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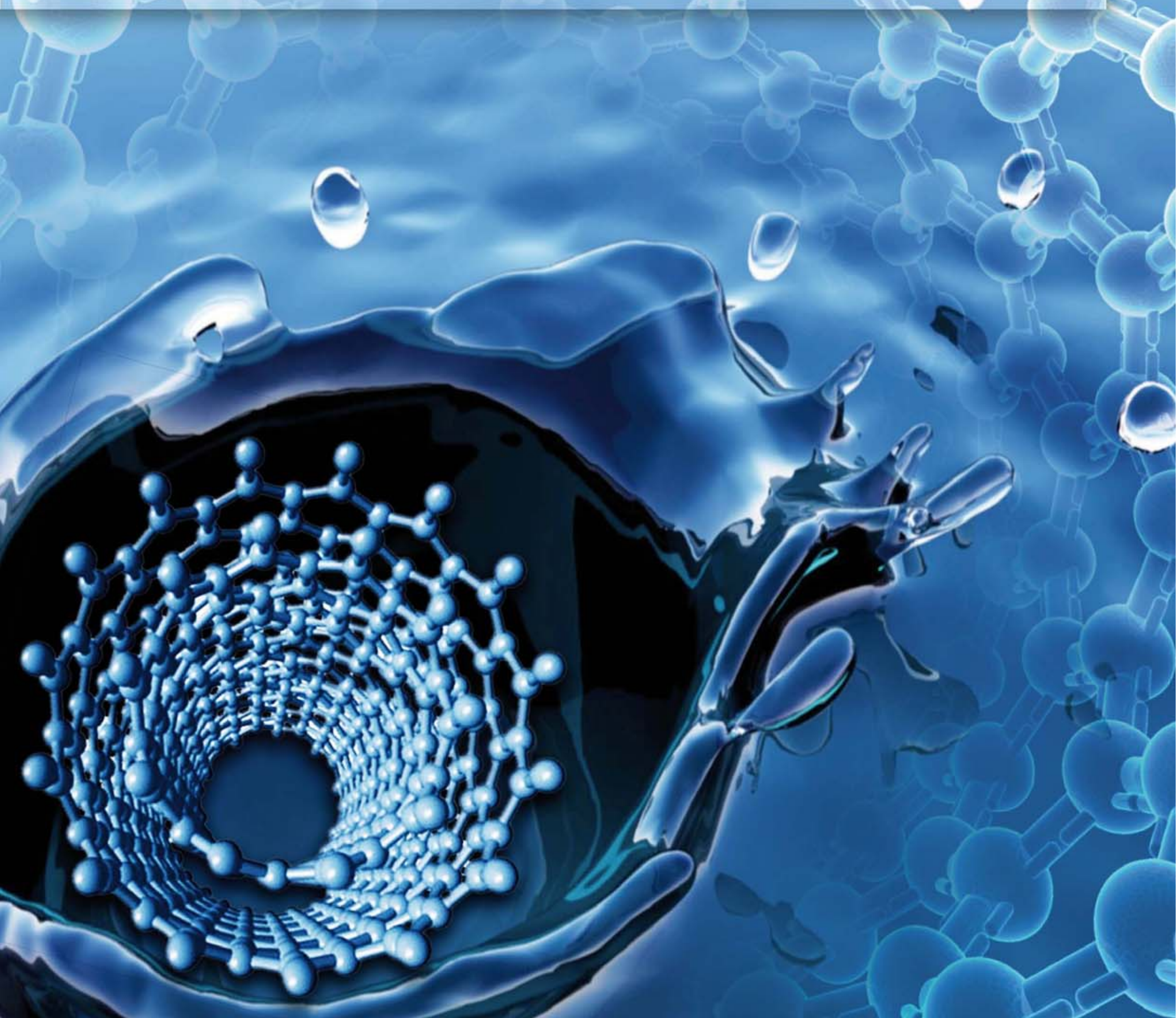

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Distribution and contamination status of chromium in surface sediments of northern Kaohsiung Harbor, Taiwan

