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REVIEW ARTICLE

Single and combined metal contamination in coastal environments in China: current status and potential ecological risk evaluation

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Abstract With the development of industrialization and urbanization, metal and metalloid pollution is one of the most serious environmental problems in China. Current contamination status of metals and metalloid and their potential ecological risks along China's coasts were reviewed in the present paper by a comprehensive study on metal contents in marine waters and sediments in the past few decades. The priority metals/metalloid cadmium (Cd), mercury (Hg), chromium (Cr), lead (Pb), and arsenic (As), which were the target elements of the designated project "Comprehensive Prevention and Control of Heavy Metal Pollution" issued by the Chinese government in 2011, were selected considering their high toxicity, persistence, and prevalent existence in coastal environment. Commonly used environmental quality evaluation methods for single and combined metals were compared, and we accordingly suggest the comprehensive approach of joint utilization of the Enrichment Factor and Effect Range Median combined with Pollution Load Index and Mean Effect Range Median Quotient (EEPME); this battery of guidelines may provide consistent, internationally comparable, and accurate understanding of the environment pollution status of combined metals/metalloid and their potential ecological risk.

Keywords Heavy metal · Combined contamination · Environmental quality evaluation · Coastal environment · China

Introduction

In China, the rapid development occurs on a huge scale in every part of the country, especially in coastal regions such as the Pearl River Delta and the Yangtze River Delta along the east coast (Fig. 1). Gross domestic product (GDP) from coastal regions accounts for ~ 60% of the total (He et al. 2014). However, coastal areas have paid a high environmental price for the fast economic development and are most often prone to the ultimate anthropogenic pollutants (Gao et al. 2013). A wide range of organic and inorganic compounds including metals and metalloid cause environmental deterioration and ecosystem degradation in coastal areas. Their resources include controlled and uncontrolled disposal of waste, smelting of metallic ores and mining, process and accidental spillage, and sewage sludge application to agricultural soils (Huang et al. 2013). In 2015, the estimated amount of industrial sewage discharged into the coastal environment in China was 21 billion tons (http://www.coi.gov.cn/gongbao/huanjing/). In most of the cases, serious metal contamination can be caused due to these direct discharges of industrial sewage water into coastal areas.

The increased discharge of metals and metalloid to estuarine and coastal environments caused by economic development and rapid industrialization in coastal regions has elevated the sediment metal concentrations in coastal regions in China. Sediments in aquatic environments usually act as a

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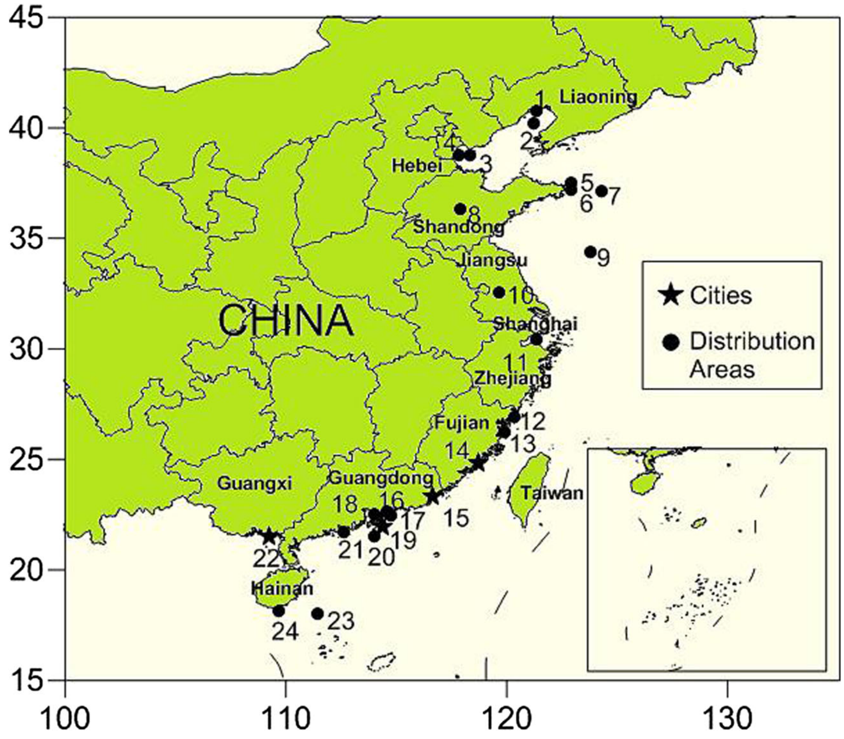

