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REVIEW ARTICLE

Distributions and sources of heavy metals in sediments of the Bohai Sea, China: a review

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Abstract This paper summarizes the recent research results from studies concerning heavy metals in the Bohai Sea in recent decades. The temporal and spatial variations and potential sources of the heavy metals in the surface sediments were analyzed. Based on these obtained data, the average concentrations in surface sediments collected in recent years (summarized 3171 samples) were 0.31, 87.0, 25.7, 25.8, 0.11, 16.9, 52.2, and 27.7 μg/g for Cd, Zn, Cu, Pb, Hg, As, Cr, and Ni, respectively. In the samples collected in the 1980s, the concentrations were 22.6, 21.3, 69.13, 0.26, and 57.5 μg/g (summarized 218 samples) for Cu, Pb, Zn, Cd, and Cr. The concentrations of Cu, Pb, Zn, and Cd increased slightly. Generally, higher concentrations were measured in the Bohai Bay and central Bohai Sea. The distribution patterns of heavy metals were significantly different between samples collected after the year of 2000 and those in the 1980s. In the 1980s and recent years, higher concentrations of Zn, Cd, and Pb were measured in the samples collected from Bohai Bay and Liaodong Bay, respectively. This indicated that the sources of heavy metals in Bohai Sea were changed significantly during the past decades.

Keywords Heavy metal · Bohai Sea · Distribution · Potential source · Sedimentation environment

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Introduction

Metals are ubiquitous in the general environment and considered as nondegradable pollutants (He et al. 2015). Before the industrial revolution, heavy metals' cycle in the environment was controlled by the natural processes, such as natural weathering (Zheng et al. 2015), forest fire, and volcanic eruption. But in the latest century, heavy metals in the general environment were largely originated from anthropogenic sources, such as fossil fuel combustion, metal smelting, coal, and mineral mining (Zhang and Liu 2002; Li et al., 2014a). The concentrations of heavy metals in the general environment were increased sharply during the last century (Veerasingam et al. 2015).

A large part of heavy metals from the anthropogenic sources were long range transported by river and/or air, finally deposited to the coastal and shelf region (Cloquet et al. 2015; Catalan 2015). Heavy metals from natural sources were contained in the mineral crystal lattice and unavailable for organisms (de Vries et al. 2013). However, most of heavy metals from anthropogenic sources were bioavailable (Davis et al. 2003). Therefore, it caused significant negative effects on the marine ecosystem (Järup 2003). According to recent studies (Chen et al. 2016; NBSC, 2017), although water and sediment discharged into the Bohai Sea were strongly influenced by the dams and reservoirs in the river basin (Wang et al. 2007; Table S1), more than thousands of tons of heavy metals were discharged into the natural environment from anthropogenic sources in China (More information was given in Table S2). The influences of heavy metals from anthropogenic sources on the natural environment, especially the semi-closed bays, should not be underestimated.

Bohai Sea, the innermost gulf of the Yellow Sea, is now facing increasing metal pollution pressures because of the elevated metal discharges from various sources (such as sewage discharges and smelting). It is bounded by the Changshan Island chain between the Shandong and Liaodong Peninsulas. Laizhou Bay, Liaodong Bay, and Bohai Bay are three major bays inside the Bohai Sea (Fig. 1). Many studies have been focused on the heavy metal pollution in the Bohai Sea since 30 years ago (Li et al. 1994; Gao and Chen 2012; Gao et al., 2014). But previous studies only focused on the regional pollution levels, so there are still plenty of unanswered questions. So far, the distribution patterns of heavy metals in the entire Bohai Sea and the relationships between the distributions and sedimentation in environment/sources were unclear. Therefore, this review summarizes available data in the literature (the 1980s–2010s) on heavy metals in surface sediments of the Bohai Sea. The distribution patterns of heavy metals in the Bohai Sea in the 1980s and after the 2000s were systemically summarized based on published papers. The potential sources and the factors affecting heavy metal distributions were analyzed.

Distributions of heavy metals in surface sediments

Heavy metals in surface sediments of Bohai Sea have been extensively studied since the 1980s. This paper summarized the distributions of heavy metals in surface sediments of the Bohai Sea according to the published papers concerning heavy metal distributions in the 1980s and in recent 15 years.

In the 1980s

In the 1980s, the average concentrations of Cu, Pb, Zn, Cd, and Cr were 22.60, 21.33, 69.13, 0.26 and 57.50 μg/g. The detailed information is listed in Table 1. The concentrations were comparable with the values measured in the coastal sediments of China (Pan and Wang 2012). Generally, higher concentrations were measured in the samples from Bohai Bay, followed by in Liaodong Bay and Laizhou Bay (Fig. 2). In the Bohai Bay, the concentrations decreased eastward. Yellow River and Haihe River were considered as the most important sources of heavy metals in the Bohai Sea. Therefore, the distributions of heavy metals in the Bohai Bay were probably controlled by the distances to the sources.

In the central Bohai Sea, the concentrations of Pb were very high (Fig. 2). Before the 2000s, atmospheric Pb was largely originated from leaded gasoline combustion (Pacyna and Pacyna 2001), followed by other industrial activities, such as fossil fuel combustion and cement production. Before and after the leaded gasoline was phased out, the level and isotope composition of atmospheric Pb were changed significantly (Cheng and Hu 2010). Therefore, Pb in the general environment was significantly correlated with atmospheric deposition. The distribution of Pb in the central Bohai Sea was probably controlled by the sedimentary environment.

