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SEDIMENT AND OTHER POLLUTIONS (R DATTA AND P ZHANG, SECTION EDITORS)

Status and Trends of Sediment Metal Pollution in Bohai Sea, China

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Abstract This paper summarizes the advancement in the Bohai Sea sediment metal pollution studies in China. It includes spatial metal distributions, anthropogenic sources, and pollutant transport as well as factors affecting concentrations and potential ecological risk due to metal contamination. The results indicate that the pollution in the Bohai Sea is serious in coastal areas and, if no protection procedures are implemented, the situation can become worse with the economic development in the Bohai Sea rim. It is found that the metal distributions are quite different due to different pollutant sources in the coastal areas as well as along-shore current transport. The study shows that metal pollution is the most serious in the northern Liaodong Bay, followed by Bohai Bay and Laizhou Bay. The pollution in these three bays is much more serious than that in the central basin of the Bohai Sea. Hg, Cd, and Pb are the predominant pollutants commonly found in the Bohai Sea although the degree of the pollution varies with different regions of the Bohai Sea. Finally, the paper points out the current environmental concerns with the Bohai Sea sediment metal pollution.

Keywords China · Bohai Sea · Sediment · Heavy metal · Pollution

Introduction

The Bohai Sea in China is a semi-enclosed inland sea, including three bays (i.e., Liaodong Bay in the north, Bohai Bay in the west, and Laizhou Bay in the south) and a central basin and is connected with the Yellow Sea via the Bohai strait in the east (Fig. 1). It covers an area of 1.60×10^4 km^2 with a population of about 70 million living in its coastal area. The average water depth in the Bohai Sea is 12.5 m with a maximum depth of 32 m [1, 2]. Both water exchange capacity and self-purification ability are poor in the Bohai Sea. With the rapid social and economic development around the Bohai Sea rim, discharge of heavy metal pollutants into the Bohai Sea is increasing, which has rapidly worsened the Bohai Sea environmental quality [3]. Yellow River, Haihe River, Liaohe River, and Luanhe River also discharge into the Bohai Sea, carrying different materials from different land-based sources. It is reported that rivers and streams around the Bohai Sea are the main source input of heavy metals [4].

Before the early 1990s, there were very few sediment studies in the Bohai Sea. Investigations were limited to some estuaries and harbors [5, 6–8] until August–October 1998 when the National Oceanic Administration of China organized the second campaign of sediment pollution investigation in all China seas. The survey parameters included heavy metals (such as Hg, Cd, Pb, and Cs), total nitrogen and phosphorus, organic matters, sulfide, and organic pollutants (such as DDTs, PCBs, PAHs, and phthalate). This investigation provided a reference for study of sediment pollutant distributions, sources, transport, chemical speciation, and sedimentation in the Bohai Sea [9, 10]. The results of the survey showed that

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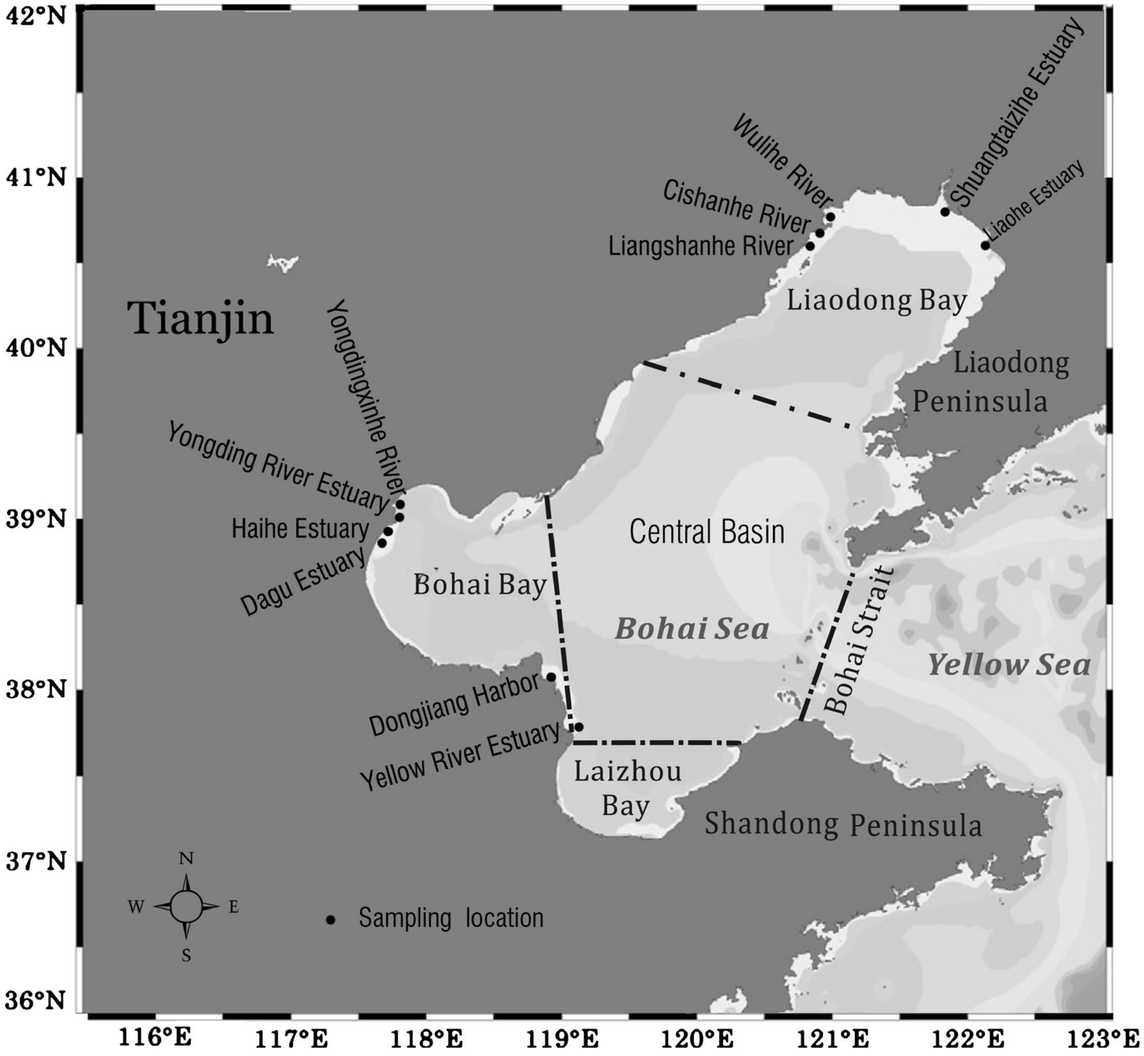

