

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

Metal Pollution in Coastal Sediments

Yu Qian^{1,3} · Weiguo Zhang² · Lizhong Yu² · Huan Feng¹

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Abstract Coastal sediment is a vital habitat for aquatic and marine life in coastal ecosystem. However, urbanization and economic development in coastal areas have resulted in environmental problems globally. Due to coastal development such as new industrial facilities and commercial port expansion, anthropogenic metals are introduced to the adjacent areas. Therefore, metal pollution in coastal areas is one of the focused environmental concerns. Sediment quality in coastal zone reflects the long-term environmental status because it keeps a record of the development in the area. In this review paper, sediment metal concentrations in 52 selected sites worldwide are summarized for evaluation of the coastal environmental quality. The results from this study can be applied to science-based policy formulation and ecological restoration/rehabilitation practices in an integrated coastal zone environmental management.

Keywords Coastal sediment · Metal · Enrichment factor · Hazardous quotient · Risk assessment

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Huan Feng fengh@montclair.edu

- ¹ Department of Earth and Environmental Studies, Montclair State University, Montclair, NJ 07043, USA
- ² State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai 200062, People's Republic of China
- ³ Present address: School of Ecology and Environmental Sciences, Yunnan University, Kunming, Yunnan 650091, People's Republic of China

Overview

Metals with different speciations and oxidation states are naturally present in the earth crust [1]. Depending on the rock type, geophysical condition, and geographical location around the world, metal concentrations in sediments, soils, and water present as naturally weathered products from the earth crust can be different from place to place [2, 3]. Trace amount of metals is essential micronutrients for the growth and metabolism of many organisms [4]. However, metals in excessive amount can be toxic to organism and cause ecological toxicity [4]. Anthropogenic metal input associated with the urbanization and industrialization in the twentieth century has increased dramatically in the coastal areas and is drawing increasing attention around the world because about 80 % of the pollutants from human activities are introduced into coastal environment [1, 5]. Therefore, metal pollution in the coastal sediments has become a major environmental problem because it threatens the economic and ecological value of the coastal area.

Coastal environment is a complex system involving physical, chemical, and biological processes that play important roles in metal biogeochemical cycle. However, anthropogenic input has caused metal pollution in the coastal area. The sources include mining, metal product fabrication, solid waste disposal, fossil fuel combustion, and municipal/industrial waste effluent [1, 6]. Industries, such as foundry, paper processing, laundry, tannery, and dye works, can emit toxic metals and discharge wastewater to the adjacent estuaries and coastal areas [7, 8]. Acid rain can leach metals and increase the mobility of toxic metals from pollutant sources to the environment [9]. Once enter coastal environment, the toxic metals will mainly accumulate in the sediments because of particle scavenging and settling. Therefore, high concentrations of toxic metals are often found in sediments in many industrialized urban coastal areas as a consequence of industrialization and urban development with population growth in the areas [10-14]. It is well known that fine-grained sediments are the main carriers of the toxic metals because of its higher specific surface area. Many studies have shown that coastal sediments are repository for metal pollutants and provide time-integrated records of pollution history [11, 15–19]. This paper summarizes the metal concentrations in coastal sediments in selected areas and reflects the environmental pollution and the potential ecological risk associated with urbanization and economic development.

Metal Concentrations in Coastal Sediments

In this review, sediment metal concentrations in 52 selected coastal sites around the world were summarized based on the published literatures (Table 1 and Fig. 1). These publications are based on original field studies in six continents (Africa, Asia, Australia, Europe, North America, and South America) including 20 countries, e.g., France [36], Spain [42, 43], Greece [37, 38], Turkey [24], Albania [35], Italy [39, 41], USA [7, 47-49], French Guiana [53], Mexico [46], China [12, 26–28], India [30, 31], Korea [29], Azerbaijan [23], Iran [23], Kazakhstan [23], Russia [23], Fiji [34], Australia [33], and Africa [20]. Table 1 summarizes sediment metal concentrations in selected areas around the world. Average metal concentrations at each site range from 0.096 to 10.9 % for Al, 0.21–13.8 % for Fe, 3–81 kg⁻¹ for Ag, 0.04–998 mg kg⁻¹ for Cd, 1.0–463 mg kg⁻¹ for Cr, 0.5– 604 mg kg⁻¹ for Cu, 0.01–1.8 mg kg⁻¹ for Hg, 0.4–4, 643 mg kg⁻¹ for Mn, 2–240 mg kg⁻¹ for Ni, 3–2, 369 mg kg^{-1} for Pb, and 7–4,430 mg kg⁻¹ for Zn, respectively (Table 1). The wide variations in toxic metal concentrations reflect the different natural mineral compositions in the sediments as well as anthropogenic input in some of these coastal areas.

Sediment Pollution Assessment

Excess amount of toxic metals in environment can cause pollution problems in coastal areas. To evaluate metal pollution in the sediments, metal enrichment factor (EF) has been widely used as the assessment criteria to screen sediment metal concentrations of environmental concern (e.g., [7, 11, 12, 28, 55–60]). Because EF values can be used to distinguish natural metal concentrations from those of anthropogenic origin, it is widely used for sediment quality assessment (e.g., [7, 11, 12, 55, 57, 58, 60, 61]). Mathematically, it is expressed as (e.g., [62]):

$$EF = \frac{\left(\frac{Me}{Al \text{ or } Fe}\right)_{Sample}}{\left(\frac{Me}{Al \text{ or } Fe}\right)_{Background}}$$
(1)

where Me is the metal concentration of concern, $\left(\frac{Me}{Al \text{ or } Fe}\right)_{Sample}$ is the metal to Al or Fe ratio in the sample, and $\left(\frac{Me}{Al \text{ or } Fe}\right)_{Background}$ is the metal to Al or Fe ratio in the background. Al and Fe are used here as geochemical normalization elements because both Al and Fe are the most abundant elements in the earth crust [2, 3]. The metal background concentration should be derived from the sampling site if available [63]. However, the background data are not readily available in most of the cases. For metal EF calculation, when the local metal background values are not available, the upper continental crust values can be used as the background values [64–71]. In this case, metal concentrations in the upper continental crust [3] were adopted as alternatives for the background values.

Metal EF values indicate the extent of metal enrichment in the sediments and can be used as sediment assessment reference criteria [60, 72, 73]. As a simple guideline, $EF \approx 1$ indicates natural crustal origin, whereas EF>10 suggests anthropogenic source [74]. Zhang and Liu [60] also recommend that 0.5<EF<1.5 suggests that the metals may be entirely from crust natural weathering processes, whereas EF>1.5 indicates that a significant portion of the metal is delivered from non-crustal materials. In another classification, metal EF values are divided into five categories based on the degree of enrichment, i.e., (1) EF<2 suggests deficiency to minimal enrichment, (2) EF=2-5, moderate enrichment, (3) EF=5-20, significant enrichment, (4) EF=20-40, very high enrichment, and (5) EF>40, extremely high enrichment [72].

The estimation of metal EF relies on the data availability of the geochemical normalization element in the site of interest. We managed to estimate the metal EF values for 38 selected coastal areas around the world. The results show that metal EF values vary widely from minimal to extremely high enrichment (Fig. 2). Specifically, the EFs calculated from the average metal concentrations at each site range from 7.2 to 43 for As (n=9), 0.5–1,582 for Cd (n=26), 0.1–15 for Cr (n=31), 0.07–29 for Cu (n=34), 0.02–5 for Mn (n=26), 0.3–56 for Ni (n=33), 0.6–130 for Pb (n=31), and 0.8–44 for Zn (n=38), respectively (Fig. 2).

