mean: 25.42) for Ni, 2.22 to 38.94 (mean: 19.99) for Cu, 0.02 to 0.48 (mean: 0.20) for Cd, 8.93 to 37.67 (mean: 24.03) for Pb, 2.75 to 15.97 (mean: 9.18) for As, and 0 to 0.13 (mean: 0.04) for Hg.

The mean heavy metal concentrations in different regions of the Bohai Sea are shown in Table 2. We found that the mean Cd, As, Cu, Ni, Cr, Zn, and total heavy metals in Bohai Bay were higher than those in the other analyzed regions. In addition, the mean levels of Hg in Laizhou Bay and Pb in Liaodong Bay were substantially higher than those in the other tested regions. Furthermore, Bohai Strait contained the lowest concentrations of all tested heavy metals with the exception of Pb, the levels of which were lowest in Laizhou Bay.

We found that the spatial distributions of Cu, Pb, Ni, Cr, and Zn were similar (Fig. 2). Levels of these metals were highest in Bohai Bay, areas of fine-grain sediment, northwestern Laizhou Bay (YRD), northwestern Liaodong Bay (Jinzhou Bay), and southeastern Liaodong Bay (Fuzhou Bay). Levels of these metals were relatively low in the Qinhuangdao-Caofeidian offshore area, the tidal sand ridge of Liaodong Bay, and the Bohai Strait. In contrast, the spatial distributions of Cd, Hg, and As were distinct from each other and from those of other analyzed heavy metals. Cd concentrations were high in Jinzhou Bay and Fuzhou Bay, while Hg concentrations were high in Jinzhou Bay and Laizhou Bay, and As levels were high in Bohai Bay and Laizhou Bay.

We found that all heavy metal concentrations were significantly correlated with Mz, Al_2O_3, and Fe_2O_3 (Table 3). Pearson's correlation coefficients between heavy metals and Fe_2O_3 were higher than for those between heavy metals and Mz or Al_2O_3. The weakest correlation was between Cd and Fe_2O_3 (< 0.40), while those between Cr, Zn, Ni, Cu,

Table 2
Mean concentrations (μg/g) of heavy metals in surface sediments from different geomorphic regions in the Bohai Sea.

Regions	Cd	Hg	As	Cu	Pb	Ni	Cr	Zn	Sum
Liaodong Bay	0.23	0.04	8.13	18.49	24.86	23.45	53.04	64.74	193.02
Bohai Bay	0.25	0.03	11.81	28.02	24.34	33.00	72.36	87.63	257.50
Laizhou Bay	0.13	0.05	11.47	18.59	20.74	26.72	61.44	57.20	196.38
Bohai Basin	0.17	0.04	9.70	21.14	24.00	27.07	62.08	66.83	211.07
Bohai Strait	0.14	0.03	7.72	14.28	22.01	18.89	46.88	48.80	158.76
Bohai Sea	0.20	0.04	9.18	19.99	24.03	25.42	57.95	66.15	202.96