Fig. 1 Map of the Bohai Sea, China

the pollution in The Bohai Sea was very serious and became worse. In December 1998, the Bohai Blue Sea Plan [11] was officially launched with a focus on the comprehensive control of some key areas, estuaries, and pollutants. Since then, studies have been paying more attention to sediment pollution in the Bohai Sea.

Many studies have indicated that the interaction among heavy metals, biological macro-molecules group, and genetic materials may cause deformity, mutation, and cancer [12–14]. Most of the heavy metals in water body are scavenged by suspended particles falling down to the sediments. However, metals adsorbed on the sediments can be released into the overlying water due to contaminated sediment resuspension under certain environmental conditions, e.g., current and wave activities. Furthermore, toxic metals can threaten the ecological system directly and indirectly due to biological accumulation and magnification [15]. Therefore, heavy metal pollution research is very critical in order to protect the ecological system. In the past two decades, environmental scientists have conducted numerous heavy metal pollution studies in the Bohai Sea, including characterization of heavy metal sources, transport, and distributions, as well as the controlling factors by tracking the pollution history in this area [16, 17].

Characterization of Sediment Metal Concentrations and Spatial Distributions

The ^{210}Pb radioisotope dating technique is useful to study the history of metal (e.g., Cu, Pb, Zn, and Cd) concentrations and distributions in the Bohai Sea sediments [6, 18–20] because the concentrations and spatial distributions of heavy metals in the sediments are important indicators of the aquatic environment [21, 22]. The results of some heavy metal concentrations in the Bohai Sea sediments from the selected studies are summarized in Table 1. As shown in Table 1, the heavy metal

Table 1 The summary of heavy metal concentrations (mean±SD mg/kg) in surface sediments of the Bohai Sea and its adjacent bays and estuaries