As shown in Table 2, cases of moderate to high metal enrichment (EF>2) account for 19–100 %, depending on the metals, of which relatively high

Table I Sum	mary or meral conce	summary or mean concentrations (unit, ing kg) / in selected rivenite, estuarine, and marine sediments around the world	eu livellie, esui						
Continent	Country/region	Location		Al	Fe	As	Hg	Cd	Cr
Africa	Egypt	Suez Gulf (sandy sediment)	Mean±SD		$2,240\pm3,789$			9.92 ± 3.74	32.0 ± 18.3
			Range		385 - 13,549			5.63 - 17.1	18.1 - 85.0
	Egypt	Suez Gulf (muddy sediment)			$7,497 \pm 8,902$			13.34±6.5	64.2 ± 29.0
	Morocco	Lagoon of Oualidia	Mean±SD	108.600 ± 25.400	010,9–012 69.100±8.900		0.7 ± 0.3	Q.42-1C.0	52.5±6.1
		0	Range	52,600–143,600	46,200–79,000		0.1 - 1.2		46.0-62.0
	Morocco	Loukkos			$20,800\pm4,100$			1.34 ± 0.49	
	Morocco	Sehon			16,000-25,900 29.200 ± 2.100			0.90-2.17 2.30 ± 0.70	
		5			24,100–39,000			1.50 - 3.10	
	Morocco	Bou Regreg			$33,700\pm 2,500$			$2.30 {\pm} 0.70$	
	Managao	Otime an Dhio			26,800-40,300 14 000+4 €00			1.10-3.00	
Asia	Azerbaijan	Caspian Sea	Mean±SD	$69,200\pm7,690$	$37,100\pm3,800$	14.7 ± 4.2	0.2 ± 0.1	0.14 ± 0.03	85.3 ± 11.3
			Range	50,000 - 81,300	29,300-43,500	8.9–22.6	0.1 - 0.5	0.08 - 0.19	56.4 - 100
	Iran			$60,500\pm11,400$	$35,500\pm 5,950$	12.5 ± 3.0	0.1 ± 0.0	0.16 ± 0.03	85.2±15
	V azalchatan			37,500-77,500	22,200-44,000 6 730+5 130	7.0-20.1	0.0-0.1	0.10-0.24	59.6 - 128 2.14 ± 10
	Nazakustan			1.070 ± 5.100	$0,70\pm 3,150$ 1.940-28,000	2.1±2.0.2	0.0±0.0 ≪0.01−0.0	0.01-0.25	1.9 - 103
	Russia			$18,800\pm 8,510$	$5,520\pm 1,840$	3.0 ± 2.0	0.0 ± 0.0	0.06 ± 0.02	32.0 ± 19
				2,650 - 38,300	1,600-9,680	0.4 - 6.7	0.0 - 0.1	0.02 - 0.10	2.1–69
	Turkey	Marmara Sea	Mean±SD		$10,909 \pm 4,548$			$0.50 {\pm} 0.12$	47.8 ± 18.1
			Range		5,956 - 14,896			<0.02-0.62	27.2–61.5
	Israel	Mediterranean Coastal Region			2,090±852 1.000 5 806		0.0 ± 0.2	0.29±1.44	
	China	Minoijano Estuary	Mean+SD		1,000,0-000,1		1.0-0.0	0.04-008	
	СШПА	IVIIIIBJIAIIB LOUALY	Range					0.09-0.34	
	China	Xiamen-Jinmen	Mean±SD					0.2055 ± 0.05044	
			Range					0.15-0.285	
	China	Pearl River estuary	Mean±SD Pance						
	China	Western Bohai Bay	Mean±SD		$37,180\pm 2,647$			0.13 ± 0.01	52.0±2.9
			Range		28,700-40,500			0.09-0.16	38.2-63.2
	China	Haihe River Estuary			29,5002,828			0.170.01	57.55.4
	Chino	Dout of Tioniin			24,00036,000 24 7222 862			0.160.19	51.064.6 62 10 4
	СШПА				28.40036.400			0.120.25	52.079.8
	China	Yongding River estuary			31,75010,111			0.130.01	54.52.4
					22,20041,400			0.100.17	46.464.2
	China	Yangtze River intertidal	Mean±SD	$65,375\pm11,467$	$33,394\pm6,257$			0.26 ± 0.13	78.9±19.7
	71		Kange	40,803-97,213	20,238-49,627			c/.0-71.0	50.9-175
	Norea	Masan Bay	Mean±>D Range					0.10-7.47	0/.1 30.5–99.8
	India	Alang-Sosiya coast intertidal	Mean±SD		$137,990{\pm}78,573$			32.7±2.8	290.2 ± 63.2
		Gulf of Mannar	Mean±SD		$12,422\pm 7,246$			$0.20 {\pm} 0.06$	183.0 ± 199.2
		;	Range		3,600 - 34,600			0.06-0.35	34.0-1,038.0
	Russia	Barents Sea surface sediments		$46,700\pm14,600$	29,600±19,300 5 100 50 900	55.0±77.0	0.0±0.0	0.04 ± 0.03	65.0 ± 35.0
				21,300-00,000	0,00-001,0	0.000-0.0	0.0-0.0	60.0-70.0	1/.0-11/.0

Table 1 (continued)	ned)								
Continent	Country/region	Location		Al	Fe	As	Hg	Cd	Cr
Australia Australia	Australia Fiji	Bells Creek catchment Suva Harbor	Range Mean±SD	955–9,821	9,400-41,000 $31,533\pm9,300$	3.0-10.8			1.0–26.0
Europe	Montenegro	Adriatic Albanian coast	Mean±SD Pange		14,000-46,700 44,060±14,809 24.600-61-100		0.2 ± 0.3	0.22 ± 0.13	313 ± 52
	France	Cajarc Site, Lot River	Mean		Z4,000-01,100		711.0-000.0	125	10+
	France	Marcenac Site, Lot River	Kange Mean					0.81	
	France	Temple Site, Lot River	Kange Mean					0.33–1.40 20.4	
	ereere	Candarli Gulf surficial	Range Mean+SD	18 241+6 438	26 781+5 259	23 4+6 4	1 0+1 4	0.56–180	41 2+12 7
		sediments	Range	3,800-25,300	16,200-36,000	11.0–37.0	0.2-6.3		15.8-71.1
	Greece	Keratsini Harbor, Saronikos	Mean±SD			109.9 ± 359.6		998 ± 329	463.5 ± 145.4
	Italy	Gulf of Manfredonia	Kange Mean±SD			n.d1, &15.0 16.0 ± 7.0		190-/63	264.0-860.0 81.0±24.0
	•		Range			2.0-57.0			15.0-122.0
	Italy	Naples City Port	Mean±SD P			15.7±3.3		0.90 ± 0.64	72.5±42.7
	Italy	Taranto Gulf	Kange Mean±SD	58,564±11,247	$31,566\pm 2,903$	0.12-0.0	0.10.1	00.2-02.0	10.5-101.8 85.9±7.7
	•		Range	36,866–72,020	26,313 - 36,098		0.00.4		75.2-102.8
	Spain	Estuary of Huelva and	Mean±SD			180.7±255.6	1.8 ± 3.2	3.25 ± 3.06	52.0±39.9
		adjacent Atlantic shelf	Kange			12.0-850.0	0.1-11.6	1.00-9.00	16.0-150.0
	umde	Guil 01 Caulz	Range		271-39.820	9.3±2.0 5.1–13.7	0.1-1.0	0.39–1.24 0.39–1.24	39.7–283.9
	Spain	Odiel River	Mean±SD		$31,862\pm21,718$				54.7±52.4
			Range		11,601-88,429				7.9–234.6
	UK	Tees Estuary						6.13 ± 1.93	242.7±139.9
North America	USA	Baia California. California	Mean±SD	57.000 ± 14.927	27.200 ± 7.200			2.600−9.80 0.17±0.12	$0.7.0\pm164.6$
			Range	36,100-85,500	13,600-46,000			0.08 - 0.64	55.9-802.0
	USA	Hudson River	Mean±SD	$51,264\pm 5,980$	$34,348\pm 5,598$			1.00 ± 0.56	
	V STI		Kange	41,000-63,200	20,300-42,800	C F - F O F		0.18-2.29	702-107
	ASU ASU	California, 110al sait marsn	Mean±>D Ran∞e	21,220±8,218	$52,080 \pm 10,514$	10.1 ± 4.2 5 1-15 9		0.46 ± 0.11 0.18-0.26	(0.5 ± 19.0)
	USA	Upper Gulf of California	Mean±SD		$17,298\pm 8,091$				28.1 ± 13.0
	USA	Colorado River Delta	Mean±SD		$16,740\pm 6,138$				27.6±8.8
	USA	Oyster Rock Landing salt	Mean±SD	46,067±6,754					
	¥ SI I	marsh in Delaware	Range	43,000–55,900					
	AGU	wolle Ulade Delaware sait	Mean±SU Dango	20,0/0±±/,01/ 17 000 20 700					
	Canada	ulatsu Vancouver Harbor	Mean	1 /,000-20, /00				0.37	62.0
South America	Brazil	Jacarepagua Basin	Mean±SD					1	91.7 ± 69.1
) 4	Range						25.0-163.0
	Brazil	Lower Paraiba do Sul estuary	Mean±SD		$9,500\pm900$				90.0±13.0
	French Guiana	Kaw mangroves	Kange Mean±SD		9,000-11,400 47.245 ± 4.077		0.3 ± 0.0		/5.0-114.0 55.6±8.3
		þ	Range		39,762-59,922		0.2 - 0.3		39.5-67.1