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Abstract

The distribution, enrichment, accumulation, and potential ecological risk of chromium (Cr) in the surface sediments of northern Kaohsiung Harbor, Taiwan, China were investigated. Sediment samples from ten locations located between the river mouths and harbor entrance of northern Kaohsiung Harbor were collected quarterly in 2011 and characterized for Cr, aluminum, water content, organic matter, total nitrogen, total phosphorous, total grease, and grain size. Results showed that the Cr concentrations varied from 27.0 to 361.9 mg/kg with an average of (113.5 ± 87.0) mg/kg. High Cr concentration was observed near the Jen-Gen River mouth. The mean Cr concentration was high at 255.5 mg/kg, which was at least 2 to 7 times than that of other sites. This might imply significant Cr contribution from upstream receiving tanneries wastewater into the Jen-Gen River. The spatial distribution of Cr reveals relatively high in the river mouth region, especially in Jen-Gen River, and gradually diminishes toward the harbor entrance region. This indicates that the major sources of Cr pollution from upstream industrial and municipal wastewaters discharged along the river bank; and Cr may drift with sea current and be dispersed into open sea. Moreover, Cr concentrations correlated closely to the physical-chemical properties of the sediments, which suggested the influence of industrial and municipal wastewaters discharged from the neighboring industrial parks and river basins. Results from the enrichment factor and geo-accumulation index analyses imply that the Jen-Gen River sediments can be characterized as moderate enrichment and none to medium accumulation of Cr, respectively. However, results of potential ecological risk index indicate that the sediment has low ecological potential risk. The results can provide valuable information to developing future strategies for the management of river mouth and harbor.

Key words: accumulation; chromium; ecological risk; enrichment; sediment

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Introduction

The anthropogenic activities may cause severe environmental pollution problems of metals, such as chromium (Cr), lead (Pb), zinc (Zn), and copper (Cu) (Fukue et al., 2007). These metals being transported from the terrestrial to the marine environment, and the estuary environmental is the last place (Scott and Wright, 1988). Hence, the river mouth region, harbor, and seashore with dense population and industries usually become heavily polluted by toxic metals, including Cr (Pertsemli and Voutsa, 2007). Cr is moderately toxic to aquatic organisms; its presence threatens the water ecological environment. Therefore, much research effort has been directed toward the distribution of Cr in water environment. Anthropogenic activities including tanning, mining, smelting, domestic and industrial wastewaters, steam electrical production, and sewage sludge are the major source of Cr pollution (Callender, 2003; Pertsemli and Voutsa, 2007). In receiving water body, Cr is presented essentially as hydroxy-complexes of low solubility associated with the water-borne suspended particles (Kotaś and Stasicka, 2000; Pawlikowski et al., 2006). After a series of natural processes, the water-borne Cr finally accumulates in the sediment, and the quantity of Cr contained in the sediment reflects the degree of pollution for the water body (Selvaraj et al., 2004).

Kaohsiung Harbor is located on the southwestern shore, and it is the largest international harbor in Taiwan, China. However, it receives effluents from four contaminated rivers, including Love River, Canon River, Jen-Gen River, and Salt River. Study has shown that the Kaohsiung Harbor is heavily polluted with Cr, and the Love River, Canon River, and Jen-Gen River are the major pollution sources (Chen et al., 2007). These three rivers flow through the downtown area of Kaohsiung City and finally discharged into Kaohsiung Harbor (**Fig. 1**). Kaohsiung City is the largest industrial city in Taiwan with 2.9 million residents. During earlier years, the lack of sanitary sewer

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system forces discharge of un-treated raw wastewater directly to adjacent water bodies, which leads to serious deterioration of river water quality. Although in recent years, Kaohsiung City actively promotes the construction of wastewater collection and treatment systems, in 2010, the wastewater system only serves 60% of the city (http://web.trying.com.tw/sewer/index.htm). Additionally, Kaohsiung City also actively involves in public projects on river restoration (e.g., Love River, and Jen-Gen River) by constructing river intercepting stations near the middle section of the river to divert the upstream polluted river water to a wastewater treatment facilities for alleviating the downstream pollution problems. However, during the wet season, the river water intercepting gate is opened for by-passing the sudden surge of river flow brought over by storms that will discharge the upstream pollutants to downstream sections. Love River, Canon River, and Jen-Gen River are located in Kaohsiung City's northern basin area of about 60% of the entire Kaohsiung City, and regions along river have dense population with prosperous business and industrial establishments. The major pollution source includes domestic wastewater discharges, industrial wastewater discharges (e.g., tanning, paint and dye, chemical production, metal processing, electronic and foundry), municipal surface runoff, and transportation pollution (Chen et al., 2007). All the pollutants will eventually be transported to the river mouth and/or harbor and become deposited and accumulated in the bottom sediments.

The objective of this study is to investigate the Cr distribution in the surface sediment of the water body between river mouths (i.e., Love River, Canon River, and Jen-Gen River) and harbor entrance of Kaohsiung Harbor so that the degree of Cr enrichment, accumulation, and potential ecological risk can be evaluated.

