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Distribution of heavy metals in tidal flat sediments and their bioaccumulation in the crab *Macrophthalmus japonicas* in the coastal areas of Korea

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ABSTRACT: Levels of heavy metal concentrations in crab, *Macrophthalmus japonicas*, and in surface sediments from tidal flats in the western and southern coastal area of the Korean Peninsula were assessed in terms of metal pollution and bioaccumulation. Metal concentrations in tidal flat sediments varied considerably at various sites, mostly because of the high variability of pollutants commonly found in the area. Levels of some metals in the sediments exceeded the minimum concentration guidelines effects-range, leading to the expectation of adverse biological effects. The pollution index associated with different metals varied greatly and indicated that As, Cr, Cu and Pb contamination is relatively high in the coastal areas of Korea. The combined pollution index ranged from 0.49 to 2.88 and spatial distribution analysis indicated that runoff from an abandoned metal mine and from a variety of other industries are the main sources of metal pollution. Bulk metal concentrations in crabs showed no significant relationship with size but significant differences with sex: levels of most non-essential metals were significantly higher in females than in males, but levels of essential metals such as Cu and Zn showed no significant difference associated with sex. The concentrations of most metals in crabs were significantly positively related to those in sediments, indicating the usefulness of crabs as a bioindicator for metal pollution assessments in tidal flats. Bioaccumulation factors (BAF) were in the order of Cu>Cd>Zn>As>Ni>Pb>Cr and tended to be significantly inversely related to exposure concentrations. This indicates that other factors, besides BAF, also need to be taken into account when assessing the hazard potential of particular metals.

Key words: tidal flat, sediments, heavy metal, crabs, bioaccumulation

1. INTRODUCTION

Heavy metals, transported via drainage channels and direct discharges, tend to become trapped in coastal areas and accumulate in coastal sediments. Although many metals are essential for life, some are harmful, particularly at high concentrations, and may adversely affect aquatic biota, either through leaching into the aqueous phase or by direct contact.

Assessing ecological risks and environmental impacts of metals in sediment presents several challenges. Heavy metal concentrations in sediments are indicators of potential toxic effects because metal bioavailability can vary considerably between sediments (Chapman et al., 1998; Borgmann et al., 2004). A number of pollution monitoring studies have reported on the biological effects of pollution in marine organisms and shellfish are widely used to assess the bioavailability of metals in coastal areas (Sanders, 1984; McPherson and Brown, 2001; Reichmuth et al., 2010). Crustaceans are known to accumulate metals from surrounding waters and sediments by adsorption or direct ingestion from food, water and/or sediment (Bryan, 1971, 1979). Nevertheless, few studies have investigated the bioaccumulation of heavy metals in crabs.

The coastal areas are an important ecosystem because of their many functions that are useful to humans (Mitsch and Gosselink, 1993). Despite international recognition for the ecological importance of tidal flats as wetland habitats, increasing urban and industrial development in surrounding areas continuously pose a threat to these ecologically-sensitive areas (Kueh and Chui, 1996; Ongche, 1999). Many tidal flats are distributed along the west and south coasts of the Korean Peninsula, collectively making up an area of approximately 2,400 km^2. Most of these tidal flats have fine-grained sediment, transported from surrounding landmasses. Over the past several decades, industrialization and urbanization have taken place in Korea and this has been accompanied by an increase in heavy metal and organic pollution in the coastal environment.

This study aims to assess the levels of heavy metals and their spatial distribution in tidal flat sediments as well as their bioaccumulation in crabs. It also evaluates the degree to which human disturbance and various anthropogenic factors (such as urbanization and industrialization) affect Korea's coastal areas. The significance of regional differences, pollution of sediments, interactions between metals found in sediments and in crabs, and bioaccumulation factors are evaluated and discussed.