Location	Sampling date	Sample size	As	Hg	Cd	Cr	Cu	Ni	Pb	Zn	References
Liaodong Bay	2007	56	9.11±3.15	28.6±8.7	0.10±0.10	54.5±22.0	15.8±6.4	21.6±9.4	20.4±5.1	57.8±18.8	Hu et al. [23]
Liaodong Bay	2009	128	8.3	0.04	N/A	46.4	19.4	22.5	31.8	71.7	Hu et al. [24, 25]
Liaohe River Estuary	1985	N/A	N/A	N/A	0.123	N/A	13.9	N/A	9.2	46.8	Zhou et al. [26]
Liaoning coastal region	2000.05	12	13	0.08	0.44	64	24	34	29	96	Li and Li [27]
River mouths Liaodong Bay	2000.03	22	N/A	N/A	1.16	N/A	17.6	N/A	23.9	105	Zhou et al. [26]
ShuangtaiziheEstuary	2008.07	2	N/A	N/A	0.21	50.5	19.9	25.1	20.8	61.4	Liu et al. [28]
Wulihe River	2005.05	10	N/A	8.668	7.947	N/A	56.6	N/A	80.5	525	Zhang et al. [29]
Cishanhe River	2005.05	6	N/A	33.07	250	N/A	217	N/A	454	5595	Zhang et al. [29]
Liangshanhe River	2005.05	4	N/A	1.59	9.73	N/A	73.1	N/A	105	451	Zhang et al. [29]
Jinzhou Bay	2006.09	14	397	N/A	248	60.6	417	N/A	753	6419	Zhang et al. [29]
Jinzhou Bay	2009.10	25	N/A	N/A	26.8±22.2	N/A	74.1±68.5	43.5±11.9	124±115	689±569	Li et al. [30]
Southern sea Huludao City	N/A	35	12	0.169	N/A	75	25.9	N/A	32.2	144	Li et al. [30]
Bohai Bay	N/A	18	N/A	N/A	0.15	79.7	28.7	36.56	25.6	72.8	Gao et al., [1]
North Bohai Bay	2008	33	8.50±1.90	0.028±0.028	0.15±0.14	47.0±22.0	13.0±10.0	N/A	25.0±9.2	60.0±40.0	Luo et al., [31]
North Bohai Bay	2008	35	N/A	N/A	0.13±0.18	N/A	N/A	N/A	N/A	50.0±39.0	Luo et al., [32]
NW Bohai Bay	2008	42	N/A	N/A	0.22	101	38.5	40.7	34.7	131	Gao and Chen [33]
Bohai intertidal zone	2008	15	N/A	N/A	0.12	68.6	24.0	28.0	25.6	73.0	Gao et al., [34]
Dongjiang Harbor	2009	11	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	Guo et al., [35]
South Bohai Bay	2008	119	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	Hu et al. [24, 25]
Haihe estuary	2008	15	N/A	N/A	0.19	63.3	61.9	38.3	22.1	112	Wu et al., [36]
Coast of Bohai Bay	2012	90	12.70	0.40±0.30	0.13±0.06	102±25	51.0±11.2	55.0±17.0	37.0±10.3	144±82	Wua et al., [37]
Bohai Bay	2011	15	N/A	0.12	0.25	N/A	33.0	N/A	29.0	118	Zhou et al., [38]
Bohai Sea	1983–1985	25	N/A	N/A	0.12±0.08	N/A	21.8±6.5	N/A	14.4±4.5	67.6±17.0	Li et al., [20]
Dagu & Beitang Estuary	N/A	25	N/A	N/A	0.13±0.03	N/A	28.8±5.0	N/A	20.2±4.0	75.3±18.7	Li and Hao [19]
Dagu Estuary	2003	21	7.24	26.34	1.48	46.99	14.7	19.98	16.9	275.5	Meng et al. [39]
Bohai Bay	2003	21.00	6.66	0.57	0.12	N/A	27.2	N/A	17.3	99	Qin et al. [40]
Yongding Estuary	2004	12	N/A	0.12	0.10	N/A	21.3	N/A	2.78	62.9	Zhang et al. [41, 42]
south of Bohai	2009	21	N/A	N/A	0.66	43.67	17.3	N/A	25.3	61.7	Zhang et al. [4, 43]
Bohai	2008	11	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	Zhang et al. [4, 43]
Dagu Estuary	2008	5	20.24±1.00	N/A	0.11±0.00	38.1±2.7	34.2±18.2	25.2±1.6	14.7±2.1	61.0±9.9	Zhang et al. [4, 43]
Bohai Bay	1997	N/A	N/A	0.05±0.03	0.24±0.17	N/A	28.1±8.1	N/A	21.2±18.3	103	Zhan et al. [44]
Bohai Bay	2003	N/A	N/A	N/A	N/A	25.4	25	26.9	22.1	88	Qin et al. [45]
Bohai Bay	2008	N/A	N/A	N/A	0.3	83.7	35.32	39.99	30.6	97.3	Qin et al. [45]

Table 1 (continued)

Location	Sampling date	Sample size	As	Hg	Cd	Cr	Cu	Ni	Pb	Zn	References
Tianjin coastal region	2000.05	10	16	0.15	0.19	82	32	47	29	112	Li and Li [27]
Hebei coastal region	2000.05	22	8	0.03	0.13	63	20	29	23	58	Li and Li [27]
NW coastBohai Bay	2003.06	20	8.22	0.33	0.63	94.3	19.4	N/A	22.7	132	Meng et al. [46]
Estuary intertidal Bohai Sea	2004.01	12	N/A	0.12	0.1	N/A	21.3	N/A	2.78	62.6	Zhang et al. [42]
Intertidal Bohai Bay	2008.05	15	N/A	N/A	0.12	68.6	24	28	25.6	73	Gao and Li [47]
Coastal Bohai Bay	2008.05	42	N/A	N/A	0.22	101.4	38.5	40.7	34.7	131	Gao and Chen [33]
Western Bohai Bay	2007.07	5	N/A	N/A	1.29±0.12	53.1±3.3	27.9±1.8	31.4±3.4	20.5±1	83.6±4.4	Feng et al. [18]
Port of Tianjin	2007.07	3	N/A	N/A	0.17±0.05	61.5±9.5	38.2±16.9	36.1±3.8	23±2	110±30	Feng et al. [18]
Port of Tianjin	2010.09	20	3.73	0.31	1.24	N/A	6.37	N/A	14.21	N/A	Zhang et al. [48]
Dagu Draige Cal	2010.09	16	4.87	0.87	11.3	N/A	69.9	N/A	55.9	N/A	Zhang et al. [48]
Yongdingxinhe Estuary	2007.07	2	N/A	N/A	0.124±0.004	51.9±3.5	31.4±1.6	31.2±3.1	20.4±1.3	92.9±2.3	Feng et al. [18]
Haihe River Estuary	2007.07	2	N/A	N/A	0.173±0.009	57.6±5.1	29.5±3.5	33.6±2.5	25.6±0.7	86±13.7	Feng et al. [18]
Southern Bohai Bay	2008.08	119	N/A	N/A	0.14	33.5	22.7	30.5	21.7	71.7	Hu et al. [24]
Dongjiang Harbor	2009.03	11	17.13	0.07	0.06	N/A	19.4	N/A	4.34	88.4	Guo et al. [35]
Coastal N Bohai Yellow Seas	2008.03	35	8.5±1.9	0.028±0.028	0.15±0.14	47±22	13±10	N/A	25±9.2	60±40	Hu et al. [25]
Coastal watersheds N Bohai Sea	2008.10	35	N/A	0.027±0.028	N/A	N/A	N/A	N/A	N/A	N/A	Luo et al. [49]
Upstream coastal N Bohai Sea	2008.10	17	N/A	N/A	0.13±0.18	N/A	N/A	N/A	N/A	50±39	Luo et al. [50]
Downstream coastale N Bohai Sea	2008.10	12	N/A	N/A	0.11±0.082	N/A	N/A	N/A	N/A	34±33	Luo et al. [50]
Laizhou Bay	2012.05	18	N/A	N/A	0.22	50.6	11.5	18.9	17.9	48.3	Gao et al. [1, 2]
Laizhou Bay	2012.09	18	N/A	N/A	0.30	53.5	11.9	20.6	18.6	58.7	Gao et al. [1, 2]
Laizhou Bay	2007	32	13.10±2.30	N/A	0.10±0.10	57.1±11.5	13.3±5.8	19.4±6.6	20.2±5.1	59.4±15.4	Hu et al. [51]
Yellow River Estuary	2004–2005	27	9	0.04	0.1	19.7	19	N/A	13	31	Wu et al. [52]
Laizhou Bay	2007.08	42	13.1	0.053	0.081	57.1	13.3	19.4	20.2	59.4	Hu et al. [53, 54]
Internal of Bohai Bay	1983–1985	10	N/A	N/A	0.12±0.04	N/A	25.4±2.3	N/A	16.3±1.4	74.0±5.7	Li et al. [19]
Estuary of Yellow River	N/A	25.00	N/A	N/A	0.15±0.03	N/A	20.1±7.4	N/A	11.9±4.3	58.4±18.1	Li et al. [19]
middle of Bohai	2006	386	N/A	N/A	N/A	59.7±16.0	22.5±8.93	30.6±13.0	24.3±5.0	64.3±23.1	Liu et al. [55]