Table 1 (continued)	(pər								
Continent	Country/region	Location		Al	Fe	As	Hg	Cd	Cr
Earth's crust ERL ERM	French Guiana French Guiana	Guiana shoreface sediments Sinnamary mangroves	Mean±SD Range Mean±SD Mean	80,400	50,144±558 49,530−51,317 44,062±5,361 35,000	1.5 8.2 70.0	$\begin{array}{c} 0.2\pm0.0\\ 0.2-0.2\\ 0.4\pm0.5\\ 10^{\prime}a\\ 0.2\\ 0.7\\ 0.7\end{array}$	0.098 1.2 9.6	64.5±1.0 63.4-65.5 59.8±11.4 35.0 81.0 370.0
Continent	Country/region	Location		Cu	Mn	Ni	Pb	Zn	References
Africa	Egypt	Suez Gulf (sandy sediment)	Mean±SD Range	33.2 ± 37.3 6.9-133	116±204 22-727	71.4±12.7 58.8–96.7	70.4±20.3 35.5–95.6	159±138 41-428	[20]
	Egypt	Suez Gulf (muddy sediment))	157.9 ± 205 27.1-719	295 ± 612 17-2.034	82.4 ± 40.1 71.8 -186.0	93.7 ± 32.0 42.6-151.3	133±60 64–277	[20]
	Morocco	Lagoon of Oualidia	Mean±SD Range	36.5 ± 22.9 20.0-90.0			54.6±16.6 30.6−88	228±16 203-260	[21]
	Morocco	Loukkos	0	32.4±7	53 ± 21	56.1±7.3		131±9	[22]
	Morocco	Sebou		24.0–42 51.5±12	37–89 117±26	49.0-66.0 101.0 ± 24.0		116−142 179±24	[22]
	Morocco	Bou Regreg		38.0-66 45.2±9 22.0 55	87-150 144±22	66.0-135.0 97.0±7.0		150-206 172±14	[22]
:	Morocco	Oum er Rbia		35.2±5	101 ± 53	80.0±6.0		173±38	[22]
Asia	Azerbaijan Iron	Caspian Sea	Mean±SD Range	31.9±9.6 14.5–57.6 34 7±11 0	832±124 543–971 815±100	50.1 ± 8.9 34.5-68.0 51.6+11.8	19.6±3.9 12.2–28.6 18.0+4.2	83±14 51-110 85+18	[23]
	Kazakhstan			0.1.1.1.0 13.2−50.9 6.4±8.3	470−1,110 196±107	29.4–67.8 10.4±10.8	10.0±±±± 11.3−24.6 5.8±3.3	56-146 56-146 11±11	[23]
	Russia			1.2–49.5 8.3±5.4 2.5–21.9	45-630 200±96 90-455	1.8-54.8 14.0 ± 7.6 5.4-34.2	1.4-14.6 4.2 ± 2.2 0.7-8.0	1-60 17±13 3-53	[23]
	Turkey	Marmara Sea	Mean±SD Range	20.0 ± 9.4 12.7-30.6	344±61 274–384	38.6 ± 16.8 20.5-53.9	25.4±5.7 21.6–31.9	43±8 34−51	[24]
	Israel	Mediterranean Coastal Region		2.0±1.3 1.2−8.6			5.8 ± 1.7 3.6-10.9	7.3±4.5 2.6–28.4	[25]
	China	Mingjiang Estuary	Mean±SD Range	27.9±7.4 16.4−37.2			43.8±11.9 34.4–69.6	100±13 92–128	[26]
	China	Xiamen-Jinmen	Mean±SD Range	16.61 ± 4.20409 11-24.5			52.63±14.4746 36-80.3	103.95±26.3579 77.5−136	[26]
	China	Pearl River estuary	Mean±SD	37.7±1.7		32.3 ± 0.9	50.9 ± 3.3	104 ± 7	[27]

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Continent	Country/region	Location		Cu	Mn	Ni	Pb	Zn	References
			Range	33.9-40.4		31.2–34.6	47.0-59.5	95–115	
	China	Western Bohai Bay	Mean±SD	28.1 ± 1.6		30.3 ± 2.4	$20.4 {\pm} 0.9$	83±3	[12]
			Range	22.5-36.5		26.4-41.6	18.3-23.4	67–97	
	China	Haihe River Estuary		28.52.8		32.71.6	25.20.1	84±13	[12]
				22.934.9		29.537.7	23.026.6	62-107	
	China	Port of Tianjin		38.121.0		34.50.8	22.92.8	108 ± 34	[12]
				20.078.9		30.345.4	20.030.7	71–156	
	China	Yongding River estuary		29.80.1		30.72.1	20.82.3	$91{\pm}4$	[12]
				17.8 33.3		27.039.1	18.624.8	67–101	
	China	Yangtze River intertidal	Mean±SD	30.7±9.7	766 ± 177	31.8 ± 7.2	27.3±5.6	94±24	[28]
			Range	6.9-49.7	413-1,112	17.6 - 48.0	18.3-44.1	48-154	
	Korea	Masan Bay	Mean±SD	43.4		28.8	44.0	206	[29]
			Range	13.5-90.7		10.2 - 40.4	13.0-82.2	80-379	
	India	Alang-Sosiya coast intertidal	Mean±SD	214.4 ± 53.5	$4,643\pm 1,770$	173 ± 48	170.0 ± 69.2	$1,222 \pm 147$	[30]
		Gulf of Mannar	Mean±SD		301 ± 169	23.7±11.4	15.7±3.5	74±27	[31]
			Range		70-818	11.0-63.0	7.0-21.0	41–184	
	Russia	Barents Sea surface sediments		13.0 ± 7.0	377.0 ± 156.0	25.0 ± 14.0	14.0 ± 4.0	47.0 ± 30.0	[32]
				4.0-25.0	154.0-684.0	6.0-47.0	9.0-22.0	7.0–97.0	
Australia	Australia	Bells Creek catchment	Range	0.5-5.8	0-229	1.5 - 6.4	3.0-7.3	18-68	[33]
Australia	Fiji	Suva Harbor	Mean±SD	54.0 ± 29.8			45.4±17.4	110 ± 48	[34]
			Range	21.4–143			22.1–93.5	40–269	
Europe	Montenegro	Adriatic Albanian coast	Mean±SD	103.3 ± 91.6	$1,940{\pm}2,364$	240 ± 65	$18.4{\pm}6.4$	107 ± 58	[35]
			Range	31.6–255	499–6,149	168-311	13.1–29.2	48-193	
	France	Cajarc Site, Lot River	Mean	97.7			523	4,430	[36]
			Range	40.6–264			111 - 1, 280	909-10,000	
	France	Marcenac Site, Lot River	Mean	269.0			105	134	[36]
			Range	17.9-42.0			27.9-68.0	82–206	
	France	Temple Site, Lot River	Mean	30.7			105	949	[36]
			Range	16.7 - 78.0			31.3-593	82-7,230	
	Greece	Candarli Gulf surficial	Mean±SD	24.4 ± 10.7	400 ± 256	42.0 ± 20.8	35.5±20.7	94±53	[37]
		sediments	Range	2.7-46.1	173-1,423	7.6-100.3	14.5–138	55-358	
	Greece	Keratsini Harbor, Saronikos	Mean±SD	288±75	$204{\pm}185$		648 ± 151	$1,435\pm 1,438$	[38]
		Gulf	Range	195-518	95 - 1, 101		521-1,263	409–6,725	
	Italy	Gulf of Manfredonia	Mean±SD	47.0 ± 30.0		47.0 ± 15.0	$17.0 {\pm} 6.0$	82±21	[39]
			Range	11.0-135		12.0-82.0	4.0 - 33.0	23–119	
	Italy	Naples City Port	Mean±SD	131 ± 84	389±94		123.0 ± 67.6	303 ± 274	[40]