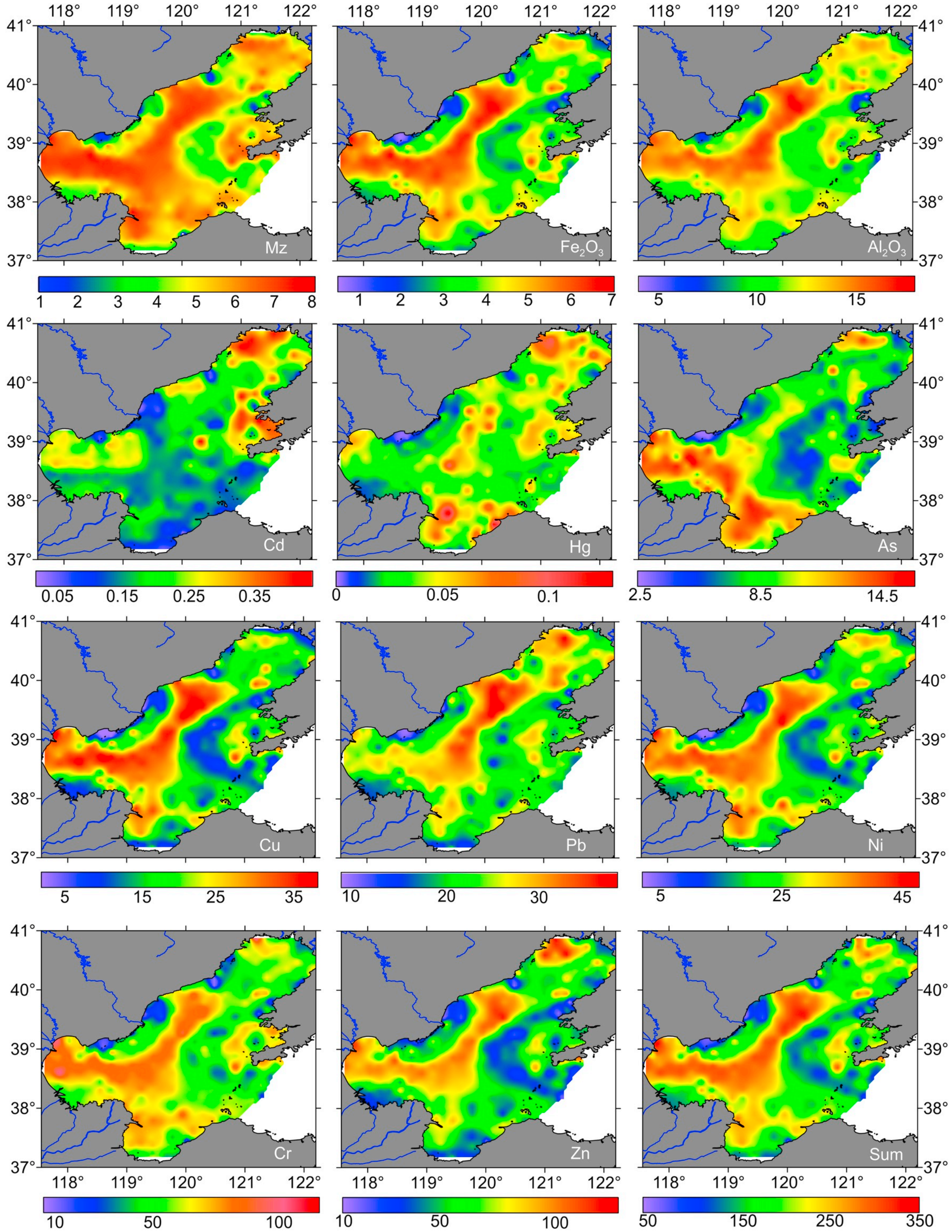

Fig. 2. Spatial distribution of Mz (Φ), Fe_2O_3 (%), Al_2O_3 (%), and heavy metals (μg/g)in the surface sediments from the Bohai Sea.

Table 3
Pearson's correlation coefficients for relationships between Mz, major oxides (Al_2O_3 and Fe_2O_3), and heavy metals concentrations in the surface sediments from the Bohai Sea (n = 404).

	Mz	Al_2O_3	Fe_2O_3	Cr	Zn	Ni	Cu	Cd	Pb	As	Hg
Mz	1.00										
Al_2O_3	0.80**	1.00									
Fe_2O_3	0.91**	0.89**	1.00								
Cr	0.87**	0.76**	0.92**	1.00							
Zn	0.86**	0.87**	0.93**	0.83**	1.00						
Ni	0.91**	0.84**	0.96**	0.92**	0.91**	1.00					
Cu	0.89**	0.87**	0.97**	0.87**	0.94**	0.95**	1.00				
Cd	0.36**	0.32**	0.34**	0.41**	0.41**	0.31**	0.32**	1.00			
Pb	0.70**	0.79**	0.79**	0.63**	0.83**	0.75**	0.80**	0.29**	1.00		
As	0.59**	0.52**	0.67**	0.63**	0.60**	0.70**	0.66**	0.10*	0.44**	1.00	
Hg	0.46**	0.38**	0.41**	0.44**	0.48**	0.45**	0.41**	0.37**	0.48**	0.31**	1.00

* $P < 0.05$.
** $P < 0.01$.

and Fe_2O_3 were > 0.90.