concentrations are different from area to area because of different local pollutant sources along the coast. For example, the concentrations of Hg, Cd, and As in the coast of Tianjin and Liaoning were much higher than that in Yellow River Delta [27, 45, 56–58], which could be attributed to industrial waste and sewage discharging through Haihe River and Liaohe Reiver in Tianjing and Liaoning area. The concentrations of Hg, Cd, and Zn in some areas in the Bohai Sea were 12 to 150 times higher than the background concentrations. The pollution was mainly caused by rapid economic development in the areas around the Bohai Sea [18]. The studies on sediment metal pollution in the intertidal zone of the Bohai Sea showed that heavy metal (As, Cd Cr, Cu, Pb, and Zn) concentrations were the highest in Liaodong Bay, followed by Bohai Bay (ranked as the second highest) and Laizhou Bay (ranked as the third highest) [4, 43]. The correlation analysis suggested that Cr, Cu, Zn, Cd, and Pb may have similar sources, but the major sources of As may be different from other metals [4, 43]. Studies also showed that sediment heavy metal concentrations, especially Pb and Hg, were increasing in many estuaries around the Bohai Sea [59].

Besides the large scale studies conducted in the entire Bohai Sea, there were many intensive regional studies conducted in many localized areas in the Bohai Sea to obtain important local information. For example, spatial analysis of metal concentrations in the west Bohai Sea indicates that heavy metal concentrations decreased from near-shore to off-shore and from south to north [60]. The concentrations of Cr, Zn, Cu, Pb, and Cd were higher in estuaries and in the middle section of Tianjin coastal area [61]. Due to the different contaminant sources along the coastal area, the vertical profiles of heavy metal concentrations showed wavy distributions [62]. The metal enrichment in the sediments was obviously caused by human activities (Gao et al., 2012 [45, 58]). High levels of Pb and Zn appeared especially in the estuaries, indicating that river discharge was the main pollution source [46]. In Dagukou of Haihe River estuary, Cu, As, Pb, and Cd were major contaminants in the sediments. The results indicate that the sediments in the estuary or nearshore were easily affected by land-based pollutant input [4, 43]. Two minimum heavy metal concentrations were found in a core collected from the Haihe River estuary. According to age determination, these two minimum concentrations respond to the time periods of 1938–1942 and 1960–1967 when the flooding events occurred in the northern part of Haihe River water system in 1939 and 1963, respectively [63]. In the intertidal zone of Yongding River estuary, it was found that the sediment mental concentrations changed with tides—the concentrations of Cu, Pb, and Hg were higher in spring tidal period but Zn and Cd were higher in neap tidal period because of the different sources. However, the vertical profiles of the heavy metal concentrations were always higher in the upper section of sediments than that in the lower section no matter in spring or neap tidal period [42]. The results suggest high metal input in recent years possibly due to urbanization and economic development in the area in last two decades [18].