2. MATERIALS AND METHODS

2.1. Sampling Methods

Sediments and crabs were collected from nine tidal flat localities on the western and southern coasts of Korea from

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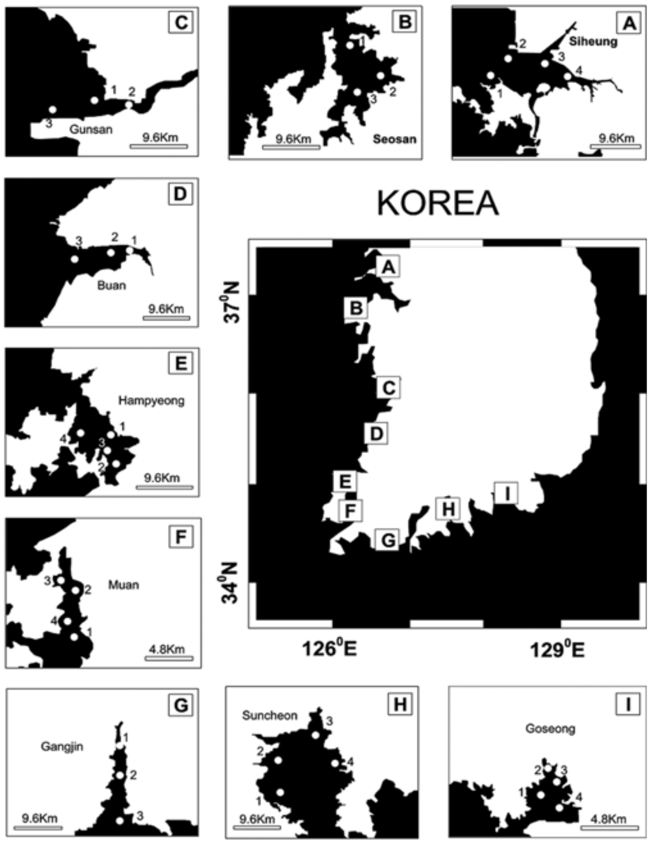

Fig. 1. Map showing the tidal flats studied and the sampling sites from the coastal areas of Korea.

September to October 2008–2009 (Fig. 1). The tidal flats were classified into two groups according to the most common human activities affecting them: those near industrial complexs/urban sewage outfalls (Site A and Site C), an urban sewage outfall (Site H) and an abandoned copper mine (Site I), discharges from stockbreeding (Site B), small pottery factories (Site G) and rural villages (Sites D, E and F). Details relating to geographical location, area, and degree of human disturbance in each tidal flat are summarized in Table 1. Intertidal surface sediments (the upper 5 cm) were collected, during the ebb tide, from 32 points in the near

Table 1. Geographical locations and human activities of the study areas

Site	Location	Area (km^2)	Human activities
A	Banweol	265.9	industrial complex/urban sewage
B	Garolim	307.9	stockbreeding/farming
C	Janghang	196.5	industrial complex/urban sewage
D	Gomso	120.8	farming/rural villages
E	Hampyeong	146.3	farming/rural villages
F	Muan	95.1	farming/rural villages
G	Gangjin	20.3	small pottery factories/farming
H	Suncheon	1023.1	urban sewage/tourism
I	Goseong	26.9	abandoned Cu mine

shore area above tidal flats. A small amount of the surface sediment was taken (using a plastic scoop) from more than 10 points randomly selected within an area of approximately 10 m^2. The samples were then mixed in a polyethylene bag to avoid the influence of local disturbance and to obtain a homogenous representative of the site. This mixed sediment was counted as one sample representing a site, stored in an ice box, and transported to the laboratory.

The crab, *Macrophthalmus japonicas,* was selected because this species is widely distributed along the western and southern coasts of Korea and commonly inhabits the tidal flats where the study took place. Ten to 15 crabs with approximately similar carapace width (12–14 mm) were captured at each sampling point. Additional assessments on the effects of gender and size, in terms of tissue concentrations, were carried out on crabs collected at Site H. In addition to carapace width (12–20 mm) these crabs were also classified into gender categories. Captured crabs were carefully washed with seawater, placed into polyethylene bottles containing local seawater, and stored in an ice box, prior to transport to the laboratory.

2.2. Analytical Methods

Sediments were washed with deionized water to remove seawater and dried at 105 °C for 48 h. A subsample of the dried sediments was characterized for grain size using Sedigraph techniques (Lum et al., 1996). The samples were placed in beakers and a 10% solution of H_2O_2 was added to digest organic matter in the sediments. After a complete reaction, the excess H_2O_2 solution was removed by evaporation, cooled, and 0.5% of sodium hexameta-phosphate was added. The mixture was then analyzed to determine sediment grain size distribution, using an X-ray automatic grain size analyzer (SediGraphy 5100, Micromeritics Co., USA). Loss on ignition (LOI) was obtained from samples ashed at 550 °C in a muffle furnace for 1 h (Barille-Boyer et al., 2003). For analysis of metal concentrations, the sediments were prepared for digestion by grinding to a fine powder (100 mesh) using an agate mortar. The samples (0.500 ± 0.002 g) were digested with a mixture of $HClO_4$/HF (1:5) in a Teflon bottle at 140 °C for 4 h (Pempkowiase et al., 1999). After digestion, the samples were diluted with a 0.1 M HNO_3 solution to a final volume of 50 mL. To analyze labile metals in the sediments, an extraction solution was prepared by mixing 7 g of dried sediment with 25 mL of 0.5 M HCl and shaking it at room temperature for 30 min. After 24 h, the solution was filtered (Sutherland and Tolosa, 2001).