Table 1 (continued)	ined)								
Continent	Country/region	Location		Cu	Mn	Ni	Pb	Zn	References
			Range	40-415	95–535		37.0–314	41 - 1, 196	
	Italy	Taranto Gulf	Mean±SD	47.4±2.7	893±537	53.3 ± 3.8	57.8 ± 8.4	102 ± 14	[41]
			Range	42.4–52.3	552-2,826	47.9–60.7	44.7–74.8	87–129	
	Spain	Estuary of Huelva and	Mean±SD	$407{\pm}603$		21.2 ± 14.9	191 ± 283	535 ± 694	[42]
		adjacent Atlantic shelf	Range	17-1,830		10.0-46.0	12.0-926	61-2,300	
	Spain	Gulf of Cadiz	Mean±SD	53.4 ± 19.8	293 ± 87	29.4 ± 12.3	45.0 ±21.5	118 ± 52	[43]
			Range	13.2–92.4	79-452	1.2-43.8	6.9 - 84.6	19–188	
	Spain	Odiel River	Mean±SD	604±751	$1,086\pm 1,212$	29.7±17.4	$2,369\pm1,925$	$2,874\pm 1,647$	[44]
			Range	52-2,843	54-5,482	15.2–95.1	512-5,778	1,264-8,286	
	UK	Tees Estuary		123.3 ± 72.7		40.2 ± 8.3	330.1 ± 230.1	407.4 ± 218.0	[45]
				25.0-262.0		21.0-49.0	37.0-680.0	65.0-777.0	
North America	USA	Baja California, California	Mean±SD	12.8 ± 3.9	$889{\pm}1,096$	25.3 ± 8.0	13.6 ± 3.8	68±32	[46]
			Range	4.9-22.7	392-5,282	15.5-44.4	6.0-21.3	39–188	
	USA	Hudson River	Mean±SD	77.8 ± 31.7			81.5 ± 33.5	182 ± 41	[7]
			Range	18.0–149			24.0–177	101 -	
	USA	California, Tidal salt marsh	Mean±SD	37.9 ± 26.6		53.4±21.9	39.1 ± 33.0	120 ± 87	[47]
			Range	15.1–78.3		20.3-75.1	6.8-87.9	67–273	
	NSA	Upper Gulf of California	Mean±SD			45.1 ± 44.0		44±27	[48]
	NSA	Colorado River Delta	Mean±SD			117 ± 117		39±9	[48]
	USA	Oyster Rock Landing salt	Mean±SD	29.6±4.7			$36.1{\pm}11.8$	66±18	[49]
		marsh in Delaware	Range	23.0–38.0			19.0-55.0	42–89	
	NSA	Wolfe Glade Delaware salt	Mean±SD	37.8 ± 36.8			130.4 ± 73.5	78±14	[49]
		marsh	Range	13.0–130			22.0-215	51 - 100	
	Canada	Vancouver Harbor	Mean	168.0		42.0	33.0	138	[50]
South America	Brazil	Jacarepagua Basin	Mean±SD	60.7 ± 25.3	$278.0 {\pm} 38.6$	55.7±28.3	55.7±13.1	$330.0{\pm}65.6$	[51]
			Range	32.0-80.0	244.0-320.0	32.0-87.0	41.0-66.0	270.0 - 400.0	
	Brazil	Lower Paraiba do Sul estuary	Mean±SD	63.0 ± 11.0	$895.0 {\pm} 57.0$			192.0 ± 15.0	[52]
			Range	54.0-87.0	850.0-1,020.0			166.0-212.0	
	French Guiana	Kaw mangroves	Mean±SD		529 ± 160	35.2 ± 4.7	26.9 ± 4.1	$174{\pm}28$	[53]
			Range		240–901	27.0-48.7	16.6-55.9	128–267	
	French Guiana	Guiana shoreface sediments	Mean±SD	24.8 ± 1.9	$1,040{\pm}40$	35.8 ± 1.8	30.0 ± 2.1	141 ± 15	[53]
			Range	22.9–26.7	1,001-1,089	34.0–37.6	26.9–33.2	123–154	
	French Guiana	Sinnamary mangroves	Mean±SD	17.8 ± 6.4	539 ± 509	31.7 ± 5.9	26.9 ± 4.1	164 ± 71	[53]
Earth's crust			Mean	25.0	600	20.0	20.0	71	[3]
ERL				34.0		20.9	46.7	150	[54]
ERM				270.0		51.6	218.0	410	[54]



Fig. 1 Selected sites of estuaries and coastal areas worldwide

percentages of enrichment occur to As (100 %), Cd (73 %), Ni (58 %), and Cr (55 %). The results show that metal EFs reflect the impact of external metal sources on the sediment quality. Metal enrichment among the selected coastal areas is shown in Fig. 2 and summarized below:

- As—Barents Sea surface sediments (Russia) and Candarli Gulf (Greece) show extremely high enrichment of As (EF>40), and Caspian Sea, Gulf of Cadiz (Spain), Bells Creek catchment (Australia), and California tidal salt marsh (USA) show significant enrichment of As (5<EF<20).
- Cd—Suez Gulf (Egypt), Alang–Sosiya coast intertidal zone (India), Oum er Rbia (Morocco), and Mediterranean Coastal Region (Israel) show extremely high enrichment of Cd (EF>40). Sebou (Morocco), Bou Regreg (Morocco), and Loukkos (Morocco) show very high enrichment of Cd (20<EF<40). Marmara Sea (Turkey), Hudson River (USA), Gulf of Cadiz (Spain), Masan Bay (Korea), Gulf of Mannar (India), and California tidal salt marsh (USA) show significant enrichment of Cd (5<EF<20). Caspian Sea (Russia) and Yangtze River intertidal zone (China) show moderate enrichment (2<EF<5) of Cd.
- Cr—Gulf of Mannar (India), Suez Gulf (Egypt), Lower Paraiba do Sul estuary (Brazil), and Adriatic Albanian coast (Montenegro) show significant enrichment (5<EF <20) of Cr. Caspian Sea, Marmara Sea (Turkey), Gulf of Cadiz (Spain), Taranto Gulf (Italy), Yangtze River intertidal zone (China), Barents Sea (Russia), California tidal salt marsh (USA), and Alang–Sosiya coast intertidal zone (India) show moderate enrichment (2<EF<5) of Cr.

very high enrichment of Cu (20<EF<40). Lower Paraiba do Sul estuary (Brazil) shows significant enrichment (5< EF<20) of Cu. Oum er Rbia (Morocco), Adriatic Albanian coast (Montenegro), Hudson River (USA), Gulf of Cadiz (Spain), Marmara Sea (Turkey), Sebou (Morocco), Suva Harbor (Fiji), Loukkos (Morocco), Alang–Sosiya coast intertidal zone (India), Caspian Sea (Russia), and Taranto Gulf (Italy) show moderate enrichment (2<EF<5) of Cu.
5. Mn—Lower Paraiba do Sul estuary (Brazil) shows significant enrichment (5<EF<20) of Mn (Fig. 2). Suez Gulf