In recent 15 years

Figure 3 shows the distributions of heavy metals in surface sediments collected from the Bohai Sea in recent 15 years. The average concentrations were 0.31, 87.0, 25.7, 25.8, 0.11, 16.9, 52.2, and 27.7 μg/g for Cd, Zn, Cu, Pb, Hg, As, Cr, and Ni, respectively. Compared with the results observed during the 1980s (Fig. 2 and Table 1), the average concentrations were increased and the distribution patterns were changed significantly.

Increasing of heavy metal concentrations in recent decades was caused by the increasing emissions from industrial activities and land use/land cover changes. This point has been proved by many previous studies (Facchinelli et al. 2001; Xu et al. 2009). According to the results of Tian et al. (2012a), total atmospheric emissions of Cd, Cr, and Pb from coal combustion in China have promptly increased almost tenfold from 1980 to 2006. There were 36.7 t of Hg, 9.4 t of Pb, 4.4 t of Cr, 2.8 t of Ni, 0.9 t of Cd, and 0.2 t of As from municipal solid waste incineration in China during 2003 to 2010 (Tian et al. 2012b). Therefore, the increase was largely caused by the rapidly growing industrial and municipal emissions.

The most important finding was the changes of distribution patterns (Figs. 2 and 3), the relatively enriched areas of Pb, Zn, and Cd were shifted from the Bohai Bay to Liaodong Bay. This change was resulted from the joint effects of the sources and sedimentary environment changes. During the past decades, the sedimentary environment was changed significantly, such as the silt and sand transported by the rivers to the sea was reduced, due to both the coastal and upper stream human activities. In the Bohai Bay, the concentrations of As and Zn gradually decreased from 2007 to 2012, Hg and Cu concentrations increased, and Pb and Cd concentrations decreased from 2007 to 2009 or 2010 and increased after 2010 (Peng, 2015). This indicated that heavy metals discharged into the Bohai Bay continue to fall in recent years.

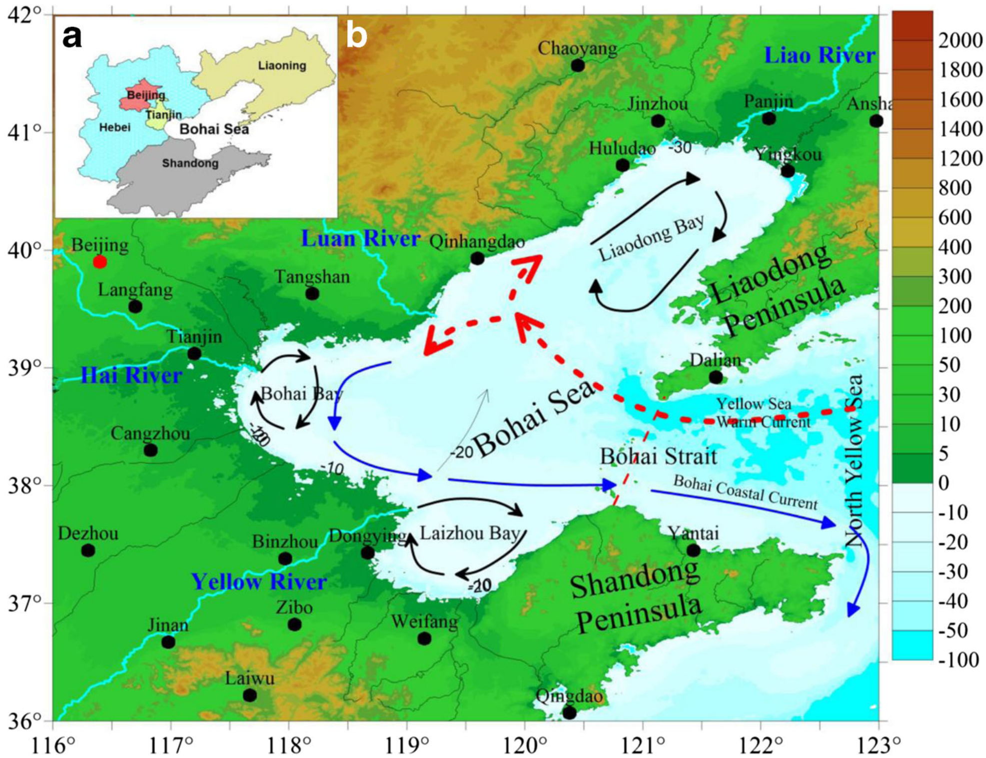

Fig. 1 Administrative map (a) and geological setting of the Bohai Sea, China (b). Cities are indicated by dots. Major currents are indicated by arrows. Gridded bathymetric data is from http://www.bodc.ac.uk/data/online_delivery/gebco/

Historical records of heavy metals in BS

As discussed above, distributions of heavy metals in the surface sediments of BS were changed significantly during the past decades (Figs. 2 and 3).