Fig. 1 Sampling sites of the cited studies in main coastal regions in China: 1. Jinzhou Bay. 2. Liaodong Bay. 3. Bohai Bay. 4. Haihe Estuary. 5. Swan Lake lagoon. 6. Rongcheng Bay. 7. Yellow Sea. 8. Yellow River. 9. East China Sea. 10. Yangtze River Estuary. 11. Hangzhou Bay.12. Luoyuan Bay. 13. Quanzhou Bay. 14. Xiamen. 15. Shantou. 16. Daya Bay. 17. Dapeng Bay.18. Shenzhen Bay. 19. Hong Kong. 20. Pearl River Estuary. 21. Hailing Bay. 22. Beihai. 23. South China Sea. 24. Sanya Bay

sink for contaminants, as they receive contaminants through the sedimentation of suspended matters that adsorb a significant amount of elements. Metal accumulation in upper sediments has potential toxicity to sediment-dwelling organisms. Hence, the monitoring and evaluation of metal distribution in sediment is important in the marine environment (Huang et al. 2013). The pool of bioavailable metals in sediments is typically much smaller than the total metal concentration and is strongly influenced by metal-binding with acid-volatile sulfide (AVS), particulate organic carbon (OC), and iron and manganese oxide solid phases (Campana et al. 2013; Huang et al. 2013). In Fig. 1 bellow, common coastal bays of China are mentioned that were mainly selected in different studies for determining the contamination caused by priority metals.

Cadmium (Cd), lead (Pb), mercury (Hg), arsenic (As), and chromium (Cr) are commonly considered as the priority metals in China in view of their current contamination status, high toxicity, and prevalent existence in the environment. Metals in sediments are associated with different geochemical fractions, which comprise the exchangeable, carbonatic, reducible (hydrous Fe/Mn oxides), oxidizable (e.g., sulfides) and residual fractions (i.e., mineral bound) (Tessier et al. 1979). This fractionation of the element in sediments influences the mobility, thus the bioavailability and toxicity of the element to organisms. The non-residual fractions are the most mobile, thus the most bioavailable/toxic for aquatic organisms. Generally, elements of anthropogenic sources mainly distributed in more biolavailable fractions (Huang et al. 2013); this claims the necessity of a bioavailability/toxicity-related evaluation approach besides the mere chemical assessment of total concentration of the element in environments.

Various evaluation techniques have been applied for examining the environmental contamination and potential ecological risk, including the Enrichment Factor (EF), Contamination Factor (CF), Effects Range-Median (ERM) guidelines, Mean Effect Range Median Quotient (mERM-Q), Mean Probable Effect Level Quotient (mPEL-Q), and Pollution Load Index (PLI). Studies have shown that different or even contradictory conclusions may be drawn by using different assessment methods for the same sample or for different elements within the same sample (Huang et al. 2013; Yu et al. 2011). Therefore, in the present study, the metal spatial distribution along coastal regions in China were reviewed with special focuses on the combined contamination caused by priority elements Cd, Pb, Hg, As, and Cr. Moreover, the differences and relationships of the most commonly used evaluation methods were examined, in order to provide useful information on exploring one set of internationally comparable, accurate, and comprehensive approaches for evaluating the contamination status and the potential ecological risk posed by combined elements.

Metal concentrations in coastal environments

Though it is well known for decades that metal/metalloid contamination is one of the most severe environmental problems, the amount of metals and metalloid produced and consumed in China is still significantly increasing with the development of industrialization and urbanization, especially in coastal regions (Huang et al. 2013), and metal/metalloid pollution in the aquatic environment has become a classical environmental problem in China. China's national seawater quality standards classify seawater into four grades (SEPA 2002): Grade I is suitable for marine fisheries, marine sanctuaries, and protected areas for rare and endangered marine organisms; Grade II can be used for aquaculture, bathing beaches, entertainment districts, and industrial water associated with human food processing; Grade III can only be used for industrial water and coastal scenic areas; and Grade IV is only suitable for ocean ports and engineered structures.

Metal/metalloid concentrations in coastal regions vary with the local economic development, pollution sources, and geographical conditions (Huang et al. 2013; Pan and Wang 2012). In China, the government has built up three most densely populated and industrialized economic zones and made it a priority by integrating all nearby cities/regions to foster economic development, this includes building an advanced communications network, better highways, increased education and scientific resources. The Pearl River Delta Economic Zone is in the expansive delta lands of the Pearl River at the South China Sea that consists of the most economically dynamic region since the launch of China's reform program in 1979, such as Guangzhou, Shenzhen, and Zhuhai. The Bohai (Bay) Economic Rim (BER) is an emerging economic powerhouse of Northern China surrounding the Bohai Sea, including areas in Liaoning, Hebei, and Shandong, etc. (Fig. 1). The BER has traditionally been involved in heavy industry and manufacturing, and now is becoming a significant growth cluster for the automobile, electronics, and petrochemical sectors. This region is recently undergoing major economic and infrastructural changes and is rivaling the Pearl River Delta in the south and the Yangtze River Delta in the east. The Yangtze River Delta Economic Zone (YRDEZ) is the economic region that encompasses the Shanghai municipality, Jiangsu, and Zhejiang province. Being a hub for the automobile industry, electronics, energy, iron, and steel industries, this region accounts for 20% of China's gross domestic product. The port of Shanghai is the world's busiest, and its volume is still increasing rapidly; also, Shanghai predominates in the automobiles and logistics industries.