4. Cu-Suez Gulf (Egypt) and Odiel River (Spain) show

- icant enrichment (5<EF<20) of Mn (Fig. 2). Suez Gulf (Egypt), Adriatic Albanian coast (Montenegro), and Caspian Sea (Russia) show moderate enrichment (2<EF <5) of Mn.
- Ni—Suez Gulf sandy sediment (Egypt) shows very high enrichment (20<EF<40) of Ni. Suez Gulf muddy sediment of (Egypt), Colorado River Delta (USA), Oum er Rbia (Morocco), Adriatic Albanian coast (Montenegro), Marmara Sea (Turkey), Sebou (Morocco), and Bou Regreg (Morocco) show significant enrichment (5<EF< 20) of Ni (Fig. 2). Loukkos (Morocco), Upper Gulf of California (USA), Caspian Sea (Russia), Gulf of Mannar (India), Taranto Gulf (Italy), tidal salt marsh in California (USA), Candarli Gulf surficial sediments (Greece), Caspian Sea (Kazakhstan), Caspian Sea (Iran), Caspian Sea (Azerbaijan), and Alang–Sosiya coast intertidal (India) show moderate enrichment (2<EF<5) (Fig. 2).
- Pb—Odiel River (Spain) and sandy sediment of Suez Gulf (Egypt) show extremely high enrichment (EF>40) of Pb. Muddy sediment of Suez Gulf (Egypt) shows very

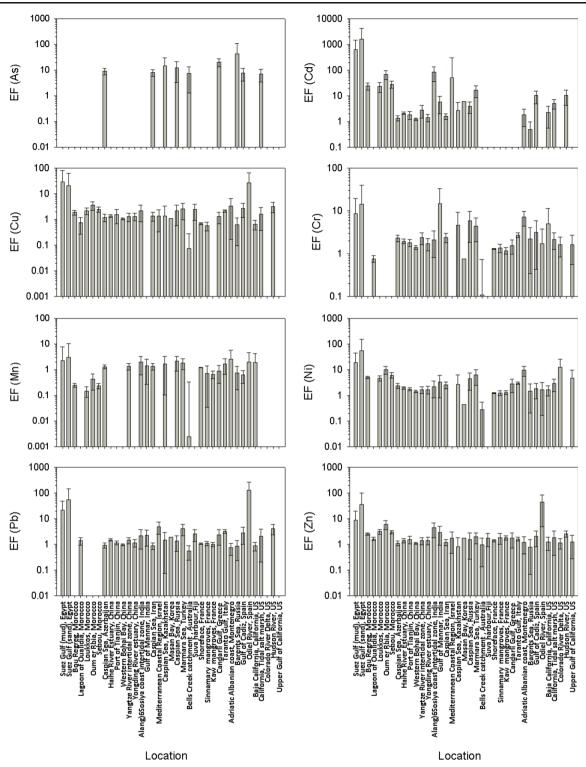


Fig. 2 Wide variations of metal (Cd, Cr, Cu, Ni, Pb, and Zn) enrichment factors (EF) reflect different sources and enrichment levels of metals in the sediments from different regions. High enrichment (EF>2) suggests various degrees of metal pollution

high enrichment (20<EF<40) of Pb. Mediterranean Coastal Region (Israel), Hudson River (USA), Marmara Sea (Turkey), Taranto Gulf (Italy), Gulf of Cadiz (Spain), Suva Harbor (Fiji), Candarli Gulf surficial sediments (Greece), Gulf of Mannar (India), Alang-Sosiya coast intertidal (India), and California tidal salt marsh (USA) show moderate enrichment (2<EF<5) of Pb.

Zn—Odiel River (Spain) shows extremely high enrichment (EF>40) of Zn. Suez Gulf sandy sediment (Egypt) shows very high enrichment (20<EF<40) of Zn. Lower

 Table 2
 Percentage of coastal areas that have different degrees fo potential contamination caused by As, Cd, Cr, Cu, Mn, Ni, Pb and/Zn

Elements	Total cases	Percentage	of coastal areas wit	h EF values of			Total
	(n)	<2	2~5	5~20	20~40	>40	
As	9	0.0	0.0	77.8	11.1	11.1	100
Cd	26	26.9	23.1	19.2	11.5	19.2	100
Cr	31	45.2	32.3	22.6	0.0	0.0	100
Cu	34	55.9	32.4	2.9	8.8	0.0	100
Mn	26	80.8	15.4	3.9	0.0	0.0	100
Ni	33	42.4	30.3	18.2	6.1	3.0	100
Pb	31	58.1	32.3	0.0	3.2	6.5	100
Zn	38	68.4	18.4	7.9	2.6	2.6	100

Five categories of metal enrichment are defined based on metal enrichment factor (EF): 1) deficiency to minimal enrichment (EF<2), 2) moderate enrichment (EF=2–5), 3) significant enrichment (EF=5–20), 4) very high enrichment (EF=20–40), and 5) extremely high enrichment (EF>40)

Paraiba do Sul estuary (Brazil), Suez Gulf muddy sediment (Egypt), and Oum er Rbia (Morocco) show significant enrichment (5<EF<20) of Zn. Alang-Sosiya coast intertidal (India), Loukkos (Morocco), Sebou (Morocco), Gulf of Mannar (India), Hudson River (USA), Bou Regreg (Morocco), and Gulf of Cadiz (Spain) show moderate enrichment (2<EF<5) of Zn (Fig. 2).

Metal Pollution and Ecological Impact

Ecotoxicity and biogeochemical circulation of metals in an ecosystem are determined by the metal concentration that organisms are exposed to. When the metal concentration in sediment exceeds certain threshold level, adverse biological effects frequently occur. At molecular level, excessive metal accumulation in the organism might stimulate biological counter stress processes such as induction of antioxidant enzymes, physiological impairment, and extra energy consumption [75]. As a result, organism mortality rises continuously when sediment metal concentrations increase [54].

To avoid adverse ecological effects from sediments, several approaches have been developed to assess the metal hazard potential to organisms as guidance for coastal ecosystem protection and restoration [54, 76, 77]. Generally, the potential ecotoxicological risk of metals on organisms in the sediments can be evaluated using the sediment guidelines (e.g., [54, 76, 78–80]). In order to better predict the toxicity of contaminants, Long et al. [54] defined the effect range-low (ERL) and effect range-median (ERM) system based on the compilation of matching biological and chemical data from numerous modeling, laboratory, and field measurement in marine and estuarine sediments. According to Long et al. [54], ERL is defined based on the concentration when 10th percentiles of organisms are influenced by the toxicity of a specific contaminant,

while ERM value is decided when 50th percentiles of organisms are influenced by the toxicity of the contaminant in that concentration. Three effect ranges of contaminant level could be set up based on ERL and ERM. When the contaminant concentration is below ERL, a very rare biological adverse effect is anticipated, and when the concentration is between ERL and ERM, an occasional biological adverse effect is expected. However, when the concentration is greater than ERM value, a frequent biological adverse effect (>50 % chances) could occur [54, 76, 78]. To evaluate the potential biological adverse effect, hazard quotient (HQ) is one kind of singlevalue estimate and is also the simplest method to estimate toxicity potential of the selected pollutants in the sediments [77, 81]. According to Urban and Cook [77], the HQ is defined as

$$HQ = \frac{SCC}{SQG}$$
(2)

where SCC is the sediment chemical concentration and SQG stands for the concentration defined by the sediment quality guideline. Both SCC and SQG are in milligram per kilogram. Since ERL is more reasonably predictive of non-toxic condition (SQGs), the SQG is set at ERL levels: Cd=1.2, Cr=81, Cu=34, Ni=20.9, Pb=46.7, and Zn=150 [54]. If HQ<0.1, no adverse effects are expected; if 0.1 < HQ < 1, low potential hazards are expected; in the range of 1.0 < HQ < 10, some adverse effects or moderate hazards are probable; and when HQ>10, high hazard potential is anticipated [81, 82].