In Liaodong Bay, the concentrations of Zn, Pb, Cd, and Hg increased abruptly after the late 1970s (Xu et al. 2009). This indicated that high metal concentrations could result from increased inputs due to rapid industrial development and urbanization since the 1980s. Before the 1970s, this region mainly received inputs from the natural environment.

In Bohai Bay, dozens of cores were collected to analyze the historical records of heavy metals (Li et al. 1990; Qi et al. 2004; Qin et al. 2006; Zhao et al. 2009). In the coastal region of Bohai Bay (Qin et al. 2006), the concentrations of Pb, Zn, and Cd were increased sharply in recent decades and significantly higher than the background values, while the concentrations of Cu and Ni were lower than the background values and have remarkable correlations with the concentrations of Fe, Al, and Mn. This indicated that Pb, Zn, and Cd concentrations were influenced by human activities in surrounding areas. Cu and Ni originated from natural sources, such as

Table 1 Metal concentrations (mg/kg dry wt) in surface sediments of Bohai Sea collected in the 1980s. Values in the table were the reported average values

	Cu	Pb	Zn	Cd	Cr	Sampling size	Sampling date	Reference
Yellow River Estuary	20.40	/	80.40	/	/	35	1984	Xue et al. 1988
	28.30	/	66.30	/	/	44	1984	Ma et al.1989
	22.00	21.60	71.30	/	63.70	39	1983	Zhang et al., 1988
Tangshan coast	11.16	11.32	45.69	0.53	/	20	N.A.	Zhang et al., 1988
West Bohai Sea	28.30	37.02	81.70	/	51.30	11	1983	Zhang et al., 1988
Bohai Bay	26.00	22.40	73.60	0.15	/	41	1978–1980	Wu and Li 1985
Bohai Sea	22.03	14.30	64.90	0.11	/	28	1979–1985	Li et al., 1994
Mean value of BS	22.60	21.33	69.13	0.26	57.50	(218 samples were summarized)		

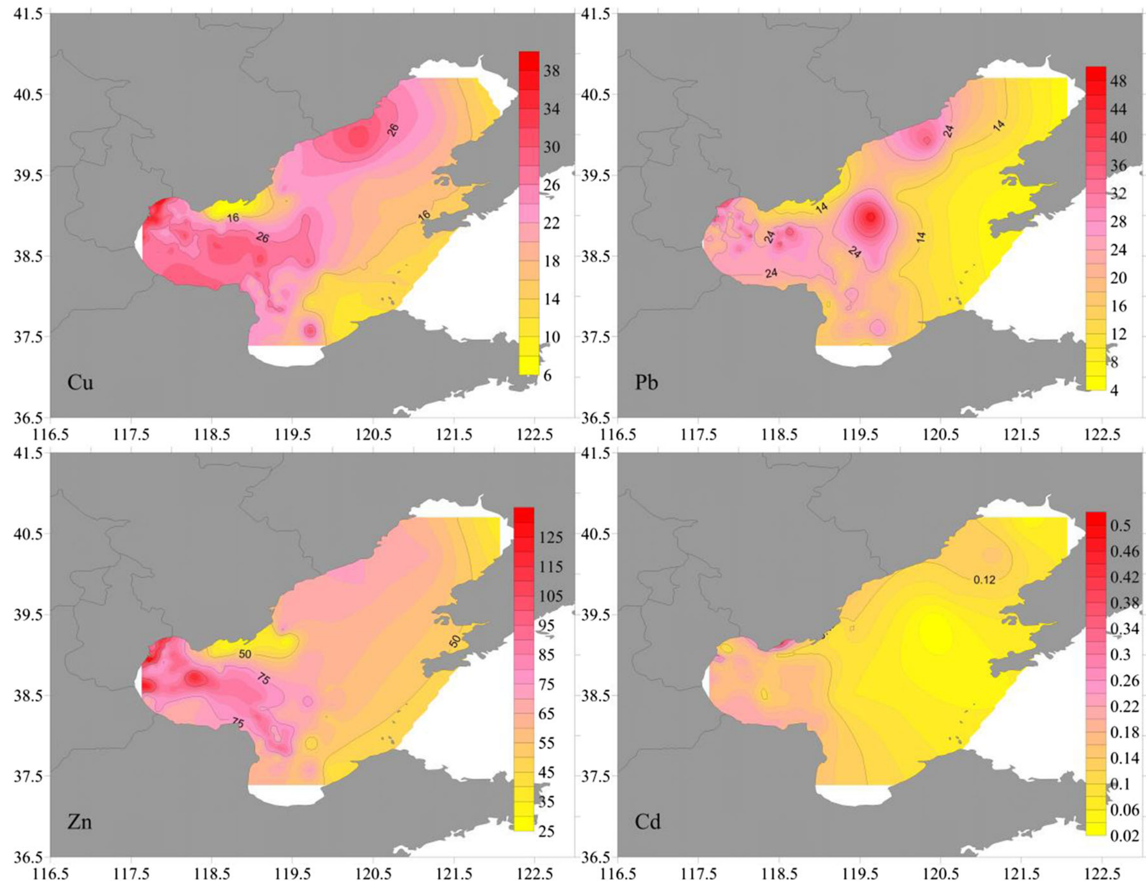

Fig. 2 Heavy metal distribution patterns in surface sediments of Bohai Sea in the 1980s (data are from Xue et al. 1988; Ma et al.1989; Zhang et al., 1988; Wang et al.1988; Zhang et al., 1988; Wu and Li 1985; Li et al.1994)

weathering and erosion. In the offshore and central region of the Bohai Bay (Li et al. 1990; Qi et al. 2004; Zhao et al. 2009), the concentrations of heavy metals have fluctuated greatly in the past decades and no significant increasing occurred. In some sites of the central Bohai bay (Zhao et al. 2009), the concentrations of As and Hg were increased slightly since the late 1970s.