Studies showed that generally seawater metal concentrations in south China was higher than in east and north China (Mao et al. 2009; Wan et al. 2007); the reason for the higher metal concentration can be the severer urbanization and anthropogenic activities near the coastal area. Generally, metal contamination is more serious in coastal regions in south China compared with north China both for seawater (Table 1) and sediments (Table 2). Metal concentrations in seawaters from South China Sea showed contamination to a higher extent (seawater as grade IV with Pb > 10 μg/L) than those from the north regions where seawater was classified as grade II with Pb within 5 μg/L) (Table 1). The highest concentration of Pb (14 μg/L at Sanya Bay, grade IV) was found in the South China Sea. Pb is known to be the most frequently encountered toxic metal in China, especially for long-term exposure (Cheung and Wang 2008; Dong et al. 2011). Metalloid As has caused great concern with high toxicity in terrestrial as well as aquatic environments. The highest concentration of As as 4.71 μg/L was reported at the Pearl River Estuary which was clean enough to be classified as grade I.

The highest concentration of Cd (1.04 μg/L at Liaodong Bay, grade II) and Hg (0.05 μg/L at Bohai Bay, grade II) was recorded at Bohai Sea in north China (Table 1). The health of the Bohai Sea has attracted a huge amount of attention in recent years due to the high input of anthropogenic pollutants from adjacent terrestrial areas (Huang et al. 2013).

Cr contents in the surface waters of Jiaozhou Bay were reported recently in the range of 0.44–1.56 μg/L (Yang et al. 2015). Cr concentration in the Yangtze River Estuary of east China was 4.54 μg/L (Yin et al. 2016) as shown in Table 1, higher than other coastal areas. The high population and dense industrialization has caused high metal discharge to the eastern coastal areas. Generally, in coastal seawater, the Hg concentration was very low except in Bohai Bay; it reached the upper limit of the first class criterion 0.05 μg/L (Table 1).

The National Sediment Standard of China (SEPA 2002) has defined marine sediment quality into three grades (Table 2): the first class quality is suitable for mariculture, nature reserves, endangered species reserves, and leisure activities; the second class quality is suitable for industry and tourism sites; and the third class can only be used for harbors.

Table 2 shows metal concentrations present in sediments from different coastal areas of China. Generally, all sediment showed high contamination of Cd and Pb, which was consistent with the status of metal contamination in seawater (Table 1). In the aquatic environment, As is derived from natural (volcanism and weathering) and anthropogenic sources. Though for both As and Cr, the seawater showed no contamination with the water of grade I quality; in sediment, however, marked pollution by As and Cr was observed; the highest As concentration (397 mg/kg) exceeded the upper limit of the grade III criteria by 327%. Riverine input and atmospheric deposition of As from natural and anthropogenic sources transport it into the aquatic environment. In north China, the As concentration in sediments from Jinzhou Bay is higher than other areas, which was polluted mainly by a smelting plant and the surrounding land-based sources (Chalmers et al. 2011).

Both of the highest concentration for Pb and Cr was recorded in sediments from the South China Sea, which was respectively 1088 and 437 mg/kg dry wt being of quality worse than grade III. Domestic and industrial products are rich in Cr. Metal concentration of Cd, Pb, and As in sediments from the Bohai Sea also recorded the quality of worse than grade III, with the value of 248, 753, and 397 mg/kg dry wt, respectively

Table 1 Metal concentrations in water from coastal areas of China and guideline values used to distinguish water quality (μg/L)

	Sample Sites	Cd	Pb	Hg	As	Cr	References
Bohai Sea	Jinzhou Bay	0.92	0.61	0.030	2.19		Wang et al. (2012)
	Liaodong Bay	1.04	4.91				Wan et al. (2007)
	Bohai Bay	0.20	4.43	0.05			Mao et al. (2009)
Yellow Sea	Yellow Sea	0.078	0.37	0.0036			He et al. (2008)
	Yellow River	0.68	0.51		0.92		Tang et al. (2010)
	Yangtze River	0.092	1.34	0.0078	3.05	4.54	Yin et al. (2016)
East China Sea	Hangzhou Bay	0.12	1.74				Jin and Shao (2003)
	Luoyuan Bay	0.04–1.0	0.35–1.70		1.41–2.98		Wang et al. (2010a)
South China Sea	Shantou	0.23–0.42	0.2–5.9		1.2–1.2		Zhang et al. (2015)
	Dapeng Bay	0.08	0.34				Huang et al. (2005)
	Hong Kong	0.0545	0.2376				Chan (1995)
	Pearl River Estuary	0.20	3.08		4.71		Zhang et al. (2013)
	South China Sea	0.04–0.91	0.03–10.2		0.20–2.3	0.31–0.86	Li et al. (2013)
	Sanya Bay	0.07–0.69	1.98–14.0			0.17–4.38	Yang et al. (2004)
Guideline values (GB3097-1997)	Grade I*	1	1	0.05	20	50	SEPA (2002)
	Grade II*	5	5	0.2	30	100	SEPA (2002)
	Grade III*	10	10	0.2	50	200	SEPA (2002)
	Grade IV*		50	0.5		500	SEPA (2002)