As showed in Fig. 3, HQ values calculated from the average metal concentrations in the selected coastal sediments range from 0.36 to 22 for As (n=13), 0.03 to 832 for Cd (n=36), 0.01 to 6 for Cr (n=38), 0.01 to 18 for Cu (n=49), 0.07 to 12 for Hg (n=12), 0.01 to 12 for Ni (n=36), 0.06 to 51 for Pb (n=49), and 0.05 to 30 for Zn (n=53). The percentages of the coastal areas facing various degree of hazard potential from no adverse effects (HQ<0.1) to high hazard potential

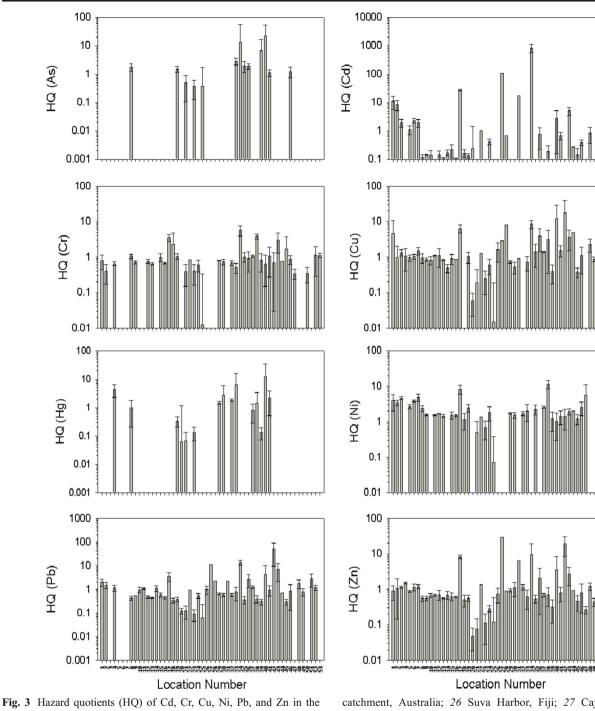


Fig. 3 Hazard quotients (HQ) of Cd, Ct, Cu, NI, Po, and Zh in the sediments from the selected coastal areas. Moderate (1 < HQ < 10) to high (HQ>10) hazard potential that can cause adverse biological effect are anticipated in some coastal areas. Location numbers: *1* Suez Gulf (mud), Egypt; *2* Suez Gulf (sand), Egypt; *3* Bou Regreg, Morocco; *4* Lagoon of Oualidia, Morocco; *5* Loukkos, Morocco; *6* Oum er Rbia, Morocco; *7* Sebou, Morocco; *8* Caspian Sea, Azerbaijan; *9* Haihe River Estuary, China; *10* Mingjiang Estuary, China; *11* Pearl River estuary, China; *12* Port of Tianjin, China; *13* Western Bohai Bay, China; *14* Xiamen-Jinmen, China; *15* Yangtze River intertidal zone, China; *16* Yongding River estuary, China; *17* Alang-Sosiya coast intertidal zone, India; *18* Gulf of Mannar, India; *19* Caspian Sea, Iran; *20* Mediterranean Coastal Region, Israel; *21* Caspian Sea, Kazakhstan; *22* Masan Bay, Korea; *23* Caspian Sea, Russia; *24* Marmara Sea, Turkey; *25* Bells Creek

catchment, Australia; 26 Suva Harbor, Fiji; 27 Cajarc Site, Lot River, France; 28 Marcenac Site, Lot River, France; 29 Shoreface, France; 30 Sinnamary mangroves, France; 31 Temple Site, Lot River, France; 32 Kaw mangroves, France; 33 Candarli Gulf, Greece; 34 Keratsini Harbor, Saronikos Gulf, Greece; 35 Gulf of Manfredonia, Italy; 36 Naples City Port, Italy; 37 Taranto Gulf, Italy; 38 Adriatic Albanian coast, Montenegro; 39 Barents Sea, Russia; 40 Estuary of Huelva and adjacent Atlantic shelf, Spain; 41 Gulf of Cadiz, Spain; 42 Odiel River, Spain; 43 Tees Estuary, UK; 44 Vancouver Harbor, Canada; 45 Baja California, California, USA; 46 California, Tidal salt marsh, US; 47 Colorado River Delta, US; 48 Hudson River, USA; 49 Oyster Rock Landing salt marsh in Delaware, USA; 50 Upper Gulf of California, US; 51 Wolfe Glade Delaware salt marsh, USA; 52 Jacarepagua Basin, Brazil; 53 Lower Paraiba do Sul estuary, Brazil

(HQ>10) are shown in Table 3. In most of the areas, only low hazard potentials (0.1 <HQ<1) are expected. However, As (15 %) and Cd (14 %) show relative high percentage in hazard potentials (HQ>10) (Table 3). The potential adverse biological effects caused by each individual metal are summarized below:

- As—High hazard potential (HQ>10) caused by As is anticipated in Estuary of Huelva and adjacent Atlantic shelf (Spain) and Keratsini Harbor, Saronikos Gulf (Greece); moderate hazard potential (1<HQ<10) in Barents Sea (Russia), Candarli Gulf (Greece), Gulf of Manfredonia (Italy), Naples City Port (Italy), Caspian Sea (Iran and Kazakhstan), California tidal salt marsh (USA), and Gulf of Cadiz (Spain); and low hazard potential (0.1<HQ<1) in Caspian Sea (Kazakhstan and Russia) and Bells Creek catchment (Australia) (Fig. 3).
- Cd—High hazard potential (HQ>10) caused by Cd is 2. anticipated in Keratsini Harbor, Saronikos Gulf (Greece), Cajarc Site, Lot River (France), Alang-Sosiya coast intertidal zone (India), Temple Site, Lot River (France), and Suez Gulf muddy sediment (Egypt); moderate hazard potential (1<HQ<10) in Suez Gulf (Egypt), Tees Estuary (UK), Estuary of Huelva and adjacent Atlantic shelf (Spain), Oum er Rbia (Morocco), Sebou (Morocco), Bou Regreg (Morocco), Loukkos (Morocco), and Masan Bay (Korea); and low hazard potential (0.1<HQ<1) in Hudson River (USA), Naples City Port (Italy), Marcenac Site, Lot River (France), Gulf of Cadiz (Spain), Marmara Sea (Turkey), California tidal salt marsh (USA), Vancouver Harbor (Canada), Mediterranean Coastal Region (Israel), Yangtze River intertidal zone (China), Adriatic Albanian coast (Montenegro), Xiamen-Jinmen (China), Gulf of Mannar (India), Port of Tianjin (China), Haihe River Estuary (China), Baja California (USA), Mingjiang Estuary (China), Caspian Sea (Iran and Azerbaijan), Western Bohai Bay (China), and Yongding River estuary (China).