1 Materials and methods

1.1 Sample collection and analysis

Ten sampling points were distributed in northern Kaohsiung Harbor, Taiwan (**Fig. 1**). On-site sampling of all 40 surface sediment was done on board a fishing boat in February, May, August, and October, 2011 at 10 stations selected in this study in northern Kaohsiung Harbor (**Fig. 1**). A global positioning system was employed to identify the precise location of each site. After collected with a 6 in. × 6 in. × 6 in. Ekman Dredge grab sampler (Jae Sung International Co., Taiwan), the surface sediment (0–10 cm) samples were temporarily placed in polyethylene bottles that had been pre-washed with acid; the bottles were stored in a dark ice chest filled with crushed ice.

After transported back to the laboratory, a small portion of the sample was subject to analysis of water content (105°C), and the remaining portion was preserved in –20°C freezer to be analyzed later. Prior to being analyzed, each sample was lightly crushed with a wooden board, and then screened through 1-mm nylon net to remove particles with diameters larger than 1 mm. One portion of the screened portion was subject to particle size analyses using a Coulter LS Particle Size Analyzer (Chen et al., 2007, 2012); the particles were classified into three groups, i.e., clay (<2 μm), silt (2–63 μm), and sand (>63 μm). Another portion was washed with ultra-pure water to remove sea salts; the salt-free particles were dried naturally in a dark place, grounded into fine powder with mortar and pestle made of agate, and then analyzed for organic matter (OM), total nitrogen (TN), total phosphorus (TP), total grease (TG), Cr, and aluminum (Al). OM was determined using the LOI (loss-on-ignition) method at 550°C; TN and TP of the sediment were analyzed according to USEPA Method 351.1 and USEPA Method 365.2 (US EPA, 1979), respectively. TG was determined according to procedures 5520E published in Standard Method (APHA, 2005).

For Al and Cr analyses, 0.5 g dry weight of the sediment sample was mixed with a mixture of ultra-pure acids (HNO_3:HCl:HF = 5:2:5, *V*/*V*/*V*), and was then heated to digest. The digested sample was filtered through 0.45-μm filter paper; the filtrate was diluted with ultra-pure water to a pre-selected final volume. The Al and Cr content were determined using a flame atomic absorption spectrophotometry (Hitachi Z-6100, Japan). Each batch of analyses was accompanied with a standard reference (marine sediment (PACS-2)) and a blank. For every 5 samples analyzed, the examination of standard solutions

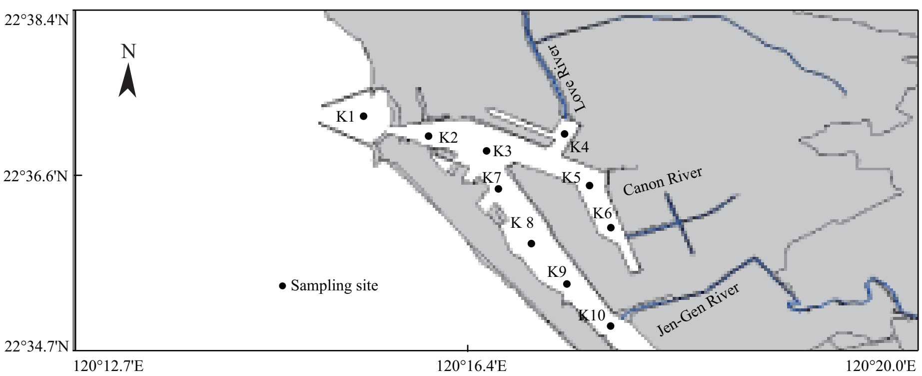

Fig. 1 Map of the study area and sampling locations (K1–K10).

was carried out to assure the stability of the instrument used. The standard reference of marine sediment (PACS-2) was found to contain (91.9 ± 2.1) mg/kg (n = 4) in our lab that is close to the certified values of (90.7 ± 4.6) mg/kg.

1.2 Data analysis

Data analysis (e.g., mean, standard deviation, maximum and minimum concentrations), using statistical methods, was performed in this study. To test the relationship between sediment characteristics (i.e., particle size, OM, TN, TP and TG) and Cr contents, Pearson correlation analysis was used with SPSS software (SPSS, version 12.0). The enrichment factor (EF) and geo-accumulation index (I_{geo}) were applied to evaluate the degree of Cr contamination; the sediment quality guidelines (SQGs) and potential ecological risk index (PERI) were employed to evaluate the sediment biological effects and potential ecological risk. EF is defined as:

$$EF = (C_{Cr}/C_{Al})_{sediment}/(C_{Cr}/C_{Al})_{crust} \quad (1)$$

where, C_{Cr} and C_{Al} is the Cr and Al content in sediment and crust, respectively.