Crab samples were washed with deionized water and dried at 65 °C to constant weight. To minimize the influence of biological discrepancies between the crabs collected, only four to five male crabs of the same carapace size (12–14 mm) per sampling point were selected for determination of metal concentrations. The samples, comprised of four to five male crabs, were homogenized by crushing and grinding them to a fine powder (≤100 mesh) using a PVC pestle and an agate mortar. The powdered samples were dried at 105 °C for 24 h, after which 0.12 g of each dried sample was placed in a Teflon bottle and digested with a mixture of $HClO_4$ (30%) and HNO_3 (10%). Samples were later refluxed on a hot plate and diluted to 25 mL with 0.1 M HCl (Al-Mohanna and Subrahmanyam, 2001).

The concentrations of heavy metals in all decomposed samples were determined using inductively coupled plasma atomic emission spectrometer (Spectroflame EOP, Spectro Analytical Instruments, Germany) and inductively coupled plasma-mass spectrometer (PQ-Excell, Thermo Elemental, USA). The metals analyzed included As, Cd, Cr, Cu, Ni, Pb and Zn. During the analysis, data were assessed for accuracy and precision using a quality control system that included reagent blanks and triplicate reference (NIST1646a) samples integral to the analytical procedure. At least one duplicate was run for every six samples to verify the precision of the analysis. The bias of the chemical analysis was less than 10% and the precision was approximately 5% for Cu and Zn and 10% for the other metals, at a 95% confidence level.

Bioaccumulation factor (BAF), which is used to assess metal concentrations in sediments and their bioaccumulation in crabs, was calculated using the following formula:

$$BAF = C_{icrab}/C_{ised} \quad (1)$$

where C_{icrab} is metal concentration (mg kg^{-1} dry wt.) in crab tissue, and C_{ised} is metal concentration (mg kg^{-1} dry wt.) in sediment (Barron, 1995). To assess the environmental quality of the sediment, the pollution index (PI) of each metal and the combined pollution index (CPI) of the metals were calculated for each site. The Interim Sediment Quality Guidelines (ISQG, Environment Canada, 1998) was used to calculate the PI and CPI:

$$PI = C_i/S_i \quad (2)$$

$$CPI = (\Sigma C_i/S_i)/n \quad (3)$$

where C_i is the concentration of metals determined from sediments, and S_i is the guideline for metals in sediments. Statistical analyses were carried out using the SPSS statistical program package for Windows.

3. RESULTS AND DISCUSSION

3.1. Physical Chemistry of Sediments

Determinations of the particle size and organic material content of sediments in marine environments are important to understand their physical chemistry. Haque and Subramanian (1982) found that relative metal adsorption capacity was in the order of sand<silt<clay, due to their increasing surface areas with decreasing the particle sizes. The average values of particle size and loss on ignition (LOI) are presented in Table 2. In the study area, the average fractions of sand, silt and clay are 3.9%, 49% and 47%, respectively, and relatively high proportions of clay were found at Site C and Site H. Although results showed a significantly overlapping range, in general the clay fraction of sediments increased in the southward direction and decreased at the southeastern areas, for example, at Site I. Site H and Site E had the highest (8.3%) and the lowest (2.9%) LOI values, respectively. In general, finer-grained sediments had higher organic matter content. The LOI content of the sediments collected near the drainage area was generally higher compared to those collected closer to the sea. This indicates organic matter input from the land, mostly associated with human-associated wastes such as sewage effluents and agricultural byproducts.