- Cr—Moderate hazard potential (1<HO<10) caused by 3. Cr is anticipated in Keratsini Harbor (Greece), Adriatic Albanian coast (Montenegro), Alang-Sosiya coast intertidal (India), Tees Estuary (UK), Gulf of Mannar (India), Baja California (USA), Jacarepagua Basin (Brazil), Lower Paraiba do Sul estuary (Brazil), Gulf of Cadiz (Spain), Taranto Gulf (Italy), Caspian Sea (Azerbaijan), and Caspian Sea (Iran); low hazard potential (0.1<HQ<1) in Gulf of Manfredonia (Italy), Yangtze River intertidal (China), Naples City Port (Italy), California tidal salt marsh (USA), Masan Bay (Korea), Barents Sea (Russia), Guiana shoreface sediments (France), Suez Gulf muddy sediment (Egypt), Port of Tianjin (China), Vancouver Harbor (Canada), Sinnamary mangroves (France), Haihe River Estuary (China), Kaw mangroves (France), Odiel River (Spain), Lagoon of Oualidia (Morocco), Western Bohai Bay (China), Estuary of Huelva and adjacent Atlantic shore (Spain), Marmara Sea (Turkey), Candarli Gulf (Greece), Suez Gulf sandy sediment (Egypt), Caspian Sea (Russia), Caspian Sea (Kazakhstan), Upper Gulf of California (USA), and Colorado River Delta (USA).
- 4. Cu—High hazard potential (HQ>10) caused by Cu is anticipated in Odiel River (Spain) and Estuary of Huelva (Spain); moderate hazard potential (1<HQ<10) in Keratsini Harbor (Greece), Marcenac Site (France), Alang-Sosiya coast intertidal (India), Vancouver Harbor (Canada), Suez Gulf muddy sediment (Egypt), Naples City Port (Italy), Tees Estuary (UK), Adriatic Albanian coast (Montenegro), Cajarc Site in Lot River (France), Hudson River (USA), Lower Paraiba do Sul estuary (Brazil), Jacarepagua Basin (Brazil), Suva Harbor (Fiji), Gulf of Cadiz (Spain), Sebou (Morocco), Taranto Gulf (Italy), Gulf of Manfredonia (Italy), Bou Regreg (Morocco), Masan Bay (Korea), Port of Tianjin (China), California tidal salt marsh (USA), Wolfe Glade Delaware salt marsh (USA), Pear River estuary (China), Lagoon of Oualidia

Elements	Total cases	Percenta	ge of coastal areas	with HQ values of		
	<i>(n)</i>	<0.1	0.1 <hq<1< th=""><th>1.0<hq<10< th=""><th>HQ>10</th><th>Total</th></hq<10<></th></hq<1<>	1.0 <hq<10< th=""><th>HQ>10</th><th>Total</th></hq<10<>	HQ>10	Total
As	13	0.0	23.1	61.5	15.4	100
Cd	36	8.3	55.6	22.2	13.9	100
Cr	37	2.7	64.9	32.4	0.0	100
Cu	49	4.1	38.8	53.1	4.1	100
Hg	12	8.3	25.0	58.3	8.3	100
Ni	39	2.6	5.1	89.7	2.6	100
Pb	46	4.4	56.5	32.6	6.5	100
Zn	53	3.8	60.4	32.1	3.8	100

The hazardous quotient value (HQ) suggests no adverse effects (HQ<0.1), low adverse effects (0.1<HQ<1), moderate adverse effects (1.0<HQ<10), or high adverse effects (HQ>10)

Table 3Percentage of coastalareas that have potential adverseeffects caused by As, Cd, Cr, Cu,Hg, Ni, Pb and/Zn

(Morocco). Oum er Rbia (Morocco). and Caspian Sea (Iran); and low hazard potential (0.1<HQ<1) in Suez Gulf sandy sediment (Egypt), Loukkos (Morocco), Caspian Sea (Azerbaijan), Yangtze River intertidal (China), Lot River Temple site (France), Oyster Rock Landing salt marsh in Delaware (USA), Haihe River Estuary (China), Western Bohai Bay (China), Mingjiang Estuary (China), Guiana shoreface sediments (France), Candarli Gulf surficial sediments (Greece), Marmara Sea (Turkey), Sinnamary mangroves (France), Xiamen-Jinmen (China), Barents Sea (Russia), Baja California (USA), Caspian Sea (Russia), and Caspian Sea (Kazakhstan). Hg-High hazard potential (HQ>10) caused by Hg is anticipated in Estuary of Huelva and adjacent Atlantic shelf (Spain); moderate hazard potential (1<HO<10) in Candarli Gulf (Greece), Lagoon of Oualidia (Morocco), Sinnamary mangroves (France), Gulf of Cadiz (Spain), Kaw mangroves (France), Guiana shoreface sediments (France), and Adriatic Albanian coast (Montenegro); low hazard potential (0.1<HQ<1) in Caspian Sea and Barents Sea (Russia) (Fig. 3).

- 5. Ni—High hazard potential (HQ>10) caused by Ni is anticipated in Adriatic Albanian coast (Montenegro); moderate hazard potential (1<HQ<10) in Alang-Sosiya coast intertidal (India), Colorado River Delta (USA), Sebou (Morocco), Bou Regreg (Morocco), Suez Gulf muddy sediment (Egypt), Oum er Rbia (Morocco), Suez Gulf sandy sediment (Egypt), Loukkos (Morocco), Jacarepagua Basin (Brazil), California tidal salt marsh (USA), Taranto Gulf (Italy), Caspian Sea (Iran), Caspian Sea (Azerbaijan), Gulf of Manfredonia (Italy), Upper Gulf of California (USA), Candarli gulf surficial sediments (Greece), Vancouver Harbor (Canada), Tees Estuary (UK), Marmara Sea (Turkey), Guiana shoreface sediments (France), Kaw mangroves (France), Port of Tianjin (China), Haihe River Estuary (China), Pearl River estuary (China), Yangtze River intertidal (China), Sinnamary mangroves (France), Western Bohai Bay (China), Odiel River (Spain), Gulf of Cadiz (Spain), Masan Bay (Korea), Baja California (USA), Barents Sea surface sediments (Russia), Gulf of Mannar (India), and Estuary of Huelva (Spain); low hazard potential (0.1<HQ<1) in Caspian Sea (Russia), Caspian Sea (Kazakhstan), and Estuary of Huelva and adjacent (Spain).
- 6. Pb—High hazard potential (HQ>10) caused by Pb is anticipated in Odiel River (Spain), Keratsini Harbor (Greece), and Lot River Cajarc site (France); moderate hazard potential (1<HQ<10) in Tees Estuary (UK), Estuary of Huelva (Spain), Alang-Sosiya coast intertidal (India), Wolfe Glade Delaware salt marsh (USA), Naples City Port (Italy), Lot River Marcenac site (France), Lot River Temple site (France), Suez Gulf muddy sediment (Egypt), Hudson River (USA), Suez Gulf sandy sediment</p>

(Egypt), Taranto Gulf (Italy), Jacarepagua Basin (Brazil), Lagoon of Oualidia (Morocco), Xiamen-Jinmen (China), and Pearl River estuary (China); and low hazard potential (0.1<HO<1) in Suva Harbor (Fiji), Gulf of Cadiz (Spain), Masan Bay (Korea), Mingjiang Estuary (China), California tidal salt marsh (USA), Oyster Rock Landing salt marsh (USA), Candarli Gulf surficial sediments (Greece), Vancouver Harbor (Canada), Guiana shoreface sediments (France), Yangtze River intertidal (China), Kaw mangroves (France), Sinnamary mangroves (France), Marmara Sea (Turkey), Haihe River Estuary (China), Port of Tianjing (China), Western Bohai Bay (China), Caspian Sea (Iran), Gulf of Manfredonia (Italy), Gulf of Mannar (India), Barents Sea (Russia), Baja California (USA), Mediterranean Coastal Region (Israel), and Caspian Sea (Kazakhstan).