In the north of the Bohai Sea, the distributions of sediment heavy metals in Liaodong Bay were controlled by four different factors, i.e., the particle size, concentration of organic materials, distance from estuaries, and ocean currents [23]. The concentrations of Hg, Cd, and Pb were higher near Qinhuang Island and argillaceous zone in Liaodong Bay. The concentrations of As, Hg, Pb, and Cd as well as Hg were particularly high in the marginal area of the Luanhe River Delta. The concentrations of Hg, Pb, and Cd were also high near Liaodong Peninsula in Liaodong Bay [23]. It was found that the concentrations of Zn, Pb, Cd, and Hg increased abruptly after 1970s. Coincident to the increase of heavy metal contents, the decreasing trend of ^{206}Pb to ^{207}Pb ratio indicated that Pb in the surface sediments mainly came from the anthropogenic activities [64, 65]. As shown in Table 1, the China State Bureau of Quality and Technical Supervision (CSBTS) [66] enacted Marine Sediment Quality (GB 18668–2002) to prevent and control marine pollution, protect marine life, ensure marine resource sustainable uses, maintain marine ecological equilibrium, and protect human health. The first level of sediment standard criteria is the most conservative with respect to metal toxicity and is used to protect marine life. The second level standards are also developed to regulate general industrial uses and protect coastal tourism. The third level standards are developed to address the management of harbors and special uses for ocean exploration. According to China National Standard for Marine Sediment Quality, the pollution of Zn, As, Cd, and Pb in Jinzhou Bay was extremely serious and some areas in Jinzhou Bay had very high metal concentrations that can constituted a long-term ecological risk (Table 1, [29]).

In the southern portion of The Bohai Sea, the concentrations of Cu, Zn, Pb, Cd, As, Ni, and Cr in Laizhou Bay decreased from nearshore to bay and are higher in northern portion than those in the southern portion with high Ag in the central circulation area that was located in southwest central Laizhou Bay. The ecological risk caused by As and Ni was most serious in northwest section of Yellow River estuary, followed by the center and north of Laizhou Bay [51]. In costal Laizhou Bay, the results showed that the Cr, Cu, Ni, and Zn were the main environmental threat according to the sediment quality guidelines. The marine area was generally in good condition with low risk from heavy metals and adverse effects on biota could hardly occur. Natural sources dominated the concentrations and distribution of Cu, Ni, Pb, and Zn [57]. In the intertidal Bohai Bay, the studies indicated that sediment grain size played an important role in controlling metal concentrations [34].

Source and Transport of Heavy Metals

The vertical distributions of heavy metals in the sediments kept the historic record of heavy metal contamination in water and can provide the information of heavy metal source changes and evolution processes [67]. Radioisotope dating of sediments is the key step to study sources of pollutants. Environmental geochemists often make use of ^{210} Pb method (e.g., [68]). Qi et al. [69] analyze the radioactivity of ^{210}Pb in the Bohai Sea and the Yellow Sea for sedimentation study. Li et al. [20] estimated the sediment deposition rate in the Bohai Sea and found that it was rapid in estuary as well as nearby areas but slow in the central basin of the Bohai Sea. Meng et al. [70] measured ^{210}Pb and ^{137}Cs spatial and temporal distribution patterns to investigate the sediment transport in western Bohai Bay. These studies indicated that sandy areas of the intertidal zone experienced rapid sedimentation from the 1950s to 1960s due to the fact that Northern China had plentiful precipitation during that time period. The sedimentation rate decreased after the 1960s because of the reduction of sand transported to the intertidal zone as a consequence of diminished rainfall in Northern China and intensive human activities in the Haihe basin. The sedimentation rate generally increased from north to south. Feng et al. [71] study the temporal and spatial variations of ^{210}Pb and ^{7}Be in western Bohai Sea and found that it was a non-steady state depositional environment. By comparing ^{210}Pb and ^{7}Be inventories in sediments with those from atmosphere source input, they found that the sediments dredged from Tianjin Harbor or eroded from estuarine, and coastal areas were retained for a relatively short time (several months), as reflected in the relatively high ^{7}Be inventory despite local variability in sediment dynamic and disturbance due to human activities. However, ^{210}Pb inventories in sediments indicated that there was a net on-shore transport of sediments and, nevertheless, the sediments were mass-balanced over a long-term (years to decades). Hu and his coworker [53, 54] studied ^{210}Pb sedimentation rate and its relationship with sediments transport. The results showed that high sedimentation rates were in estuaries around the Bohai Sea with the highest rate in the Yellow River Delta, whereas the offshore area was characterized by low sedimentation rates. In the northern Shangdong Peninsula, iso-lines of sedimentation rates extended northeastward and decreased step wise with the lowest at eastern Chengshantou. Based on the distribution pattern of ^{210}Pb and the sedimentation rates in southern Bohai Sea, most of the sediments from the Yellow River were deposited within the coastal area. There were small portions of those sediments which were transported along northern Shangdong Peninsula and through the Bohai Straits into northern Yellow Sea.