In the central Bohai Sea (Liao et al. 2015), the concentrations of Ni, Pb, and Zn were decreased, while concentrations of Cu and Cr were almost stable with slight fluctuation. Nearly all sediments in this region originated from the Yellow River. Therefore, the variations of heavy metals concentrations have a significant relationship with the Yellow River discharge. In the Yellow River estuary, the concentrations of heavy metals did not change evidently during the past decades. Yellow River was the most important source of heavy metals in the estuary region; therefore, the temporal variations were totally controlled by the river discharge.

In general, the concentrations of some heavy metals in Liaodong Bay and the coastal region of Bohai Bay were increased. While in the Yellow River estuary and central Bohai Sea, the concentrations have evident interannual fluctuation, but not interdecadal variation. This was caused by the changes of Yellow River under the impact of human activities.

Potential sources

Bohai Sea is the largest semi-enclosed marginal sea of China. The elemental geochemical cycle in this region was significantly influenced by the river inputs, atmosphere deposition, and coastal human activities.

Riverine inputs

The riverine fluxes of heavy metals were strictly monitored by China's State Oceanic Administration since the early 2000s. Table 3 shows the fluxes of heavy metals and As in the rivers around the Bohai Sea. The annual differences were very significant, especially in some small rivers but there have no obvious regularity. It is indisputable that riverine input is one of the most important sources for heavy metals in the Bohai Sea.

Table 2 Metal concentrations (mg/kg dry wt) in surface sediments of Bohai Sea collected in recent 15 years. Values in the table were the reported average values

Sampling region	Cd	Zn	Cu	Pb	Hg	As	Cr	Ni	Sampling size	Sampling date	References
Laizhou Bay	0.19	50.63	10.99	13.37	0.04	7.10	32.69	17.38	15	2013	Zhang and Gao 2015
	0.18	122.70	65.90	24.50	/	9.63	54.30	23.40	40	2011	Xu et al. 2013
	0.12	60.40	22.00	21.90	/	12.70	36.80	/	150	2012	Xu et al., 2013
	0.45	190.77	32.63	23.89	/	/	/	28.59	7	2012	Wen and Gao 2014
	0.15	63.30	20.00	22.50	/	/	64.50	27.30	71	2012	Li et al., 2014b
	/	54.12	22.85	43.05	/	/	44.09	22.97	72	2009	Sun et al. 2015
	0.22	**90.32**	**29.06**	**24.87**	**0.04**	**9.81**	**46.48**	**23.93**		*(Mean value)*	
Bohai Bay	0.13	144.00	51.00	37.00	0.40	12.70	102.00	55.00	90	N.A.	Wu et al. 2014
	0.22	131.10	38.50	34.70	/	101.40	/	40.70	48	2008	Zhang and Gao 2015
	0.63	131.76	19.36	22.31	0.33	8.21	94.25	/	11	2003	Meng et al. 2008
	0.15	90.83	30.95	21.90	/	/	55.88	31.80	12	2007	Feng et al. 2011
	0.12	73.00	24.00	25.60	/	/	68.60	28.00	15	2008	Gao and Li 2012
	0.14	71.70	22.70	21.70	/	/	33.50	30.50	119	2008	Hu et al., 2013a
	0.25	118.00	33.00	29.00	0.12	/	/	/	15	2001–2011	Zhou et al. 2014
	0.66	61.65	17.27	25.32	/	/	43.67	/	21	2009	Zhang et al. 2011
	0.23	87.22	23.18	38.00	0.03	16.09	60.47	/	24	2013	Zhou et al. 2015
	0.20	62.85	21.26	2.78	0.12	/	/	/	12	2004	Zhang et al., 2010a
	0.36	158.70	69.03	41.27	0.27	25.23	0.10	/	24	2009	Zeng et al. 2013
	0.28	*102.80*	*31.84*	*27.23*	*0.21*	*32.73*	*57.31*	*37.20*		*(Mean value)*	
Liaodong Bay	/	71.70	19.40	31.80	0.04	8.30	46.40	22.50	128	2009	Hu et al., 2013b
	/	143.90	25.90	32.20	0.17	12.00	75.00	/	35	N.A.	Wang et al. 2013
	0.27	63.41	8.87	23.96	0.01	/	21.50	34.60	30	2013	Yang et al. 2015
	1.70	104.60	19.80	28.45	0.14	10.13	54.00	/	345	2006	Lin et al. 2013
	0.37	47.54	9.26	17.67	0.02	/	40.35	15.89	30	2013	/
	0.78	**86.23**	**16.65**	**26.82**	**0.08**	**10.14**	**47.45**	**24.33**		*(Mean value)*	
Centural Bohai Sea	/	64.27	22.50	24.26	/	/	59.66	/	386	2006	Liu et al. 2012
	/	55.59	19.14	22.50	/	/	55.01	22.98	212	*N.A.*	Li et al. 2010
	0.15	65.50	20.10	23.00	0.02	9.87	58.90	26.70	40	2009	Lan et al. 2014
	0.15	**61.79**	**20.58**	**23.25**	**0.02**	**9.87**	**57.86**	**24.84**		*(Mean value)*	
Yellow River Estuary	0.12	63.90	19.97	19.85	0.03	11.36	65.03	/	429	2006	Lin et al. 2013
West coast	0.09	45.25	14.83	18.93	0.02	6.87	45.02	/	757	2006	Lin et al. 2013
Luan River Estuary	0.09	44.60	18.76	31.00	0.19	7.21	41.13	15.60	33	2013	Duan et al. 2016
Mean value of BS	**0.31**	**87.00**	**25.66**	**25.77**	**0.11**	**16.91**	**52.21**	**27.71**	*(3171 samples were summarized)*		