3.2. Heavy Metal Concentrations in Sediments

The concentrations of heavy metals in surface sediment samples are shown in Table 3. These were highly variable, depending on the sites and on sampling points within the same site. Site I, located in the section of estuary that receives drainage channels from a nearby copper mine area, has the highest concentrations of As, Cu, Pb and Zn. Site F, located near a small fishing village and small oyster beds,

Table 2. Physical properties of tidal surface sediments collected from study area in Korea

Location	Grain-size Distribution (%)			Ignition Loss (%)
	sand	silt	clay	
A	3.03 ± 3.45	69.40 ± 8.31	24.97 ± 11.73	4.73 ± 1.70
B	1.17 ± 0.74	70.73 ± 8.01	27.10 ± 7.27	4.13 ± 1.48
C	3.57 ± 0.90	43.33 ± 11.62	52.43 ± 12.30	4.99 ± 0.59
D	0.60 ± 0.22	74.63 ± 7.55	22.37 ± 7.12	4.23 ± 0.33
E	0.33 ± 0.12	74.33 ± 13.86	23.90 ± 14.46	2.93 ± 0.47
F	0.93 ± 0.78	57.43 ± 12.98	41.07 ± 12.99	3.52 ± 0.78
G	0.58 ± 0.31	51.78 ± 6.06	47.13 ± 5.73	5.91 ± 1.25
H	0.10 ± 0.07	39.58 ± 4.64	59.60 ± 4.57	8.34 ± 0.85
I	3.38 ± 3.10	78.40 ± 11.12	17.83 ± 10.31	4.06 ± 0.30

Table 3. Total concentrations of heavy metals (mean ± sd, mg kg^{-1} dry wt.) in the sediments from the coastal areas of Korea

Site	As	Cd	Cr	Cu	Ni	Pb	Zn
A	6.97 ± 2.86	0.43 ± 0.43	63.2 ± 16.5	56.1 ± 61.4	28.2 ± 9.6	40.7 ± 18.8	69.4 ± 4.5
B	5.28 ± 1.70	0.13 ± 0.05	61.6 ± 26.3	13.5 ± 5.8	30.9 ± 21.7	25.5 ± 3.8	58.5 ± 14.2
C	7.91 ± 1.74	0.21 ± 0.03	56.9 ± 5.1	38.9 ± 11.5	23.7 ± 2.5	35.9 ± 3.4	85.1 ± 6.1
D	6.54 ± 1.20	0.13 ± 0.02	46.0 ± 5.9	15.6 ± 2.8	18.1 ± 3.2	27.9 ± 6.4	61.7 ± 11.0
E	6.13 ± 0.40	0.08 ± 0.01	46.5 ± 6.4	20.4 ± 9.3	19.1 ± 3.8	25.8 ± 2.1	65.5 ± 8.7
F	4.52 ± 1.03	0.07 ± 0.02	30.8 ± 8.7	8.3 ± 1.4	12.6 ± 4.2	25.0 ± 1.9	45.5 ± 9.3
G	9.69 ± 1.33	0.13 ± 0.02	66.1 ± 3.6	17.6 ± 1.2	29.3 ± 2.5	30.3 ± 1.4	96.0 ± 6.3
H	6.33 ± 1.37	0.10 ± 0.03	54.2 ± 17.6	13.5 ± 4.8	24.3 ± 8.6	27.9 ± 2.3	75.5 ± 23.2
I	21.66 ± 1.32	0.20 ± 0.14	32.1 ± 4.7	164.4 ± 51.6	5.2 ± 1.2	83.0 ± 8.6	225.9 ± 15.1
Quanzhou Bay, China[a]	21.7 ± 3.8	0.59 ± 0.18	82.0 ± 19.4	71.4 ± 29.4	33.4 ± 7.3	67.7 ± 16.9	179.6 ± 33.2
Sydney Harbor, Australia[b]	21 ± 9	2.8 ± 2.4	81 ± 63	200 ± 190	20 ± 12	360 ± 310	1000 ± 1500
ERL–ERM[c]	8.2–70	1.2–9.6	81–370	34–270	20.9–51.6	46.7–218	150–410
ISQG[d]	7.27	0.70	52.3	18.7	–	30.2	124
Trigger value(L–H)[e]	20–70	1.5–10	80–370	65–270	21–52	50–220	200–410
P–S standard[f]	20–65	0.5–1.5	80–150	35–100	–	60–130	150–350

[a]Yu et al. (2008); [b]McCready et al. (2006); [c]effects-range-low(ERL) and –medium(ERM) from Long et al. (1995); [d]guideline values from Environment Canada (1998); [e]sediment quality guideline trigger values (low-high) from ANZECC/ARMCANZ (2000); [f]primary(P) and secondary(S) standard values from CSBTS (2002); "–" means not available.

has the lowest concentrations of heavy metals. Sites A and C are adjacent to an industrial complex and they showed significantly high levels of heavy metals such as As, Cd, Cr, Pb and Zn. The high levels of some heavy metals observed at Sites A and C appeared to be related to wastewater effluents from the nearby industrial complex. Site G has the highest concentration of Cr, as well as As and Zn. The development of the pottery industry near Site G may have caused higher levels of these metals as this industry uses metal-containing glaze. The varying levels of heavy metals in the different sediment samples can therefore be attributed to the kinds of pollutants that affect the area.