Metal Pollution Assessment

Correlation analysis is a statistical approach to study the interrelationship between variables. Many researchers judge the sources of pollutants by use of heavy metal correlations with a special element named as the reference element [72, 73]. Many investigators often use Al as the reference element because it is one of the most abundant elements on the earth and cannot be easily contaminated [74–78]. There means no anthropogenic contamination if the metal has a correlation with Al closed to the earth abundance ratio. In fact, the heavy metals in sediments always add up together and the concentrations of heavy metals decrease when the particle size increases. The statistical approaches mentioned above cannot eliminate the effect of particle size and natural source variations in heavy metal concentrations at the same time [79, 80]. In order to distinguish the natural sources from the anthropogenic sources, investigators usually normalize the metals of interest against the reference element to reduce the interference of particle size on heavy metal concentrations [81–84]. Liu et al. [55] analyzed the sources of sediment heavy metals in the central basin of the Bohai Sea by the use of Al as the reference element. The results indicated that the heavy metals were mainly from natural sources, but were impacted by human activities significantly. Qin et al. [45, 58] studied the sediment heavy metal sources in Tianjin section of Bohai Bay and found that the concentrations of Pb, Zn, and Cd were much higher than the background concentrations after the normalization, indicating obvious contamination by anthropogenic sources. In order to reach more reliable conclusions, several studies used integrated approaches to evaluate metal contamination. Meng et al. [39] studied heavy metals in sediment the intertidal zone of Bohai Bay by integrating correlation analysis with principal component analysis. They normalized metal contaminants using Li and Sc as the reference elements in order to validate the conclusions from the analysis of metal vertical distribution profile. The results suggested that As, Cd, Cr, Hg, Pb, and Zn were mainly from anthropogenic sources, while Cu, Al, Fe, and Ni were from natural sources. The metal enrichment factor approach was originally recommended by Kemp who proposed to use Al as a reference element. In principle, the higher the metal enrichment factor is, the worse the metal pollution is. Hu et al. [23, 51] combined enrichment factor approaches with principal component analysis to study the sources of heavy metals in Liaodong Bay and Laizhou Bay sediments. The results suggested that in Liaodong Bay Fe, Ti, Cu, Zn, Cr, Ni, Co, Mn, and Sc were mainly originated from terrestrial sources (rock and soil), while Ag, As, Cd, Hg, and Pb were mainly from anthropogenic sources, and Ba and Sr from halobiosin sediments. In Laizhou Bay, Fe, Cu, Cd, Cr, Ni, Co, Mn, and V are not enriched elements; As is the slight element; and Pb and Ag are moderate enriched elements [51]. Therefore, Fe, Cu, Zn, Cd, Cr, Ni, Co, Mn, V, As, and Sc in Laizhou Bay were mainly from natural sources, while Ag and Pb were mainly from anthropogenic sources. The conclusion was supported by another study [84] that V, Cr, Co, Ni, and Cu were not enriched in Bohai Bay, but Pb and Cd were slightly enriched. In Liaodong Bay, Fe, Ti, Mn, Cu, Zn, Ni, Cr, Co, Sc, and V were nearly the same as the background concentrations, As is slightly enriched, and Pb, Cd, Ag, and Hg were moderately enriched [23].

Another approach to evaluate sediment heavy metals pollution is potential ecological risk index [85], which is frequently used to evaluate the potential risk of heavy metal pollution in the sediments. It depends on the concentration of free metal ions and reflects not only pollution extent but also the potential risk degree of heavy metals. Previous studies indicate that the Bohai Bay has been slightly polluted by heavy metals, and the degree of heavy metal pollution from serious to minor is in the order of Cd, Hg, Pb, Cu, and Zn [86]. In Liaodong Bay wetland, the potential ecological risk is slightly higher than that in Dalinghe River and Liaohe River estuaries and much higher than that in Shuangtaizi River and Xiaolinghe River estuaries. The ecological risk caused by metal pollutants from high to low is in the order of Cd, Pb, Cu, and Zn [26]. In Jinzhou Bay, Zn, As, Cd, and Pb pollutions are extremely serious, which make Jiaozhou Bay in a high ecological risk for a long time [29]. In southern Bohai Sea, the potential ecological risk caused by the heavy metals from high to low is in the order of Cd, Pb, Cu, Cr, and Zn [4, 43]. In Tianjin section of Bohai Bay, the sediments quality is markedly polluted and Hg is the primary element of pollution which is seriously harmful to ecology [62]. Wang et al. [87] used the biological effect database to establish a heavy metal concentration standard for Jiaozhou Bay environmental quality control. By comprehensive use of sediment metal enrichment factor and Hakanson potential ecological risk index, several studies indicate that Hg and Cd are major ecological risk factors in southern Bohai Bay, and heavy metal pollution is more serious in the northern region than that in the southern region because of terrigenous input and coastal zone development in the northern region [24, 25, 88].

Geo-accumulation index proposed by Professor Muller [89] is an important indicator to evaluate the environmental impact due to human activities because it takes the human activities into account. By the comprehensive use of the geo-accumulation index and potential ecological risk index, several studies found that Cd, Hg, and Pb were positively correlated in the Bohai Sea. Various levels of Hg pollution were found in different areas within the Bohai Sea, but Cd pollution was found the most serious in Liaodong Bay. Hg pollution was predominant near Qinhuang Island and, to a much less extent, in Laizhou Bay. Hg pollution was not found in Bohai Bay [77]. It was found that there was a slight metal pollution near Liaodong Peninsula, moderate metal pollution in Haihe River estuary, Tianjin coast and central Bohai Bay. The pollution was mainly caused by Pb, followed by Cr, Zn, and V [55]. In the intertidal zone of the Bohai Sea, it was reported that Cd had caused moderate pollution and Pb had a slight impact. The ecological risk caused by heavy metal pollution from high to low was in the order of Cd, Pb, As, Cu, Zn, and Cr in the intertidal zone [4, 43].