*Values are the upper limit for the grades. Cd and As have only three grades

(Table 2). These metal contents demonstrated the serious pollution in the Pearl River Delta Economic Zone and the Bohai (Bay) Economic Rim. Sediment from the Yangtze River Estuary had the extremely high Hg concentration of 10.1 mg/kg dry wt, which was more than ten times the upper limit criterion (1 mg/kg dry wt) of grade III sediment quality; moreover, sediments in Daya Bay also had extremely high Hg concentration (up to 9.73 mg/kg dry wt). This is really noteworthy because exposure to mercury—even small amounts—may cause serious health problems. With the rapid economic growth, China is considered to be an important anthropogenic source of mercury (Hg) emission in the world (Pacyna et al. 2010). A significant amount of the Hg emission in China is from nonferrous metal smelting and coal consumption.

The highest Cd (248 mg/kg, worse than grade III) and As (397 mg/kg) are found in sediments from the Bohai Sea in north China (Zhang et al. 2008). The Bohai Sea in north China is one of the most important fishing grounds in this country; while in recent years, surrounded by one of the biggest economic rims of China, its ecological functions have been declining rapidly in the recent two decades under the heavy anthropogenic impacts. The remarkably high concentrations of metals were almost all recorded in numbers of estuarine sites (Gao et al. 2013).

The pollution status of metals and metalloid is serious in China. The source prevention of contamination, the precautionary principle, and the licensing of wastewater discharges by competent authorities have become key elements of successful policies for controlling, reducing, and preventing the discharge of toxic substances including metals from all sources into the coastal and estuarine environments.

Evaluation of metal contamination in coastal environments

Metals and metalloid can cause harmful effects in the environment; they are introduced and in the living body when exposed. There are many different methods for analysis of single and combined metal concentrations and their effects (Huang et al. 2013), such as the Enrichment Factor (EF), Contamination Factor (CF), and Pollution Load Index (PLI) (Table 3). Some indices are more concerning the potential risk to organisms, including Effects Range-Median (ERM) guidelines, Mean Effect Range Median Quotient (mERM-Q), and Mean Probable Effect Level Quotient (mPEL-Q) (Hirose 2006).

Evaluation of single metal contamination status and effect

Contamination factor (CF) is used to assess the contamination degree of a given toxic substance at a station according to its total content. It can be determined as follows (Long et al. 1995):

$$\mathrm{CF} = C_{\mathrm{m}}/C_{\mathrm{b}} \quad (1)$$

Table 2 Metal concentrations in sediments from coastal areas of China and guideline values used to distinguish sediment quality (mg/kg dry wt)

	Sample Sites	Cd	Pb	Hg	As	Cr	References
Bohai Sea	Jinzhou Bay	248	753	–	397	60.6	Zhang et al. (2008)
	Liaodong Bay	0–9.7	6.7–131	–	–	–	Zhou et al. (2004)
	Bohai Bay	0.22	34.7	–	–	101	Gao and Li (2012)
	Haihe Estuary	0.4–2.1	21.7–78.9	–	–	63.2–155	Liu et al. (2006)
Yellow Sea	Swan Lake lagoon	0.23–0.52	23.1–45.9			33.8–52.0	Huang et al. (2013)
	Rongcheng Bay	0.07–0.28	17.4–51.1	–	–	23.5–48.6	Huang et al. (2013)
	Yellow Sea	0.1	16.3	0.02	6.9	–	He et al. (2009)
	Yellow River	0.27–1.43	1.21–20.69	–	–	87.84–169.70	Ren et al. (2015)
	East China Sea	–	10–49	–	–	–	Fang et al. (2009)
	Yangtze River Estuary	0.23	27	10.1	–	86	Zhao et al. (2008)
East China Sea	Hangzhou Bay	0.04–8.4	2.0–175	–	–	–	Che et al. (2003)
	Luoyuan Bay	0.12–0.2	28.2–32.2	0.04–0.08	9.0–13.7	33.4–37.7	Lin (2008)
	Quanzhou Bay	0.28–0.89	34.3–100.9		17.7–30.2	51.1–121.7	Ruilian et al. (2008)
	Xiamen	1.74–17.2	1.24–5.49	–	–	0.10–1.16	Lin et al. (2014)
South China Sea	Shantou	0.30–1.8	35.6–64.9	–	–	36.1–74.2	Qiao and Huang (2009)
	Daya Bay	18.68–89.58	–	5.15–9.73	14.82–44.69	0.002–0.114	Yang et al. (2014)
	Dapeng Bay	0.2	9.4	0.03	–	24	Huang et al. (2005)
	Shenzhen Bay	1.2–6.6	12.0–63.0	–	–	40.0–168	Zuo et al. (2009)
	Hong Kong	0.00–3.5	17.1–90.7	–	–	10.5–41.6	Liang and Wong (2003)
	Pearl River Estuary	–	54.7	–	–	107	Yu et al. (2010)
	Hailing Bay	0.1–0.4	16.8–56.1	0.01–0.10	–	–	Qiu et al. (2004)
	Beihai	–	7	< 0.04	< 3	9	Vane et al. (2009)
	South China Sea	0–3.5	2–1088	0.01–0.63	0.7–42.3	8–437	Wang et al. (2013)
	Sanya Bay	0.13	17.5	–	7.1	12.4	Qiu and Yu (2011)
Guideline values	Grade I	0.5	60	0.2	20	80	SEPA (2002)
	Grade II	1.5	130	0.5	65	150	SEPA (2002)
	Grade III	5	250	1	93	270	SEPA (2002)