We next sought to assess how potentially harmful these heavy metals may be to ecosystems by calculating a heavy metal potential risk index (RI) (Hakanson, 1980) using the following equations:

$$C_f^i = \frac{C^i}{C_n^i} \tag{1}$$

$$E_r^i = T_r^i \cdot C_f^i \tag{2}$$

$$RI = \sum_{i=1}^{m} E_r^i \tag{3}$$

where $C_f^{\,i}$, $T_r^{\,i}$, and $E_r^{\,i}$ are the pollution coefficient, toxicity coefficient, and potential ecological risk for i heavy metals. C^i is the measured heavy metal level in sediment. $C_n^{\,i}$ is the reference value, for which we selected the maximum heavy metal levels in Chinese Sea sediments prior to industrialization (Zhao and Yan, 1993). *RI* is the sum of potential ecological risk for all heavy metals.

Based on this analysis, the potential ecological risk associated with each analyzed metal was, in rank order: Cd > Hg > As > Cu > Pb > Ni > Cr > Zn. Cd was associated with a mean potential ecological risk of 92.53, indicating a very strong potential risk, while Hg levels were consistent with a moderate ecological risk (mean value: 61.39). All other tested metals were associated with a slight potential ecological risk. Overall RI value ranged from 37.30–402.18, with a mean of 186.01.

In most tested regions, the potential Cd-associated ecological risk was deemed to be moderate (Fig. 3), with only samples in the Liugu River Delta, Luan River Delta, northern Caofeidian, and Yellow River Delta (YRD) regions being associated with a slight potential ecological risk. Notably, 55% of sampled sites had Cd levels with a high potential ecological risk, with these sites primarily being located in Liaodong Bay, northern and southeastern Bohai Bay, and northwestern Laizhou Bay. In addition, 6% of collected samples had Cd levels associated with an extremely strong potential ecological risk, including samples from Jinzhou Bay and Fuzhou Bay. Most collected samples throughout the Bohai Sea exhibited a moderate Hg-associated potential ecological risk. Approximately 23% of samples had Hg levels consistent with a strong potential ecological risk, with most of these samples being distributed in Jinzhou Bay, Fuzhou Bay, northwestern Bohai Bay, northern Laizhou Bay, and the region north of the fine-grained sediment area. A total of 7 samples had Hg levels consistent with an extremely strong potential ecological risk. Fortunately, ecological risk associated with Cr, Cu, Pb, Zn, Ni and As individually was deemed to be slight in the tested areas. However, 67 samples had total heavy metal levels consistent with a medium risk index value, and 4% of samples had a strong total heavy metal risk index consistent with significant ecological risk in the Jinzhou Bay region.

Muller (1979) proposed the use of the geoaccumulation index (I_{geo}) to gauge heavy metal contamination as follows:

$$I_{\text{geo}} = \log_2(C_n/k \times B_n) \tag{4}$$

Based on these calculations, we found that mean geoaccumulation index values for the tested metals were, in descending order: Cd > Hg > Cu > Pb > As > Ni > Zn > Cr. Cd had a mean geoaccumulation index of 0.91, which is near the range that would be classified as a medium level of pollution ($1 < I_{\text{geo}} < 2$; Muller, 1981). The mean geoaccumulation indexes for the other tested metals were all < 0, which is consistent with a nonpolluted state ($I_{\text{geo}} \leq 0$).

We found that a total of 48% of samples had Cd geoaccumulation indexes that were between 0 and 1, indicating that they were in the nonpolluted to moderately polluted range (Fig. 4). A total of 39% of samples had Cd geoaccumulation indexes between 1 and 2, consistent with a moderate degree of pollution. In addition, 15 samples that were primarily from Jinzhou Bay had geoaccumulation index values > 2, which is consistent with a moderate-to-strong level of pollution. Samples with Hg bioaccumulation indexes consistent with moderate pollution were mainly distributed in Jinzhou Bay and YRD. In addition, As levels in samples from Jinzhou Bay, central Bohai Bay, and YRD were in the non- to moderately polluted range (0–1), whereas all other tested samples had values < 0 (nonpolluted).