Bold value: mean values

Italicized values: the highest mean values

Atmospheric deposition

Heavy metals are ubiquitous in the general environment, such as fossil fuels and metal ores. During the high temperature-related human activities, such as combustion of fuels and incineration of wastes, some trace metals evaporate entirely or partially from raw materials, entering the ambient air with exhaust gases (Pacyna and Pacyna 2001). After being emitted to the atmosphere, trace metals are subject to transport within air masses and migration through the ecosystem, part of the heavy metals are long range transported and finally deposited in the deep sea or even polar regions (McConnell and Edwards 2008). According to the record from a Greenland ice core (McConnell and Edwards 2008), the amount of Pb deposited from atmosphere in recent decades was much higher than in the early twentieth century. It was believed that coal burning in North America and Europe was likely the source of Pb in the Arctic after 1860.

Table 3 Fluxes of heavy metals (Zn, Cu, Pb, Cd, and Hg) and As discharged into the Bohai Sea by coastal rivers (tons/year; SOAC, 2002–2015)

Year	Heavy metals (Zn, Cu, Pb, Cd, and Hg)						As					
	DL.R	STZ.R	BL.R	L.R	Y.R	XQ.R	DL.R	STZ.R	BL.R	L.R	Y.R	XQ.R
2015	13	18	3		316	46	6	2	1		28	2
2014	13	21	1		386	26	3	4	0.02		58	2
2013	221	39	1		704	26	15	3	0.1		40	3
2012	31	16	1.3		1110	29	13	2.5	0.16		56	2.1
2011		23	4		640	384		3	0.2		47	4
2010		72	0.1		692	**655**		12	0.1		30	5
2009				23	579	61				2	**152**	7
2008			2		773	33			4		23	3
2007					1040						28	
2006		121	43	25	**1199**			1	5	2	52	
2005		138	4	19	810			16	**21**	1	35	
2004		**318**		10	932			8		1	44	
2003				**120**	200					10	60	
2002		15						0.2				

DL.R Da-liao River, *STZ. R* Shuang-tai-zi River, *BL.R* Bi-liu River, *YR* Yellow River, *XQ.R* Xiao-qing River

The highest fluxes of heavy metals in the rivers

China is a large energy-consuming country. Industrial emission was the most important source of heavy metal in the general environment (Cheng 2003). As shown in Fig. S1, the coal and oil consumptions have increased by two to three times, pig iron and cement productions have more than doubled during the past 15 years. Especially in north China, large amount of coal were consumed for home-heating in winter, which caused heavy atmospheric pollution. Therefore, atmospheric particulates enriched with pollutants were always spread around us in high concentration (Huang et al. 2014). Under the effect of monsoon, large amount of atmospheric particulates were transported from north China to southeast region, which caused significant influence on the environment of mass pass over region (Li and Duan 2015). In the Yellow Sea, the atmospheric flux of heavy metals may even be more important than river inputs (Zhang et al. 1992).

Table S3 shows the atmospheric heavy metal concentrations in cities around Bohai Sea. Almost all the heavy metal concentrations were higher than the limits in China or WHO. Before 1980 A.D., the atmospheric heavy metal

Table 4 Background values of heavy metals in the Bohai Sea surrounding areas and the Yellow River basin (The relative higher values in each column are indicated in bold red font)

Regions		As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Reference
Southern BS	Shan-dong	7.7	0.118	59.8	20.7	0.029	25.3	24.0	59.6	Pang et al. 2014
		9.3	0.039	66.0	24.0	0.019	25.8	25.8	63.5	CEPA, 1990
Western BS	He-bei	13.6	0.079	68.3	21.8	0.036	30.8	21.5	78.4	CEPA, 1990
	Tian-jin	9.6	0.090	84.2	28.8	0.084	33.3	21.0	79.3	CEPA, 1990
	Bei-jing	9.7	0.074	68.1	23.6	0.069	29.0	25.1	102.6	CEPA, 1990
		7.09	0.119	29.8	18.7		26.8	24.6	57.5	Chen et al. 2004
Northern BS	Liao-ning	8.8	0.066	57.9	19.8	0.037	25.6	21.4	63.5	CEPA, 1990
	Ji-lin	9.4	0.08	46.7	18.7	0.025	22.1	27.3	56.7	CEPA, 1990
	Da-lian	9.5	0.049	46.8	22.7	0.082	27.4	17.6	56.0	CEPA, 1990
Yellow River*		10.9	0.092	57.3	21.8	0.031	29.8	20.8	63.4	CEPA, 1990
BS			0.136 ~ 0.04		26.26			17.31	75.0 ~ 55.3	Li et al. 1994
China		11.2	0.097	61	22.6	0.065	26.9	26	74.2	CEPA, 1990