The average Cr and Al content in the earth crust are 100 mg/kg and 8.23%, respectively, which excerpted from the data published by Taylor (1964). The I_{geo} values for the metals studied were calculated using the Müller's (1981) expression:

$$I_{geo} = \log_2(C_{Cr}/1.5B_{Cr}) \quad (2)$$

where, C_{Cr} is the measured content of element Cr, and B_{Cr} is the background content of Cr which is 100 mg/kg in the average shale (Taylor, 1964). Factor 1.5 is the background matrix correction factor due to lithogenic effects. The PERI is defined as (Hakanson, 1980):

$$PERI = PI \times T_i \quad (3)$$

$$PI = C_{Cr}/B_{Cr} \quad (4)$$

where, PI is the pollution index; T_i is its corresponding coefficient, i.e., 2 for Cr; C_{Cr} is the measured concentration of Cr in sediment; B_{Cr} is the background concentration of Cr (Hakanson, 1980). In this study, the average Cr concentration in earth crust of 100 mg/kg (Taylor, 1964) was taken as the Cr background concentration.

2 Results and discussion

2.1 Sediment characteristics

It has been reported that the distribution of particle size, OM, TN, TP, and TG content were correlated to Cr distribution in sediments (Chen et al., 2007). **Figure 2** presents the distribution of the major characteristics of surface sediments at ten monitoring stations studied including 4 times seasonal samplings in 2011. Results of sediment particle diameter analyses show that the major particles in all sediment samples are silt with diameter between 2 and 63 μm. The percentage compositions are 75.1%–88.5% for silt, 10.5%–21.3% for clay (<2 μm), and 0.0–11.5% for sand. Fine particles (<63 μm) that can easily adsorb and accumulate pollutants are the major component of particles found in the harbor sediments. As **Fig. 2b–f** shows, the water content, OM, TN, TP, and TG in the sediments from the study area have a similar spatial evolution characterized by the highest levels at stations K3, K4, K5, K6, and K9 and K10, which are located at the vicinity of Love River mouth, Canon River mouth, and Jen-Gen River mouth, respectively. TN, TP, and TG were relatively high in the vicinity of the river mouths compared with those at the harbor entrance areas (station K1). The results show that the anthropogenic contribution from the harbor tributaries is the major source of TN, TP, and TG (Chen et al., 2007). All surface sediment samples collected at ten monitoring stations studied contain 4.13%–5.83% of Al with an average of 4.81% ± 0.44% (**Fig. 2g**).

2.2 Distribution of Cr in sediments

The content of total Cr in the sediments of ten monitoring stations studied is between 27.0 and 361.9 mg/kg with an average of (113.5 ± 87.0) mg/kg. Concentration distributions of Cr in the northern Kaohsiung Harbor sediment shown in **Fig. 3** reveal that the sediment Cr content is relatively higher near the river mouths, especially in Jen-Gen River mouth (stations K9 and K10), and gradually decreases in the direction toward the harbor entrance (station K1). These observations clearly indicate that the upstream pollutants brought over by rivers are the major sources of harbor Cr pollution. These three rivers receive a great amount of industrial and domestic Cr from Kaohsiung City because about 40% domestic wastewater is discharged directly without adequate treatment. Moreover, several industrial plants discharge industrial wastewater effluents into the tributaries in or adjacent to Kaohsiung City (Chen et al., 2007), and the pollutants are transported by river flow and finally accumulate near the river mouth. Some pollutants may drift with sea current and are dispersed into open sea (Chen et al., 2007; Chen and Chen, 2011). Extremely high Cr concentration was observed in station K10, located near the Jen-Gen River mouth. The mean Cr concentration was high at 255.5 mg/kg, which was at least two times that of other stations. This might imply significant Cr contribution from upstream because regions along this river upstream have more than 36 tanneries that discharge their treated wastewater into the Jen-Gen River.