Enrichment factors can be used to effectively estimate the degree of heavy metal contamination and to distinguish between potential heavy metal sources (Liu et al., 2016). Aluminum or aluminum oxide is the element that is most frequently used in order to reduce grain size-associated effects in such analyses (Hu et al., 2013; Liu et al., 2014; Liu et al., 2016; Wei et al., 2008). Eq. (5) was used to calculate this index:

$$EF = (Me \div Al2O3)_{\text{sample}} \div (Me \div Al2O3)_{\text{background}} \tag{5}$$

where $(Me \div Al_2O_3)_{\text{sample}}$ is the measured ratio of metal between aluminum oxide, and $(Me \div Al_2O_3)_{\text{background}}$ is the unpolluted background ratio of metal to aluminum oxide. Based on heavy metals in Liaodong Bay, Lan et al. (Lan et al., 2018) proposed a more detailed EF representation, with EF < 1, EF = 1–2, EF = 2–5, and EF = 5–20 corresponding to no enrichment, minor enrichment, moderate enrichment, and severe enrichment, respectively.

Based on these calculations, measured heavy metal mean enrichment factors were, in descending rank order: Cd > Hg > Cu > Pb > As > Ni > Cr > Zn (Table 4). The mean enrichment factor values for Ni, Cr, and Zn were < 1 consistent with no enrichment, whereas those for Hg, Cu, Pb, and As were between 1 and 2 consistent with minor enrichment, and Cd had an index value of 2.86 consistent with moderate enrichment. The maximum measured enrichment factor was 10.88 for Cd, which is consistent with severe enrichment factor. The maximum Zn enrichment factor was 1.24. These suggested that human activity was likely to be responsible for surface

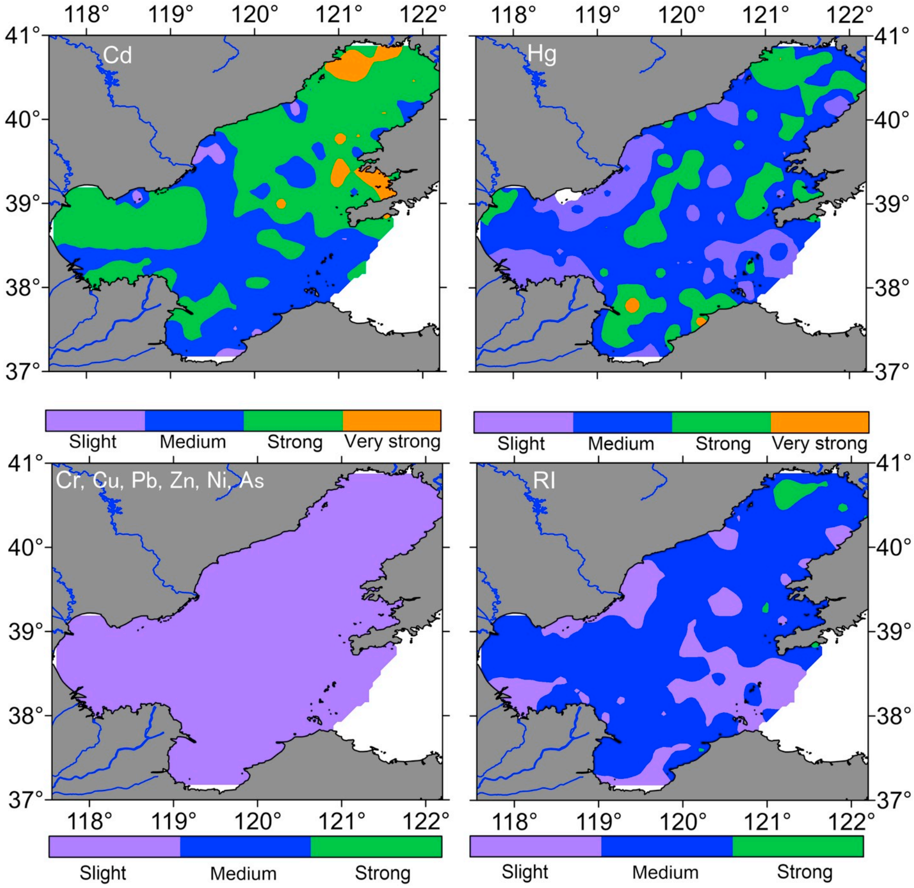

Fig. 3. Spatial distributions of potential ecological risk for heavy metals in the surface sediments from the Bohai Sea.