*the background values of the Yellow River were the mean values of Nei-meng-gu, Qing-hai, Ning-xia, Gan-su, Shanaxi, Si-chuan, Shan-xi, He-nan, and Shan-dong Provinces' background concentrations

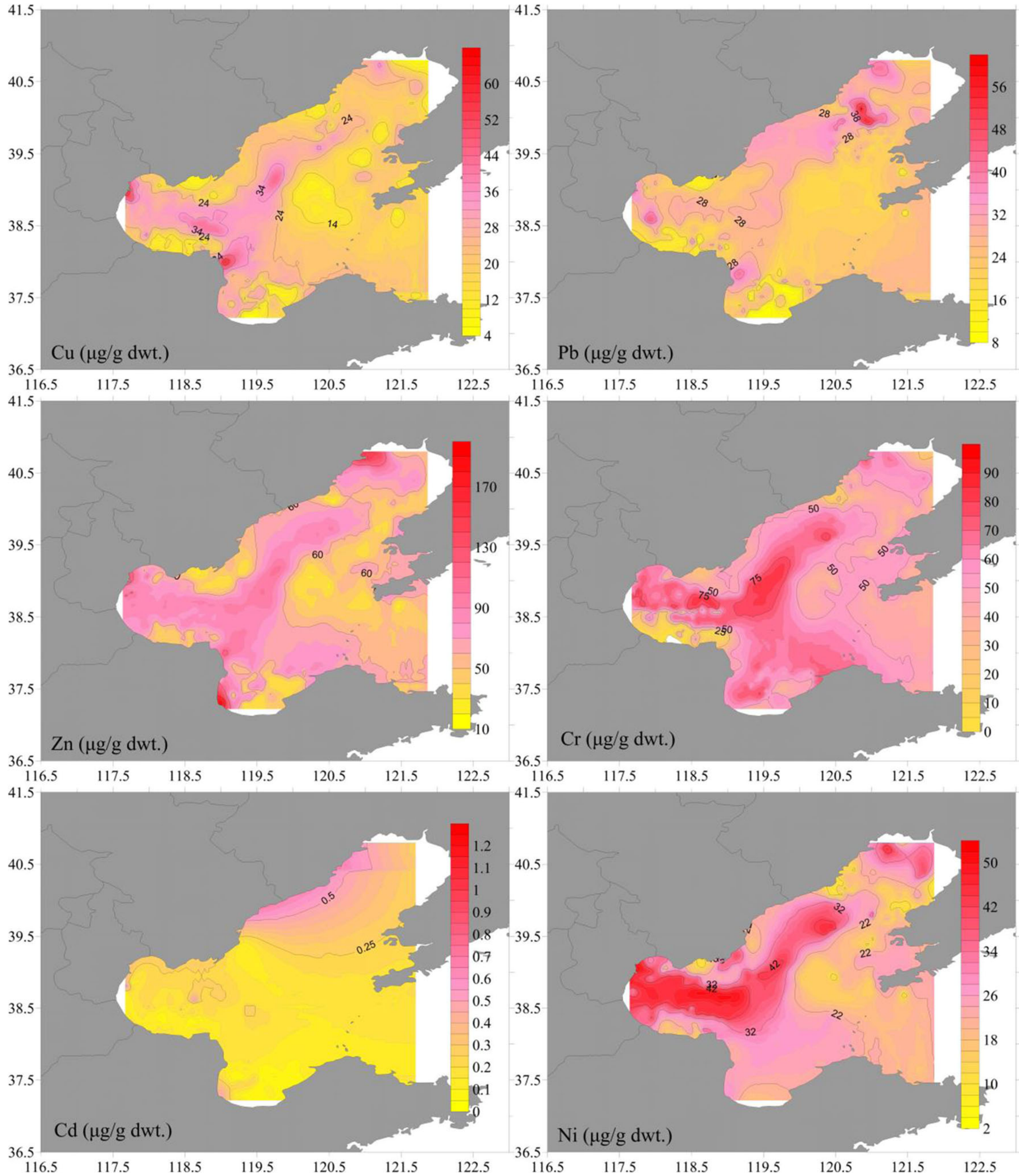

Fig. 3 Heavy metal distribution patterns in surface sediments of Bohai Sea in recent years (detailed data information is listed in Table 2)

concentrations were relatively low and they were generally natural origins. Since the 1980s, they began to increase gradually and increased the most in the 1990s (Wan et al. 2016). Since the 2000s, the emissions of atmospheric heavy metals increased at a much slower rate than coal consumption (Duan and Tan 2013). But the concentrations are still surprisingly high, especially during the haze day. According to the results of Tian et al. (2012a), total atmospheric emissions of Cd, Cr, and Pb from coal combustion in China have promptly increased from 31, 1019, and 2671 tons in 1980 to 261, 8593, and 12,561 tons in 2008, respectively. In the agricultural soils in China (Luo et al. 2009), atmospheric deposition accounted for approximately 43–85% of the total As, Cr, Hg, Ni, and Pb inputs.