Coefficient of the Pearson correlation between the sediment characteristics and Cr content is shown in **Table 1**. The surface sediment Cr content is obviously correlated to water content ($p < 0.05$), TN ($p < 0.05$), and TG ($p < 0.01$) but not to either OM or particle size ($p > 0.05$) indicating that OM and particle size doesn't seem to major

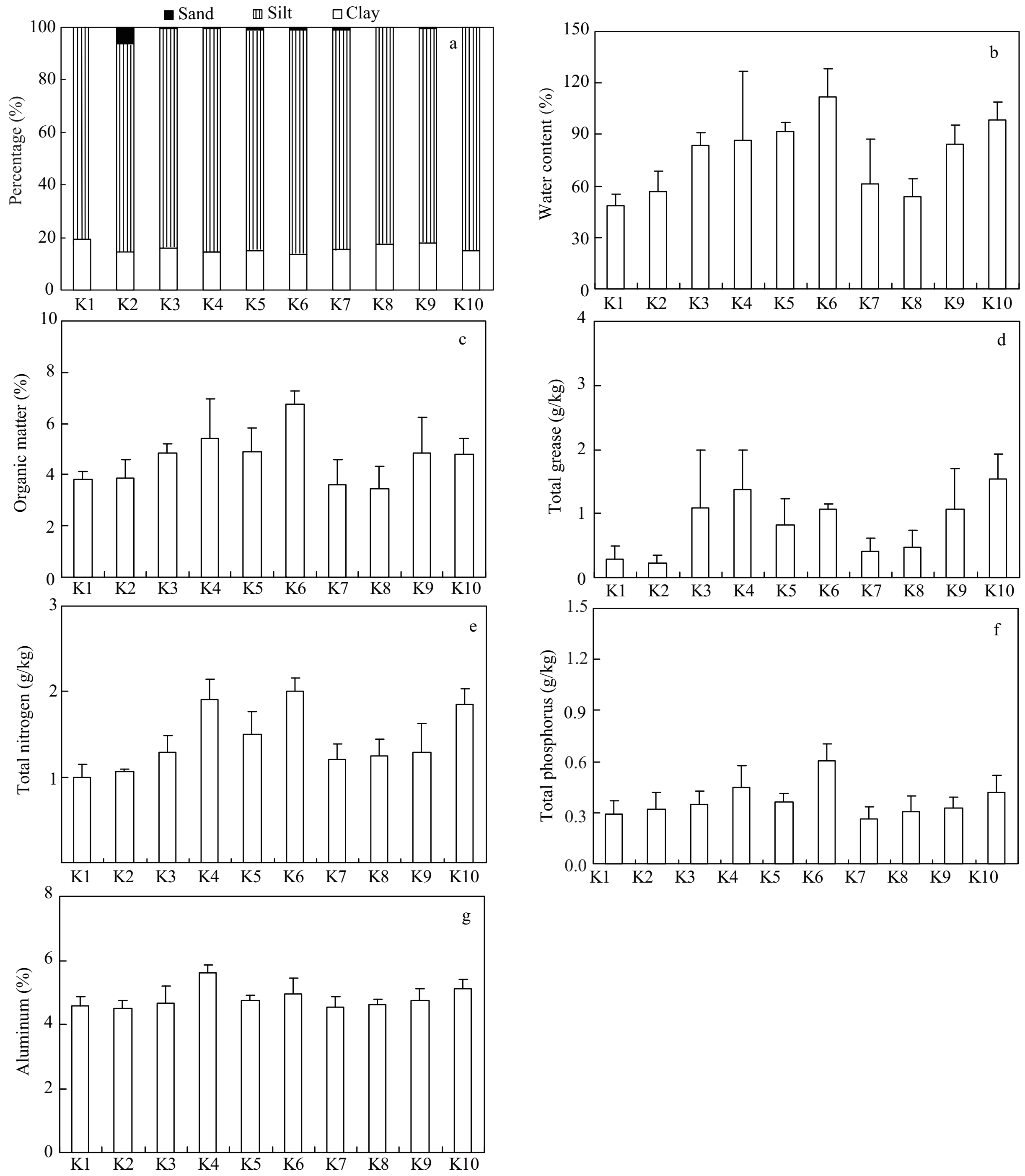

Fig. 2 Percentage compositions of particles size (a), and water content (b), organic matter (c), total grease (d), total nitrogen (e), total phosphorus (f), and aluminum (g) contents distributions in surface sediments at ten monitoring stations (n = 4).

factors to control the Cr distribution (Birkett et al., 2002). However, based on the concentrations of Cr, all stations were divided into two groups, one for low concentration (stations K1–K7), one for high concentration (stations K8–K10), and the significant correlation between Cr and organic matter content was found in stations K1–K7 (r = 0.41, $p < 0.05$), so was stations K8–K10 (r = 0.61, $p < 0.05$). The results suggest that the sediment organic matter played an important role in controlling the Cr distribution in sediments, whereas composition of organic matter can influence the partition of Cr.