In ecological risk assessment, the approach using the potential ecological risk index is preferable for a large area because it eliminates the pollution influence by regional and sources differences [40]. In Laizhou Bay, the degree of metal pollution from high to low was reported in the order of Cd, Pb, Zn, Cu, As, and Hg. However, the potential ecological risk from high to low was reported in the order of Hg, Cd, As, Pb, Cu, and Zn , [1, 2, 34, 90]. The results show that there may be some deviations in the results obtained from the different approaches in sediment heavy metals pollution assessment. To access adverse biological effects on an ecological system, a variety of guidelines have been proposed to evaluate contaminant risks to organisms (e.g., [91–94]). Long et al. [93] established two frequently used metrics for evaluating potentially adverse effects of contaminants on ecosystems based on their "concentration effects" distributions. The lower 10th percentile of the effects data is the effects range-low (ERL) concentration, while the median or 50th percentile of the effects data is referred to as the effects range-median (ERM) [93]. ERL and ERM values are usually combined to delineate three effects ranges, i.e., (1) at concentrations below the ERL, a minimal-effects range is anticipated, (2) at concentrations equal to or exceeding the ERL value but less than ERM, adverse effects might be expected to occasionally occur, and (3) at concentrations greater than or equivalent to the ERM, it is anticipated that adverse effects are likely to occur more than 50 % of the time [91, 93, 94]. In this study, ERL and ERM criteria were used to compare observed metal concentrations to the likelihood of their causing adverse biological effects. The specific potential for toxicity was estimated by calculating the hazard quotients (HQs) of the selected chemical contaminants [95]:

$$HQ = \frac{SCC}{SQG} \qquad (1)$$

where SCC is the sediment chemical concentration in milligram per kilogram, and SQG is the sediment quality guideline in milligram per kilogram. SQG values were set at ERL levels:

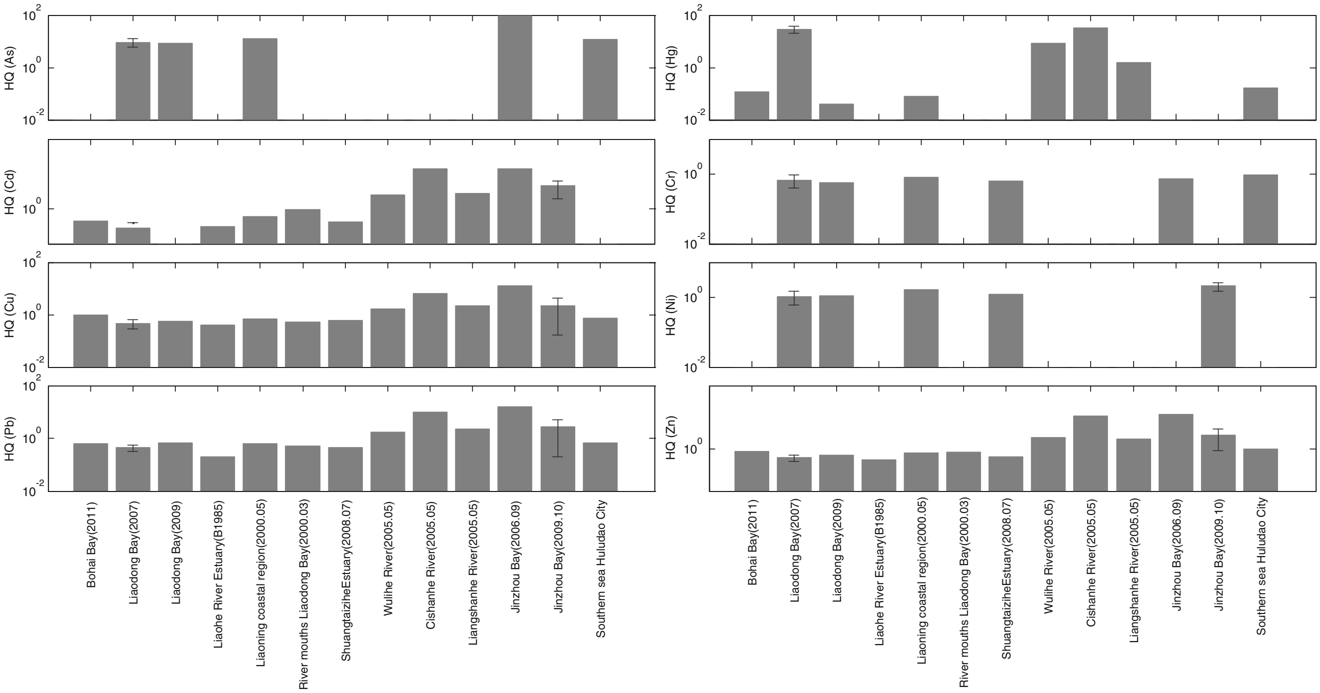

Fig. 2 Hazard Quotient of the Liaodong Bay, Bohai Sea, China

Ag=1.0, Cd=1·2, Cr=81, Cu=34, Ni=20·9, Pb=46·7, and Zn=150 [93]. For toxicity characterization, HQ values were used to express the potential risk to ecological receptors: at HQ<0.1 no adverse effects were expected; at 0.1<HQ<1 potential hazards were expected to be low, but in the range of 1.0<HQ<10, some adverse effects or moderate hazards are probable; and, finally, if HQ>10, high hazard potential is anticipated [96, 97]. Coastal ecosystem is sensitive and vulnerable adverse biological effects. Hazard quotients of selected metals in Liaodong Bay system, Bohai Bay system, and Laozhou Bay system are summarized in Figs. 2, 3, and 4. Figure 2 shows metal HQ in Liaodong Bay and associated rivers and estuaries. In Liaodong Bay system, adverse biological effects caused by As and Hg in some