− not available

where C_m is the concentration of a specific metal in the tested sample, and C_b the background concentration of that metal. The pollution status of the sample can be classified into four categories by CF values (Table 3): CF < 1, low contamination factor; 1 ≤ CF < 3, moderate contamination factor; 3 ≤ CF < 6, considerable contamination factor; and CF ≥ 6, very high

Table 3 Methods for sediment quality classification and potential risk evaluation

Index	Degree of contamination					References
	Low	Moderate	Considerable	High	Very high	
CF	< 1	1–3	3–6		≥ 6	Hakanson (1980)
EF	≤ 5	5–10	10–25		> 25	Sutherland (2000)
PLI*	< 1	> 1				Tomlinson et al. (1980)
PER	≤ 40	40–80	80–160	160–320	> 320	Hakanson (1980)
PERI	≤ 150	150–300	300–600		> 600	Hakanson (1980)
mERM-Q (risk probability)	≤ 0.1 (9%)	0.1–0.5 (21%)	0.5–1.5 (49%)		> 1.5 (76%)	Long et al. (1995)
mPEL-Q	≤ 0.1 (8%)	0.11–1.5 (21%)	1.51–2.3 (49%)		> 2.3 (73%)	Long et al. (2000)

*PLI value of < 1 indicates no pollution whereas > 1 indicates pollution of sediments contamination factor (Long et al. 1995).

Enrichment factor is another index used for evaluating the extent of heavy metal contamination through anthropogenic sources. The EF method normalizes the measured element content with respect to a sample reference metal such as Fe or Al (Ravichandran et al. 1995), which is calculated as follows:

$$\mathrm{EF} = (M/\mathrm{Fe})_{sample}/(M/\mathrm{Fe})_{background} \tag{2}$$

where $(M/\mathrm{Fe})_{\mathrm{sample}}$ is the metal to iron ratio in the sample, while $(M/\mathrm{Fe})_{\mathrm{background}}$ is the background value of metal to iron ratio. Fe is the most widely used normalizer because iron oxides show strong positive association with most trace metals and its natural concentration tends to be uniform (Ravichandran et al. 1995).

It is demonstrated that metals of anthropogenic sources are prone to be distributed in more reliable fractions in the sediments, thus of hither bioavailability, i.e., has higher EF values (Huang et al. 2013). EF also classifies the sample quality into four categories of pollution status: low, moderate, considerable, and very high (Table 3). Because it has normalized the metal content by reference element, it can more precisely reflect the given metal's origin. EF has been used to examine the metal contamination in Bohai Bay; the results showed that the order of EF values in clayey silt sediments in Bohai Bay descends in the sequence of Cr (2.84) > Ni (2.20) > Cd (1.74) > Cu (1.69) > Pb (1.66) > Zn (1.61), indicating a low level of anthropogenic contamination and the dominantly natural origin of these metals (Gao and Li 2012).

Max/Min the ration of the highest value to the lowest one

Table 4 showed the enrichment of Cd and Pb at stations St-1–St-5 in the study of Saleem et al. (2016). Index CF demonstrated very high pollution (CF > 6) of Cd and moderate pollution (1 < CF < 3) of Pb in sediments from all the 5 stations; the highest level of sediment pollution of Cd and Pb was recorded at St-5 (CF = 11.9) and St-1 (CF = 2.9), respectively. While when the sediment quality was evaluated using EF, for which the natural geographical influence was eliminated by content of Fe, the severest pollution of both Cd and Pb was consistently found at St-5, with the respective highest EF value of 19.73 (10 < EF < 25, considerable contamination) and 3.34 (EF < 5, low contamination) (Table 4).