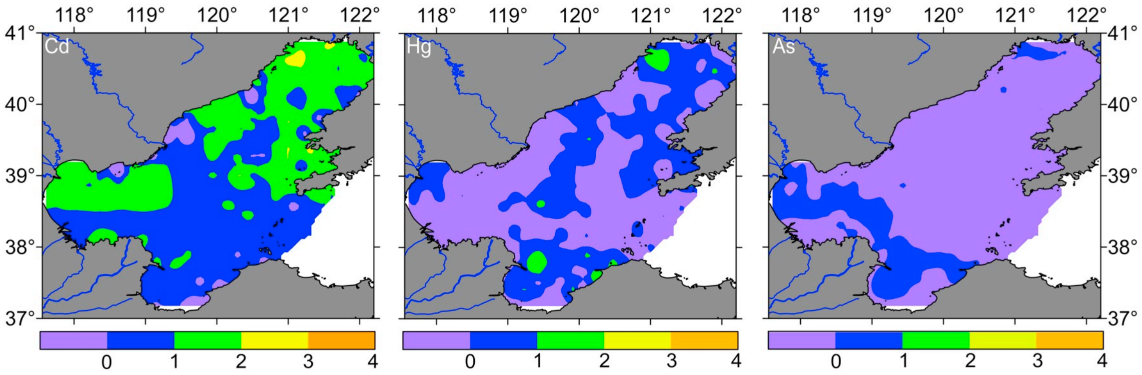

Fig. 4. Geoaccumulation index (Igeo) spatial distributions for heavy metals in the surface sediments from the Bohai Sea. Index values are not shown for heavy metals if they were < 0, consistent with a nonpolluted state.

Table 4
Parameters of enrichment factor of heavy metals in surface sediments from the Bohai Sea.

Parameter	Cd	Hg	Cu	Pb	As	Ni	Cr	Zn
Minimum	0.51	0.10	0.32	0.62	0.37	0.20	0.09	0.12
Maximum	10.88	4.79	3.28	2.83	2.43	2.71	2.48	1.24
Mean	2.86	1.41	1.18	1.11	1.10	0.94	0.86	0.44

sediment heavy metal contamination in the Bohai Sea.

Further analyses suggested that the samples most severely enriched for Cd were collected from the Jinzhou Bay and Fuzhou Bay regions (Fig. 5). Samples with moderate levels of Hg enrichment were collected primarily from Jinzhou Bay, Fuzhou Bay, and northern Laizhou Bay, while those moderately enriched for As were collected from southern Bohai Bay, Laizhou Bay, Jinzhou Bay, Fuzhou Bay, and northern Bohai Strait. There was only minor Cu and Pb enrichment in the majority of samples, with only a small number of samples exhibiting moderate levels of enrichment. Levels of Ni were minorly enriched in about half of samples, while Cr and Zn were not enriched in most samples.

In the present report, we constructed spatial distributions of heavy metal concentrations in surface sediments from the Bohai Sea. The mean concentrations of heavy metals were, in μg/g: Zn (66.15) > Cr (57.95) > Ni (25.42) > Pb (24.03) > Cu (19.99) > As (9.18) > Cd (0.20) > Hg (0.04). The potential ecological risk associated with detected Cd and Hg levels was high in Jinzhou Bay and Fuzhou Bay,