According to Wang et al. (2011), the fluxes of atmospheric heavy metal to the coastal region of Bohai Sea were 12.5 mg/(m^2 a) for Pb, 32.6 mg/(m^2 a) for Zn, 0.3 mg/(m^2 a) for Cu, and 0.6 mg/(m^2 a) for Cd. Therefore, it can be estimated that 96 t of Pb, 252 t of Zn, 2.3 t of Cu, and 4.6 t of Cd deposited into the Bohai Sea every year. This amount was comparable with or even higher than the river inputs.

Coastal human activities

Discharges from coastal and offshore human activities, such as mariculture and maritime traffic, were also important sources of heavy metals in the continental shelf sediments (Naser 2013).There is more than 6000 km^2 mud flat in the Bohai Sea. The mariculture in the mud flat and nearshore area is one of the significant pillar industries for local inhabitants. In Laizhou Bay and Zhangzidao, two important areas of mariculture in the Bohai Sea, the predominance of Cu and Zn over all the other metals was evident (Gao et al. 2013). In Hailing Bay (Zhang et al. 2012), the aquafarming zone was more significantly polluted than the non-aquafarming zone by As, Ni, Cu, and Zn. In the agricultural soils in China (Luo et al. 2009), livestock manures were responsible for 51–69% of the total Cd, Cu, and Zn inputs, respectively.

Emissions from coastal factories, ports, and other facilities were the most important point sources of heavy metals. Such as Jinzhou Bay, where multiple industries including the largest zinc plants in Asia are located, has been seriously contaminated by the sewage effluent of factories in the surrounding area (Pan and Wang 2012). As shown in Fig. 2 and Fig.3, the concentrations of Zn and Cd in this region (west coast of Liaodong Bay) increased abruptly in the recent decades.

There are a few important oil reserves in the Bohai Sea and the vicinity of the gulf. Many oil spill accidents, especially ship accidents, have occurred in recent years (Liu et al. 2015). These accidents caused significantly negative effects on the environment. In Ebocha-8 oil-spill-polluted site in Niger Delta, Nigeria (Osuji and Onojake 2004), Ni, Cu, and Pb were more enhanced in the soils. However, there is no evidence of significant increases in metal levels due to the 1991 Gulf War oil spill (Kureishy 1993) and Dalian Port oil spill in 2011 (Zhao et al. 2012).

In recent years, the differences between regions in economic structure and development level were very significant. This leads to markedly different influences of human activities on local environment. The annual discharge amounts of heavy metals in waste water and dust in south and west coast provinces were much higher than that in north coast province (Table S4). That is, more amounts of heavy metals were discharged into the environment as the by-products of human activities in the south and west coast. That was the main reason for higher concentrations of heavy metals in the Bohai Bay and Laizhou Bay.

Higher level of economic development in the west and south coast caused higher emissions of heavy metals. Almost all the available evidences support this point. However, it cannot deny the fact that the concentrations of some heavy metals, such as Zn, Pb, and Cd, in surface sediments of the Liaodong Bay were increased (Figs. 2 and 3). That was almost certainly caused by the mariculture and shipping. The mariculture area of Liaoning almost has increased fivefold during the past 20 years (Fig. S2a). The mariculture area in the Liaodong Bay was much higher than in the Laizhou Bay and Bohai Bay. The waterway freight volume of Liaoning was also increased sharply (Fig. S2b). Not counting the Yantai and Qingdao (not in the Bohai Sea) port's cargo throughput, the cargo throughput of ports in Liaoning was more than in all of Hebei, Tianjin, and Shandong. Therefore, the increasing of heavy metal concentrations in the surface sediments of the Liaodong Bay was caused by the increasing oceanic development and utilization activities, such as mariculture and shipping.

Relationships between spatial distributions and depositional environment

As reported by Gao and Chen (2012), nonpoint source inputs from both fluvial transport and atmospheric deposition were important ways for the sediment contamination in Bohai Bay. It was also the same for Laizhou Bay and Liaodong Bay (Xu et al. 2015).]The spatial distribution patterns of heavy metals were largely controlled by the depositional environment.

Background values

In the past few decades, the Chinese government provided a large injection of cash into surveying the background values of elements in soils. As shown in Table 4, highest background values were detected in the western Bohai Sea, followed by in southern. Therefore, the higher concentrations of heavy metals in the surface sediments of the west Bohai Sea (Figs 2 and 3) were inseparable with influences of higher background values in this area.

Materials sources

Yellow River is the largest river around the Bohai Sea with 1.34×10^{11} m^3 water and 3.4×10^8 t sediment discharge in 2015, followed by Liao River (detailed information are shown in Fig. S3). Before the 1980s, the Yellow River and Hai River were the main sources of land-based materials in the Bohai Sea. Therefore, higher concentrations of heavy metals were observed in the Bohai Bay. The distributions of heavy metals in surface sediments of the Bohai Sea in the 1980s were probably controlled by the riverine inputs.