2.3 Comparison with sediment quality guidelines

Several numerical sediment quality guidelines have been developed for assessing the contamination levels and the biological significance of chemical pollutants recently (Long et al., 1995; Riba et al., 2004). One of the widely used sediment toxicity screening guideline of the US Na-

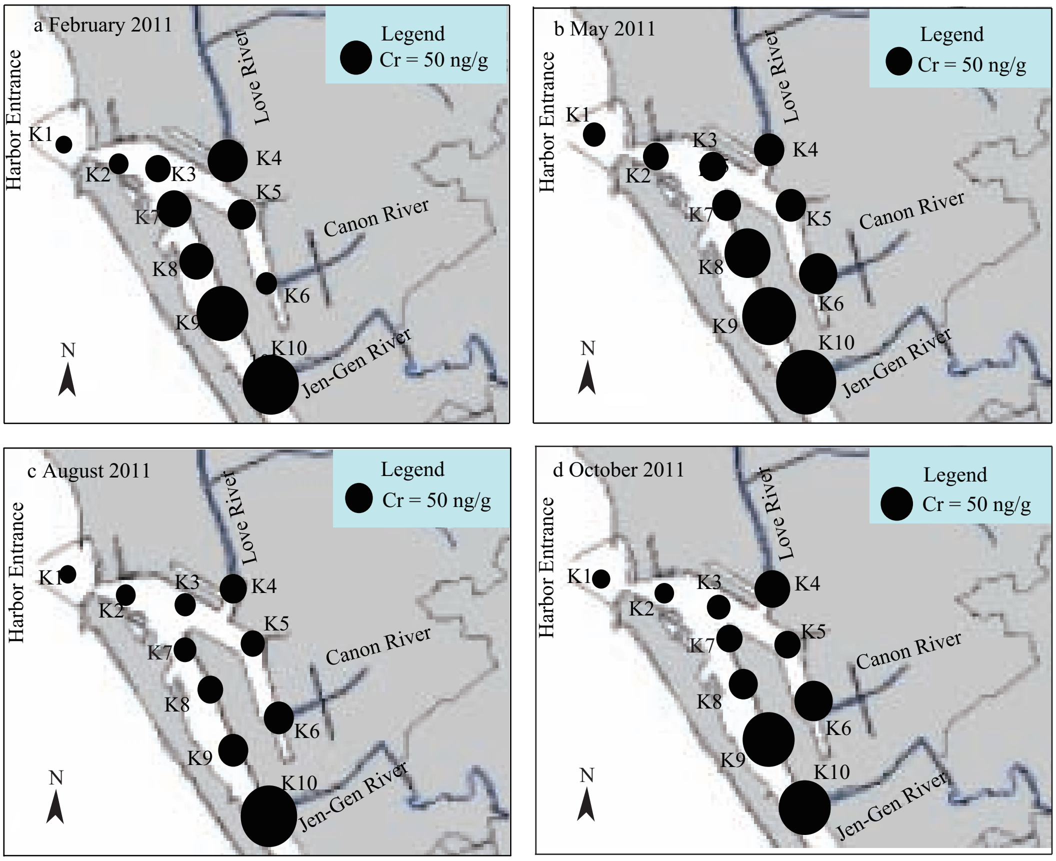

Fig. 3 Spatial distribution of Cr contents in surface sediment of northern Kaohsiung Harbor.

Table 1 Pearson correlation coefficients among sediment characteristics and Cr contents (n = 40)

	Clay	Silt	Sand	Water content	OM	TN	TP	TG	Al
Silt	–0.99[a]								
Sand	–0.88[a]	0.83[a]							
Water content	–0.03	0.05	–0.08						
OM	0.11	–0.08	–0.26	0.79[a]					
TN	0.11	–0.07	–0.33[b]	0.63[a]	0.59[a]				
TP	0.10	–0.05	–0.34[b]	0.56[a]	0.72[a]	0.69[a]			
TG	–0.09	0.11	–0.03	0.60[a]	0.35[b]	0.48[a]	0.17		
Al	0.11	–0.07	–0.30	0.32[b]	0.43[a]	0.61[a]	0.65[a]	0.25	
Cr	–0.06	0.05	0.11	0.40[b]	0.20	0.32[b]	0.12	0.54[a]	0.22