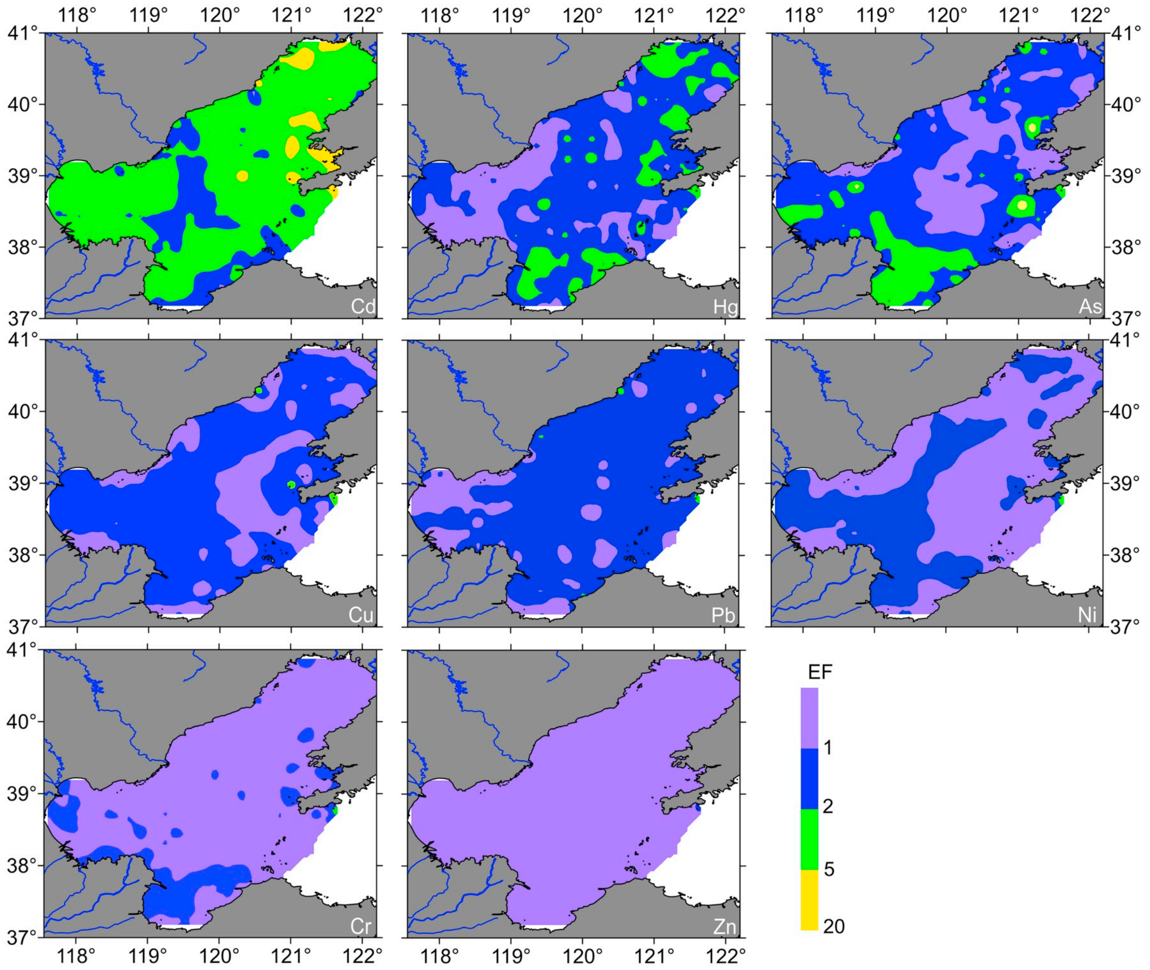

Fig. 5. Heavy metal enrichment factor spatial distributions in the surface sediments from the Bohai Sea.

which is consistent with a high potential for damage to the ecosystem. Furthermore, Cd and Hg geoaccumulation indexes and enrichment factors were high in Jinzhou Bay and the YRD, suggesting that human activity is responsible for the introduction of these heavy metals.

CRediT authorship contribution statement

Aimei Zhu:Data curation, Writing - original draft.**Jihua Liu**:Writing - review & editing.**Shuqing Qiao**:Investigation.**Hui Zhang**:Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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