However, the water and sediment discharges from the rivers flowing into the Bohai Bay, such as the Hai River and Luan River, have dramatically decreased since the 1980s due to the water diversion in the upper stream and almost reached the point of drying out in the estuaries. It was a similar story for the Liao River. In the recent decades, water consumption in the North China increased significantly which causes groundwater levels to decline and rivers to dry up. Although the Yellow River was also changed markedly by human, it is still the largest source of sediment in the Bohai Sea. Therefore, the influences of coastal human activities probably play more important roles on the levels and distributions of heavy metals in the Bohai Sea in recent years than the rivers except the Yellow River.

Sediment characteristics and sedimentary dynamics

According to the published papers (Hu et al. 2009; Zhang et al. 2010b; Li et al., 2014c; Yuan 2015), the surface sediment characteristics of the Bohai Sea was systematically concluded (Fig. 4). The mud area distribution was significantly controlled by the riverine inputs. The Yellow River sediments were transported northward to the central Bohai Sea and eastward to the Bohai Strait. Sediments from other rivers were limited compared with sediments from the Yellow River. Therefore, the fine sediments from other rivers were deposited in the estuarine regions. After comparison of the distributions of heavy metals (Figs. 2 and 3) to surface sediment characteristics (Fig. 4), it was easy to find that there was a strong correlation between heavy metal concentrations and surface sediment grain size. Generally, higher concentrations were accompanied by fine-grained sediments.

Restricted by geographical factors and ocean currents (Fig. 1), the sedimentary environment interactions among the three bays were not obvious. We have reasons to believe that the distributions of heavy metals in the Laizhou Bay, central Bohai Sea, and Bohai Strait were largely influenced by the Yellow River. The distributions in the Bohai Bay and Liaodong Bay were influenced by adjacent riverine inputs and emissions from local human activities. This helps explain why the heavy metal distribution exhibits such patterns. Higher background values and riverine inputs caused higher heavy metal concentrations in the Bohai Bay and central Bohai Sea.

Conclusions

This review summarizes available data in the literature (the 1980s–2010s) on heavy metals in surface sediments of Bohai Sea. The results indicated that:

(1) The concentrations of heavy metals in the surface sediments of the Bohai Sea were increased during the past decades.
(2) Higher concentrations were observed in the Bohai Bay. Higher concentrations were caused by the higher riverine inputs, higher anthropogenic emissions, and higher background values.
(3) The Pb, Zn, and Cd heavily polluted areas were shifted from the Bohai Bay to Liaodong Bay. The concentrations of Pb, Zn, and Cd in the Liaodong Bay were increased sharply due to the dramatic increased emissions from mariculture and shipping since the beginning of the new century.
(4) The distributions of heavy metals were highly correlated with the surface sediment characteristics. Heavy metals in the Bohai Sea were originated from riverine input, atmospheric deposition, and emissions from coastal human activities. The influence of coastal human activities

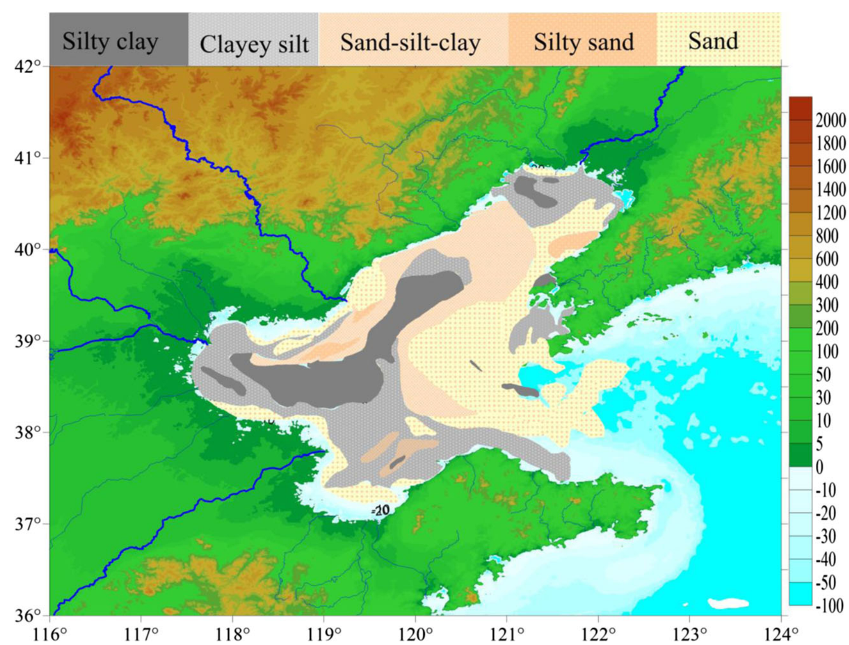

Fig. 4 The surface sediment characteristics of the Bohai Sea (Hu et al. 2009; Zhang et al., 2010a; Li et al., 2014a; Yuan, 2015). The sediment types were classified by Shepard classification method

on the concentrations and distributions of heavy metals in the Bohai Sea was increasing. However, the influence of riverine inputs was decreasing.

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