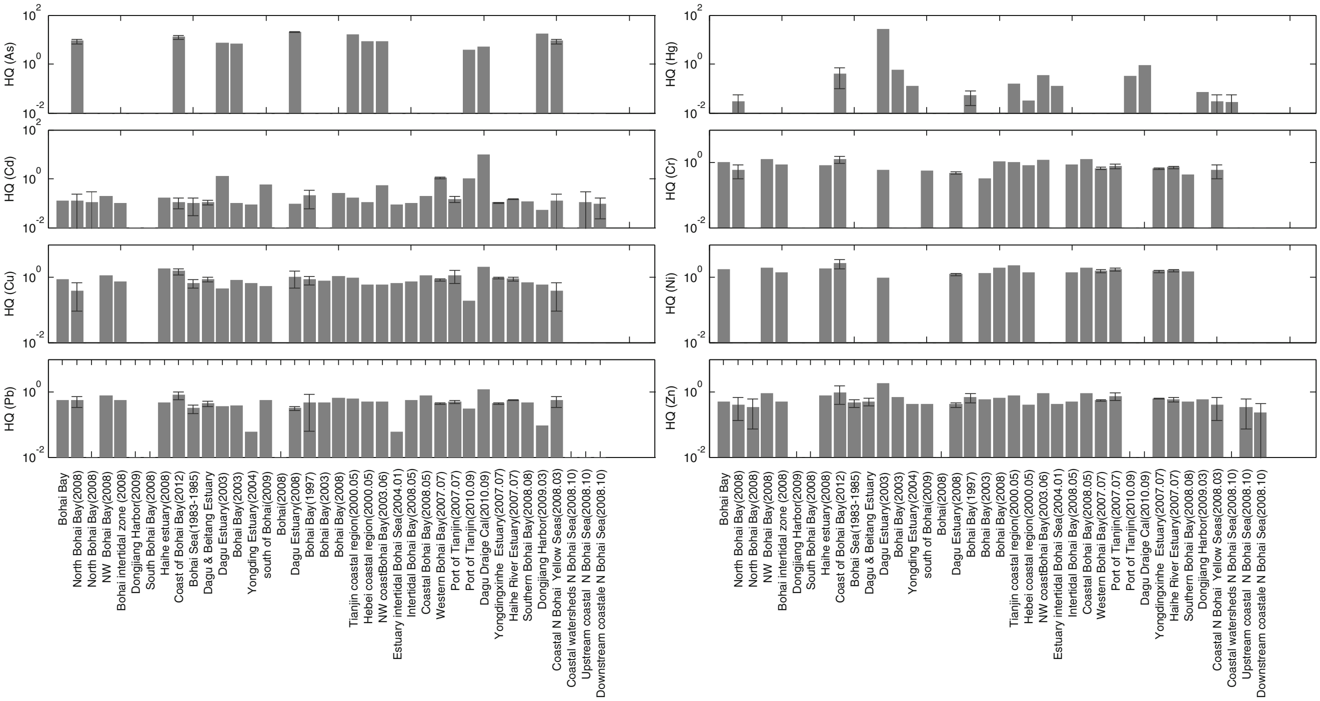

Fig. 3 Hazard Quotient of the Bohai Bay, Bohai Sea, China

Fig. 4 Hazard Quotient of the Laizhou Bay, Bohai Sea, China

areas are expected as HQ>10. The hazard potential caused by the other metals, such as Cd, Cr, Cu, Ni, Pb, and Zn, may be possible according to their HQ values (1<HQ<10) (Fig. 2). In Bohai Bay system, the situation is very similar to that in Liaodong Bay system. As and Hg have high hazard potential to cause adverse biological effect in some areas because of HQ>10 (Fig. 3). Low to moderate hazards may be caused by Cd, Cr, Cu, Ni, Pb, and Zn in some other areas (1<HQ<10) (Fig. 3). Laizhou Bay is in the southern portion of the Bohai Sea. There are limited data available for this system. As shown in Fig. 4, As is the only metal of concern, which has high hazard potential to cause adverse biological effects in Layzhou Bay and the Yellow River estuary. The adverse biological effects caused by Hg, Cd, Cr, Cu, Ni, Pb, and Zn in Laizhou Bay system are less concern according to their HQ values (HQ<1 or 1<HQ<10) (Fig. 4). According to current analysis, the northern portion of the Bohai Sea including Liaodong Bay has high hazard potential that can cause adverse biological effects due to more severe metal pollution in the region.

Current Concerns and Future Expectations in the Bohai Sea Environmental Research

Current analysis of metal source input to the sediments may distinguish the natural sources from the anthropogenic sources. However, this is still at the qualitative analysis state without knowing the proportions from each source. A quantitative analysis of metal input from natural and anthropogenic sources is expected in the future. The secondary contamination is a serious environmental problem, but there is only a few studies on the metal releasing mechanisms and people still do not fully understand the processes [98]. It is necessary to take into account of each individual factor influencing the heavy metal release such as Eh, pH, organic materials, seasonal variations, temperature, water flow velocity, and wind-generated waves. Currently, there is no globally unified method for the data comparison especially in speciation analysis. Because of this fact, there exists deviation in evaluation of sediment heavy metal pollution because the results are obtained by different analytical methods and based on different assessment criteria. The unification of analytical methods and assessment criteria is expected in the future research. The decontamination technology, especially the bioremediation technology, is still at a developing stage. It may become a "hot" topic in the field of environmental study to meet the needs of protecting environment and ecological system [99, 100].

Compliance with Ethical Standards

Conflict of Interest Yunfang Li, Lei Guo, and Huan Feng declare that they have no conflict of interest.

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