[a] Correlation is significant at the 0.01 level (2-tailed); [b] correlation is significant at the 0.05 level (2-tailed).
OM: organic matter; TN: total nitrogen; TP: total phosphorus; TG: total grease.

tional Oceanic and Atmospheric Administration provides two target values to estimate potential biological effects: effects range low (ERL) and effect range median (ERM) (Long et al., 1995). The guideline was developed by comparing various sediment toxicity responses of marine organisms or communities with observed metals concentrations in sediments. These two values delineate three concentration ranges for each particular chemical. When the concentration is below the ERL, it indicates that the biological effect is rare. If concentration equals to or greater than the ERL but below the ERM, it indicates that a biological effect would occur occasionally. Concentrations at or above the ERM indicate that a negative biological effect would frequently occurs.

Figure 4 shows the measured concentrations of Cr in comparison with the ERM and ERL values. Among the 40 sediment samples collected, the Cr is between ERL (81 mg/kg) and ERM (370 mg/kg) in 21 samples (52.2%). This indicates that the concentration of Cr found in these sediments may cause an adverse impact on aquatic lives. All other 19 samples are below ERL for Cr indicates that biological effects would rarely occur. Stations K1–K3, located at the vicinity of the mouths of harbor, all the sediment Cr contents were below ERL, whereas stations K4, K6, K9, and K10, located at the vicinity of the mouths of major rivers, the most sediment Cr contents were exceeded ERL (**Fig. 4**). This indicates that a biological effect would occur occasionally in mouths of Love River,

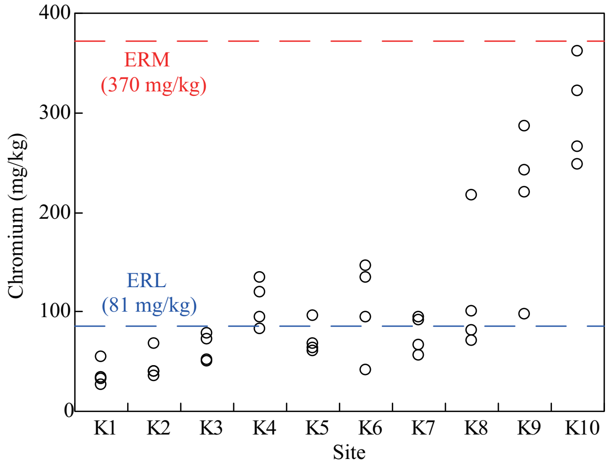

Fig. 4 Distribution of Cr contents in surface sediment of northern Kaohsiung Harbor.

Canon River, and Jen-Gen River sediments for Cr.

2.4 Enrichment factor

The enrichment factor (EF) is a useful tool for differentiating the man-made and natural sources of metal contamination (Adamo et al., 2005; Chen et al., 2007; Hung et al., 2009; Fang and Chen, 2010). This evaluating technique is carried out by normalizing the metal concentration based on geological characteristics of sediment. Aluminum is a major metallic element found in the earth crust; its concentration is somewhat high in sediments and is not affected by man-made factors. Thus, Al has been widely used for normalizing the metal concentration in sediments (Huang and Lin, 2003; Woitke et al., 2003; Chen et al., 2007; Hung et al., 2009; Fang and Chen, 2010). When the EF of a metal is greater than 1, the metal in the sediment originates from man-made activities, and vice versa. The EF value can be classified into 7 categories (Birth, 2003): (1) no enrichment for EF < 1; (2) minor for 1 < EF < 3; (3) moderate for 3 ⩽ EF < 5; (4) moderately severe for 5 ⩽ EF < 0; (5) severe for 10 ⩽ EF < 25; (6) very severe for 25 ⩽ EF < 50; and (7) extremely severe for EF ⩾ 50.

Table 2a shows EF values of the sediment Cr for ten monitoring stations studied; the Cr concentration is consistent with the EF value of Cr for all sampling stations, and except stations K1 and K2, all EF values are greater than 1. This indicates that the sediment Cr has enrichment phenomenon with respect to the earth crust and that all Cr originates from man-made sources. Stations K9–K10 are classified as moderate enrichment, stations K3–K8 are classified as minor enrichment, and stations K1–K2 are classified as no enrichment, respectively. These results demonstrate that the sediment near the mouth of Jen-Gen River experiences moderate enrichment of Cr that originates from the upstream sources of pollution. Additionally, the EF value of 4.81 obtained in station K10 (Jen-Gen River mouth) is lower than the EF value of 8.4 reported earlier (Chen et al., 2007) indicating that the upstream pollution has been reduced so that the accumulation of pollutants in sediments is not as serious as during earlier years. This observation may show the effectiveness of intercepting the river flow and dredging the river mouth.