Potential Ecological Risk (PER) is an empirical method used to evaluate the potential toxicity of one element by multiplying the CF with a parameter (i.e., Toxic Response Factor) according to the element's potential toxicity; it represents the sensitivity of the biological community to one toxic substance (Hakanson 1980). Potential Ecological Risk (PER) is determined as follows:

$$\mathrm{PER} = \mathrm{Trf} \times \mathrm{CF} \tag{3}$$

where Trf = Toxic Response Factor, the value of which depends on the sedimentological toxic factor that differs for all the metals depending on their potential toxic effect, e.g., Trf for Pb, Cd, and Cr was 5, 30 and 2, respectively.

The quality of sediments can be classified into five categories using the PER index (Table 3). Pb and Cd in the sediments around Dongjiang Harbor in Tianjin city, China, were reported originating from natural resources, and had low potential risk with PER << 40 (Guo et al. 2010), whereas the respective PER value of Cd and Pd in sediment at stations along the Helmand River was 156 and 1.7, indicating the considerable potential risk caused by Cd contamination (80 < PER ≤ 160) (Ghaleno et al. 2015).

Evaluation of combined metal contamination status and potential effects

In sediments, trace elements occur as complex mixture with great variation; therefore, the pollution load index (PLI) was proposed (Tomlinson et al. 1980) to determine and compare the combined contamination status of

Table 4 Contamination factor (CF) and enrichment factor (EF) of Pb and Cd for sediments collected from the Hub estuary (Saleem et al. 2016); based on these data, the present study calculated the Pollution Load Index (PLI), Potential Ecological Risk Index (PERI), Mean Effect Range Median Quotient (mERM-Q), and Mean Probable Effect Level Quotient (mPEL-Q) at different stations

Station	CF		EF		$\mathrm{PLI}_{\mathrm{CF\text{-}based}}$	$\mathrm{PLI}_{\mathrm{EF\text{-}based}}$	PERI	mERM-Q	mPEL-Q
	Cd	Pb	Cd	Pb					
St-1	8.4	*2.9*	10.27	2.68	4.8	5.2	267	0.15	0.32
St-2	9.95	1.88	8.24	1.56	4.3	3.5	308	0.16	0.34
St-3	8.8	2.51	6.56	1.87	4.6	3.5	277	0.16	0.35
St-4	*6.95*	*1.47*	*5.36*	*1.14*	*3.1*	*2.8*	*216*	*0.11*	*0.25*
St-5	*11.9*	2.01	*19.73*	*3.34*	*4.8*	*8.1*	*367*	*0.18*	*0.40*
Max/Min	*1.7*	*2.0*	*3.7*	*2.9*	*1.5*	*2.9*	*1.7*	*1.6*	*1.6*

Entries in italic indicate the highest and lowest values; the ratio of the highest to the lowest gives the value Max/Min in the last line complex toxicant groups. It is calculated by the *n*th root of the product of the *n* CF or *n* EF for the tested metals using the following equation.

$$\mathrm{PLI}_{\mathrm{CF}-Based} = (\mathrm{CF}_1 \times \mathrm{CF}_2 \times \mathrm{CF}_3 \times \cdots \times \mathrm{CF}_\mathrm{n})^{1/n} \quad (4)$$

$$\mathrm{PLI}_{\mathrm{EF}-Based} = (\mathrm{EF}_1 \times \mathrm{EF}_2 \times \mathrm{EF}_3 \times \cdots \times \mathrm{EF}_\mathrm{n})^{1/n} \quad (5)$$

where CF_n or EF_n is the CF or EF value of metal *n*.

The PLI values < 1 indicate that the area has not been severely impacted by anthropogenic contamination or other pollutant source. We emphasize on anthropogenic activities because the bioavailability of trace elements entering the environment through anthropogenic sources is comparatively higher than other natural sources (Deng et al. 2010). The PLI value > 1 indicates the sampling region is polluted, whereas PLI < 1 no pollution. We use the metal content at the Hub estuary as an example, (in Table 4); the EF-based PLI value of 8.1 at St-5 demonstrated it was the most seriously polluted station, followed by St-1 with the $\mathrm{PLI}_{\mathrm{EF\text{-}based}}$ of 5.2; while with the same CF-based PLI values at St-5 and St-1 (both was 4.8), the same pollution was concluded; thus, no priority site can be decided during the environmental management and pollution control. As we have discussed previously, being different from the result of CF which indicated the very high pollution (by Cd) and moderate pollution (by Pb) of the sediments with the highest pollution level at different stations, EF values demonstrated the severest pollution of both Pb and Cd at St-5 (Table 4, Saleem et al. 2016). When we chemically evaluated the combined pollution by Cd and Pb at each station, both CF-based and EF-based PLI value indicated the most severe pollution at St-5 and the slightest pollution at St-4 (with PLI value of St-5 1.5 times or 2.9 times that of St-4, Table 4). It is noteworthy that the CF-based PLI showed the multiple contamination at each station descends in the sequence of St-5 > St-1 > St-3 > St-2 > St-4, while for the EF-based PLI it was St-5 > St-1 > St2 = St3 > St4. It is well known that the EF index is a most often used technique to separate the metals of natural variability from the metal fraction associated with anthropogenic activities; the EF-based PLI value thus may be a more convenient and feasible tool for plotting the contamination trends in sediments across large geographic areas; therefore, it is recommended to use EF for determining the PLI value to study multiple contaminations of metals in combined form.