Zn-High hazard potential (HQ>10) caused by Zn is 7. anticipated in Lot River Cajarc Site (France) and Odiel River (Spain); moderate hazard potential (1<HQ<10) in Keratsini Harbor (Greece), Alang-Sosiya coast intertidal (India), Lot River Temple site (France), Estuary of Huelva (Spain), Tees Estuary (UK), Jacarepagua Basin (Brazil), Naples City Port (Italy), Lagoon of Oualidia (Morocco), Masan Bay (Korea), Lower Paraiba do Sul estuary (Brazil), Hudson River (USA), Sebou (Morocco), Kaw mangroves (France), Oum er Rbia (Morocco), Bou Regreg (Morocco), Sinnamary mangroves (France), and Suez Gulf sandy sediment (Egypt); and low hazard potential (0.1<HQ<1) in Guiana shoreface sediments (France), Vancouver Harbor (Canada), Lot River Marcenac site (France), Suez Gulf muddy sediment (Egypt), Loukkos (Morocco), California tidal salt marsh (USA), Gulf of Cadiz (Spain), Suva Harbor (Fiji), Port of Tianjin (China), Adriatic Albanian coast (Montenegro), Pearl River estuary (China), Xiamen-Jinmen (China), Taranto Gulf (Italy), Mingjiang Estuary (China), Yangtze River intertidal (China), Candarli Gulf surficial sediments (Greece), Caspian Sea (Iran), Haihe River Estuary (China), Western Hohai Bay (China), Caspian Sea (Azerbaijan), Gulf of Manfredonia (Italy), Wolfe Glade Delaware salt marsh (USA), Gulf of Mannar (India), Baja California (USA), Oyster Rock Landing salt marsh (USA), Barents Sea (Russia), Upper Gulf of California (USA), Marmara Sea (Turkey), Colorado River Delta (USA), Bells Creek Catchment (Australia), and Caspian Sea (Russia).

Discussion

Besides anthropogenic sources, metal accumulation and distribution in the estuary sediments are also influenced by the interaction between metals and sediments. Therefore, factors such as sediment grain size, sediment surface dynamic equilibrium, and exposure time can also influence the spatial accumulation of metals in marine sediments [7, 12, 46]. Among all the 52 sites summarized in this review, seven major coastal areas adjacent to highly industrialized and urbanized regions were chosen to further investigate the major factors determining metal accumulation and spatial distribution in costal sediments. These seven coastal areas including New York Harbor-Hudson River Estuary [7], Egypt Suez Gulf [20], China Tianjin Port–Bohai Bay [12], India Gulf of Cambay [30], Greece Saronikos Gulf [38], and Mexico Baja California and USA California Gulf [46] were selected because detailed information on the source and cause of metal distribution in sediments were provided by the authors. The major sources of metal contamination include upriver input (PCBs in Hudson River estuary), urban wastewater runoff (New York Harbor, Suez Gulf, Bohai Bay, Saronikos Gulf, and Baja California), port transportation (Tianjin Port and Saronikos Gulf), and industrial wastes (Suez Gulf, Gulf of Cambay, and Saronikos Gulf).

The enrichment level and spatial distribution of metals along the coastal area in the selected sites varied with contamination sources and the time of exposure. In the New York Harbor-Hudson River estuary coastal area, the distribution of metal in sediments was mainly influenced by upriver source, urban source, and sediment particle grain size. Silver was identified as tracer of urban source contamination based on correlation analysis [7] (Fig. 1). The sandy and muddy sediments in Gulf of Suez were analyzed separately, and metal concentrations were different between the two because their capacities to adsorb metals from water were different. In addition, metals in the sediments showed three different accumulation clusters (Cr, Cu, Fe, and Mn; Cd and Pb; and Co), possibly because the metals were from various contamination sources such as offshore oil fields, industrial wastes, and ballast water [30] (Fig. 1). In Tianjin Port-Bohai Bay coastal area, high metal enrichment was mostly observed in sediments collected in Tianjin Port. ²¹⁰Pb analysis indicated that metal concentrations were increasing in the recent years, suggesting that the contaminants were continuously released into the coastal area while the further transportation of contaminants into Bohai Bay was limited [12] (Fig. 1). Among all the coastal sites investigated, the Alang-Sosiya yard in the Gulf of Cambay has the highest metal contamination level. In addition, metal concentrations in the sediment close to the ship scrapping workshop were much higher than that in the sediment collected in the gulf intertidal zone, which was only 0.5 km from the ship scrapping workshop. It can be indicated that the ship scrapping industry and the domestic waste charge along it is possibly the main source of metal in the sediment. Moreover, metal concentrations in the bulk sediment lower than that in the fine sediment confirmed that metals were more bounded to the fine fraction of sediment due to its larger surface area [30] (Fig. 1). Greece Saronikos Gulf is another port that is heavily influenced by port transportation. However, compared to Tianjin Port, the level of metal enrichment was relatively high. This is possibly due to the discharge of sewage outfall into the gulf from the adjacent highly industrialized areas [38] (Fig. 1). Finally, the spatial variation of metal accumulated in the USA–Mexico (Baja California–California) estuary varied with the anthropogenic activities along the seashore. Among the 19 sampling sites that ranged from Pt. Loma Wastewater Treatment Plant in San Diego (USA) to Punta Bandera Treatment Plant in Tijuana (Mexico), sites close to Punta Bandera had higher concentration of Cu, Zn, Ni, and Cr. At the same time, Cd, Ag, and Pb showed different accumulation pattern, indicating a different metal discharge source other than Punta Bandera Treatment plant [46] (Fig. 1).

Finally, the dynamic biogeochemical circulation and accumulation of metals in sediments are also affected by the ecotoxicity of metals in sediments. The dynamic physiochemical ecological processes that mediated the transportation of substances within the ecosystem can redistribute the accumulation of metals in coastal sediments by concentrating, permanently depositing, and transporting metals to various mediums. For example, phytoplankton bloom can concentrate metal in sea water into biomass and increase bioaccumulation of metals in clams, eventually introducing significant amount of metals into food chain [75].

Future Perspectives

In summary, based on the analysis of the summarized information collected in this study, coastal sediment metal contamination should continue to raise our concern. It is not new that metal contaminations are still present in the world's estuaries and coastal areas. The current issues are how we can effectively exercise the contamination assessment, environmental protection, ecosystem restoration, and sustainable development of the coastal areas. In the future, more attention should be paid to develop more precise contamination evaluation and ecological risk assessment approaches as well as more sustainable remediation strategies for contaminated coastal sites.

Although the choice of geochemical normalization element is critical when evaluating the enrichment level of metals based on EF, a more accurate assessment should be achieved by choosing the local background values from adjacent sites with less anthropogenic disturbance [83]. It should also be noted that the HQ based on the total sediment concentration is only a conservative evaluation of the potential ecological risk of metal accumulated in the sediment because only the bioavailable fraction of metals can pose potential risk to the ecosystem [84]. Therefore, the actual ecological risk could be different from what is indicated by the HQ in this study as total metal concentration applied for the HQ calculation includes both bioavailable metal concentration and non-bioavailable metal concentration [84]. To properly estimate the actual ecological risk of metals accumulated in sediment, it is more important to determine the concentration of bioavailable metal [83].

Besides determining the fraction of bioavailable metals in total sediment metal concentration for the sediment risk assessment, metal speciation in sediment pore water is also highly concerned because not all the dissolved metals in the pore water are available for organisms to absorb. Dissolved metals in solution usually exist as free ions or associated with complexes (e.g., proton complexes, ligands, chelates) and organisms tend to uptake metals in free ion or small-molecular metal complex forms [85]. Furthermore, metal toxicity also varies with metal speciation. For example, As(III) is more toxic than As(V), Cr(VI) is more toxic than Cr(III), and methylated metals (e.g., methylated mercury) have higher toxicity [84, 86]. Therefore, special attention should be given to the development of analytical approaches that can measure the concentration and speciation of dissolved metal in pore water effectively and accurately [87].

Finally, as sediment metal contamination is becoming a worldwide environmental issue, remediation strategies are needed to eliminate the potential environmental impacts on human health [88]. Conventional remediation techniques for contaminated sediment such as in situ capping, landfill disposal, and sea dumping were once very popular for coastal sediment remediation. However, these techniques are not long-term sustainable as contaminants are potentially mobile after the treatment. In the recent decades, new remediation techniques such as biological treatment, thermal treatment, and in situ chemical treatment are under development, which significantly increased the efficiency and reliability of sediment remediation [89]. The selection of an appropriate technique for a specific remediation project usually depends on human and ecological risk before and after the remediation. In recent years, it is proposed to apply life cycle assessment to assist the selection of ideal sediment remediation technique in order to take environmental footprint into consideration [90].

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