2.5 Geo-accumulation index

Similar to metal enrichment factor, geo-accumulation (I_{geo}) index can be used as a reference to estimate the extent of metal accumulation. The I_{geo} value can be classified into 7 classes: 0, none for I_{geo} < 0; 1, none to medium for I_{geo} = 0–1; 2, moderate for I_{geo} = 1–2; 3, moderately strong for I_{geo} = 2–3; 4, strong for I_{geo} = 3–4; 5, strong to very strong for I_{geo} = 4–5; and 6, very strong for I_{geo} > 5. Based on the I_{geo} data and Müller's (1981) geo-accumulation indexes, the accumulation levels with respect to Cr at each station are ranked in **Table 2**. Stations K9 and K10 are classified as none to medium accumulation (Class 1), and stations K1–K8 are classified as none accumulation (Class 0). Chromium showed, in general, geoaccumulation indices between Class 0 and Class 1 in northern Kaohsiung Harbor. Similar results were found in the Cua Ong Harbor in Vietnam (Ho et al., 2010) and Kaohsiung Harbor in Taiwan (Chen et al., 2007).

Table 2 EF, I_{geo}, and PERI of Cr for each station studied at northern Kaohsiung Harbor

Station	Enrichment factor			Geo-accumulation index			Potential ecological risk		
	EF value	EF class	EF level	I_{geo} value	I_{geo} class	I_{geo} level	PI	PERI	Risk level
K1	0.66	1	No enrichment	–2.08	0	None	0.37	0.73	Low
K2	0.85	1	No enrichment	–1.76	0	None	0.46	0.92	Low
K3	1.14	2	Minor	–1.28	0	None	0.63	1.26	Low
K4	1.59	2	Minor	–0.50	0	None	1.08	2.16	Low
K5	1.24	2	Minor	–1.09	0	None	0.72	1.43	Low
K6	1.73	2	Minor	–0.68	0	None	1.04	2.08	Low
K7	1.42	2	Minor	–0.99	0	None	0.77	1.54	Low
K8	2.11	2	Minor	–0.50	0	None	1.18	2.35	Low
K9	3.75	3	Moderate	0.39	1	None to medium	2.11	4.23	Low
K10	4.81	3	Moderate	0.98	1	None to medium	3.00	5.99	Low

PI: pollution index; PERI: potential ecological risk index.

2.6 Potential ecological risk

The potential ecological risk index (PERI) is applied to evaluate the potential risk associated with the accumulation of Cr in surface sediments. PERI that was proposed by Hakanson (1980) can be used to evaluate the potential risk of one metal or combination of multiple metals. The calculated PERI values can be categorized into 5 classes of potential ecological risks: low risk (PERI < 40), moderate risk (40 ⩽ PERI < 80), higher risk (80 ⩽ PERI < 160), high risk (160 ⩽ PERI < 320), and serious risk (PERI ⩾ 320). **Table 2** lists the PI value, PERI value, and risk classification for the Cr contained in the surface sediment samples collected in this study. All stations are classified as low risk with respect to Cr pollution. The above evaluation results indicate that the Cr contained in surface sediments at the study area has low potential ecological risks. However, the PERI value near the Jen-Gen River mouth of stations K9 and K10 are higher than other sites.

3 Conclusions

The surface sediment samples collected between the river mouths and harbor entrance in northern Kaohsiung Harbor contained 27.0–361.9 mg/kg of Cr with an average of (113.5 ± 87.0) mg/kg. The distribution of Cr in surface sediments reveals that the Cr originates from the river upstream discharges of industrial and domestic wastewaters; it is transported along the river and finally deposited and accumulated near the river mouth. Base on the comparison with sediment quality guidelines, the concentrations of Cr in the mouths of Jen-Gen River sediments may cause acute biological damage.

Results from the EF and I_{geo} analyses imply that the Jen-Gen River sediments can be characterized as moderate enrichment and none to medium accumulation of Cr, respectively. Compared to the EF values reported earlier (Chen et al., 2007), the degree of Cr enrichment at the river mouths has been obviously reduced. This may be associated with river renovation and river mouth dredging. However, results of potential ecological risk evaluation show that the Cr contained in surface sediment of northern Kaohsiung Harbor has low potential ecological risks. The results can provide regulatory valuable information for references with the aim of extending future strategies to renovate and manage river mouth and harbor.

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