The PLI value tell us the evaluation of the contamination status at a region or station based on the total element concentration; in order to know more on the potential risk of these elements to the organisms or to ecological systems, the index of potential ecological risk (PER) can be adopted, and PERI is the sum of PER values calculated for all the metals inside one area (Hakanson 1980). PER values for Cd, Pb, Hg As, Cu, and Zn in the sediments of station 1 around Dongjiang Harbor (Tianjin) in the Bohai Sea were 11.1, 0.75, 80, 10.3, 2.60, and 1.17, respectively (Guo et al. 2010); thus, we figured out the Potential Ecological Risk index (PERI) along that station was 105.92 that is within the threshold value of low contamination limit (150) (Table 3). While the PER value for Cd, Pb, Hg, As, Cu, and Zn in the sediments of station 2 at this harbor was 17.7, 1.65, 216, 14.8 3.10, and 1.16, respectively, resulted in the PERI value of 254.41, indicating that station 2 had moderate ecological hazards and was contaminated to an higher extent compared with station 1. However, PER and PERI can only be comparable and thus can be used to distinguish among different regions or stations when they have the same studied elements.

Effects range-median (ERM) are values corresponding to the 50th (median) percentile of biological effects concentrations compiled from a variety of studies (Ritter et al. 2012). It is the threshold above which adverse effects on biota are frequently observed. The ERM guideline value for each metal is Cd 9.6 mg/kg, Pb 218.0 mg/kg, Hg 0.7 mg/kg, As 70.0 mg/kg, and Cr 370.0 mg/kg (Long et al. 1995). Below the ERM value represents a "Possible-effects" range within which effects would occasionally occur, while the concentrations equivalent to and above the ERM value represent a "Probable-effects" range within which effects would frequently occur. The incidence of adverse effects can be quantified by dividing the number of "effects" entries by the total number of entries and expressed as a percentage (Long et al. 1995).

Mean Effect Range Median Quotients (mERM-Q) have been developed from the biological toxicity test of the benthic environment to estimate possible adverse biological effects of sedimentary contaminants in coastal regions at individual levels (USEPA 2001). It can be used by calculating mean quotients adverse effects to marine organisms caused by individual chemicals included in the sediment quality guidelines (SQGs) list. The mERM-Q is designed for analyzing the potential effects of multiple trace element contamination in sediments (Pan et al. 2014), and is calculated as:

$$mERM{-}Q = \sum[C_x/\mathrm{ERM}_x]/n \quad (6)$$

where C_x is the concentration and ERM_x is the ERM value for the selected metal *x*; *n* is the number of metals.

According to the potential ecological risks of combined metal contaminations, the mERM-Q classifies the polluted sites into four levels: low priority site (mERM-Q ≤ 0.1), medium-low priority site (0.1 < mERM-Q ≤ 0.5), high-medium priority site (0.5 < mERM-Q ≤ 1.5), and high priority site (mERM-Q > 1.5), which indicate that the combined effects of contaminants have a 9, 21, 49, and 76% probability of being biologically toxic, respectively. In the Hub river, mERM-Q values less than 0.5 for the surface sediments showed the probability of toxicity to benthic organisms in the range 9–21% (Saleem et al. 2016). Many studies have proved that index mERM-Q is an effective indicator of ecosystem health under the threat of combined metal contaminations (Huang et al. 2013 and reference therein). With mERM-Q values for sediment in Table 4, it can be known that the combined contamination of Cd and Pb in the sediments gave a 21%probability of being biologically toxic.

Probable effect level (PEL) is specific guideline values for metals, e.g., the PEL for Cd is 4.2 mg/kg dry wt, and for Pb 110, Hg 0.7, As 42, and Cr 160 mg/kg dry wt (Environment Canada and Ministère du Développement durable, de l'Environnement et des Parcs du Québec 2007). The mean PEL quotient has been proposed and adopted to determine the possible biological effect of combined toxicant groups by calculating the mean quotients for a range of contaminants using the following formula (Luo et al. 2010):

$$mPEL\text{–}Q = \sum[C_x/\mathrm{PEL}_x]/n \tag{7}$$

where C_x is the sediment concentration of component x, PEL_x is the PEL for element x, and n is the sum of elements. For marine sediments, the mean PEL quotients of < 0.1, 0.11–1.5, 1.51–2.3, and > 2.3 indicate that the combined effects of contaminants have an 8, 21, 49, and 73% probability of being biologically toxic, respectively (Qiao and Huang 2009). In order to compare among all the commonly used methods, we have calculated the results and summarized them in Table 4. The mPEL-Q value for Cd and Pb in the investigated 5 stations indicated 21% probability of causing biologically reverse effect to organisms at station 5, with the highest value of 0.40 (ranged between 0.11 and 1.5). This conclusion is the same as that of assessment using mERM-Q, which also indicated 21% probability of ecological risk for the combined metal contamination.

To sum up, researchers have proposed considerable approaches to quantify the impact of anthropogenic pollution on a given site or in a given region, which has significant subsequent implications for the overall assessment, monitoring, and management of interested pollutant(s). Many kinds of trace elements usually accumulate simultaneously and cause combined contamination, among which some elements have higher toxicity than others (e.g., Hg); thus, even a region demonstrated serious contamination by elements with low toxicity, maybe the potential ecological risk caused there is much lower than a region having low level of pollution by elements with high toxicity. It is thus critical to integrately evaluate the environmental quality both through chemical/concentration and biological/toxicity aspects, and consider both the single metal contribution and the combination of all the elements.

To set up feasible evaluation methods, we thus further made a comparison of the various indices by using them to evaluate the pollution of sediments from two typical bays in China (Jinzhou Bay in North China and Sanya Bay in South China), using data in Table 2. Sediments in Jinzhou bay in North China had a very high metal concentration in the sediments, while Sanya bay from South China had a low metal concentration in sediments. All evaluation methods (CF, EF, PLI, PERI, mERM-Q, and mPEL-Q) mentioned above were used for determining the contamination status and potential risk in these two Bays. The background value for Cd, Pb, As, and Cr was 1, 70, 15, and 90 (mg/kg), respectively (Zhang et al. 2008), and Trf value for Cd, Pb, As, and Cr were 30, 5, 10, and 2, respectively (Hakanson 1980). The iron background value for calculating EF was 34,800 mg/kg (Luo et al. 2013). The sample value of iron used in Jinzhou bay and Sanya bay were 43,000 mg/kg and 26,100, respectively (Wang et al. 2010b).

The metal concentrations in sediments is very high in Jinzhou bay (Table 1); consistently values for all the evaluation indices are also significantly high (Table 5): Except for Cr, for which both CF and EF values indicated its low level of contamination thus dominantly natural origin, an EF value of 16.3 labeled Pb as considerably polluted, while a CF value of 10.8 showed Pb was polluted at a very high level. While in Sanya Bay, CF and EF consistently showed the natural origin of all the metals Cd, Pb, As, and Cr.

Both CF and EF clearly showed the sequence of contribution to the combined station contamination was Cd > > As >Pb > Cr. Multiple contaminations of all these four metals in

Table 5 Comparison of different indices to determine the pollution status and potential risk in Jinzhou Bay and Sanya Bay

Sampling site	Metals	CF	EF	PLI		PERI	mERM-Q	mPEL-Q
				(CF based)	(EF based)			
Jinzhou Bay	Cd	248	375	14.77	22.34	7759.8	8.78	18.93
	Pb	10.8	16.3					
	As	26.5	40.0					
	Cr	0.67	1.0					
Sanya Bay	Cd	0.13	0.17	0.21	0.29	10.16	0.06	0.11
	Pb	0.25	0.33					
	As	0.47	0.63					
	Cr	0.14	0.18					

Jinzhou Bay sediments resulted in the high value of PLI of > > 1 showing severe contamination and dominant anthropogenic metal resources, PERI > > 600 indicating extraordinary high risk of these combined metal contamination, mERM-Q > > 1.5 indicating 76% probability of being ecological toxic, and mPEL-Q > > 2.3 indicating 73% probability of being ecological toxic (Table 5). Whereas for sediments in Sanya bay, the indices values are all extremely low except mPEL-Q (equals 0.11) (Table 5) which in contrast showed a moderate level of contamination with 21% probability of toxic risk to organisms. While mERM-Q showed low risk probability with the value of 0.06, which is a more reasonable assessment result compared with mPEL-Q.

Conclusion

Metal contamination in coastal areas is one of the most environmental concerns all over the world. With this most serious contamination in China, prevention and management of toxic metal pollution has been set as one of the key goals of government projects recently. We detailedly compared the commonly used pollution evaluation techniques using metal data from two typical coastal bays in China; the case study results demonstrated that the EF value successfully revealed the severe pollution, while CF failed to show this information; mERM-Q gave more accurate prediction compared to mPEL-Q in potential toxicity evaluation of contaminants. Accordingly, we proposed the approach of joint indices of the Enrichment Factor and Effect Range Median combined with the Pollution Load Index and Mean Effect Range Median Quotient (EEPME). The EF technique is for single element evaluation, and PLI as chemical index for combine metal contamination evaluation; mERM-Q technique is for potential ecological risk prediction. This battery of guidelines can provide a more accurate, consistent, comparable, and comprehensive understanding of the environment pollution status of combined metals and their potential ecological risk. Further studies to elucidate the mechanism of combined metals are needed for better understanding of multiple contaminations and the accurate potential ecological risk prediction.

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