This study also shows that the concentrations of most heavy metals in the sediments collected from the tidal flats in the western and southern coasts of the Korean Peninsula were significantly lower than those recorded in other countries, such as Quanzhou Bay, China, and Sydney Harbour, Australia (Table 3). However, the concentrations of Zn in almost all of the sites sampled and of Pb at Site I, and Cu at Sites A, C and I, were relatively higher than those reported in unpolluted marine sediments around the world. Such unpolluted sediments, from marine and freshwater sediments worldwide generally contain low concentrations of heavy metals: <1 mg kg^{-1} Cd, <60 mg kg^{-1} Cr, <20 mg kg^{-1} Cu, <00 mg kg^{-1} Ni, 2–50 mg kg^{-1} Pb, and <50 mg kg^{-1} Zn (Moore and Ramamoorthy, 1984; Bryan and Langston, 1992).

Extractable concentrations of Cr, Cu, Pb and Zn (with 0.5 M HCl) and the mean ratios of extractable/total concentrations are shown in Table 4. As is the case for the total concentrations, the extractable concentrations of these heavy metals vary greatly for different metals and at different sites: 6.12 ± 2.67 mg kg^{-1} for Cr, 19.4 ± 33.2 mg kg^{-1} for Cu, 20.0 ± 16.3 mg kg^{-1} for Pb, and 2.22 ± $^{-}$1.87 mg kg^{-1}

Table 4. Extracted concentrations of heavy metals (mean ± sd, mg kg^{-1} dry wt.) from the sediments and their extraction percentages relative to total concentrations of heavy metals

Site	Cr		Cu		Pb		Zn	
	Conc.	Ex.%	Conc.	Ex.%	Conc.	Ex.%	Conc.	Ex.%
A	10.1 ± 4.7	15.4 ± 5.3	11.4 ± 6.1	57.5 ± 17.4	21.5 ± 14.5	48.7 ± 12.4	5.64 ± 3.34	8.38 ± 5.43
B	6.9 ± 2.6	11.4 ± 1.7	3.7 ± 1.4	27.4 ± 1.4	10.5 ± 4.3	39.8 ± 11.7	1.29 ± 0.30	2.21 ± 0.19
C	5.9 ± 0.7	10.6 ± 0.9	17.8 ± 7.5	44.6 ± 14.7	17.3 ± 4.0	47.8 ± 10.2	1.57 ± 0.17	1.86 ± 0.16
D	5.6 ± 0.5	12.2 ± 0.8	6.2 ± 1.4	39.6 ± 5.2	13.9 ± 5.4	48.0 ± 8.5	1.35 ± 0.16	2.20 ± 0.11
E	5.0 ± 1.0	10.7 ± 0.8	11.9 ± 9.5	50.7 ± 18.4	13.1 ± 4.7	49.6 ± 13.3	1.93 ± 0.48	2.96 ± 0.66
F	4.2 ± 0.7	14.3 ± 3.3	3.6 ± 0.5	45.6 ± 14.4	8.3 ± 0.9	33.7 ± 5.4	0.97 ± 0.03	2.24 ± 0.56
G	7.6 ± 1.0	11.4 ± 0.9	5.9 ± 0.7	33.6 ± 1.7	19.1 ± 0.9	62.9 ± 0.5	1.67 ± 0.38	1.74 ± 0.41
H	6.1 ± 2.1	11.1 ± 0.7	5.4 ± 2.2	40.8 ± 9.4	13.2 ± 4.5	46.9 ± 13.5	1.46 ± 0.62	1.91 ± 0.36
I	4.2 ± 0.4	13.1 ± 0.7	104.4 ± 33.8	63.2 ± 0.9	60.3 ± 7.4	72.4 ± 1.4	3.82 ± 0.23	1.69 ± 0.01

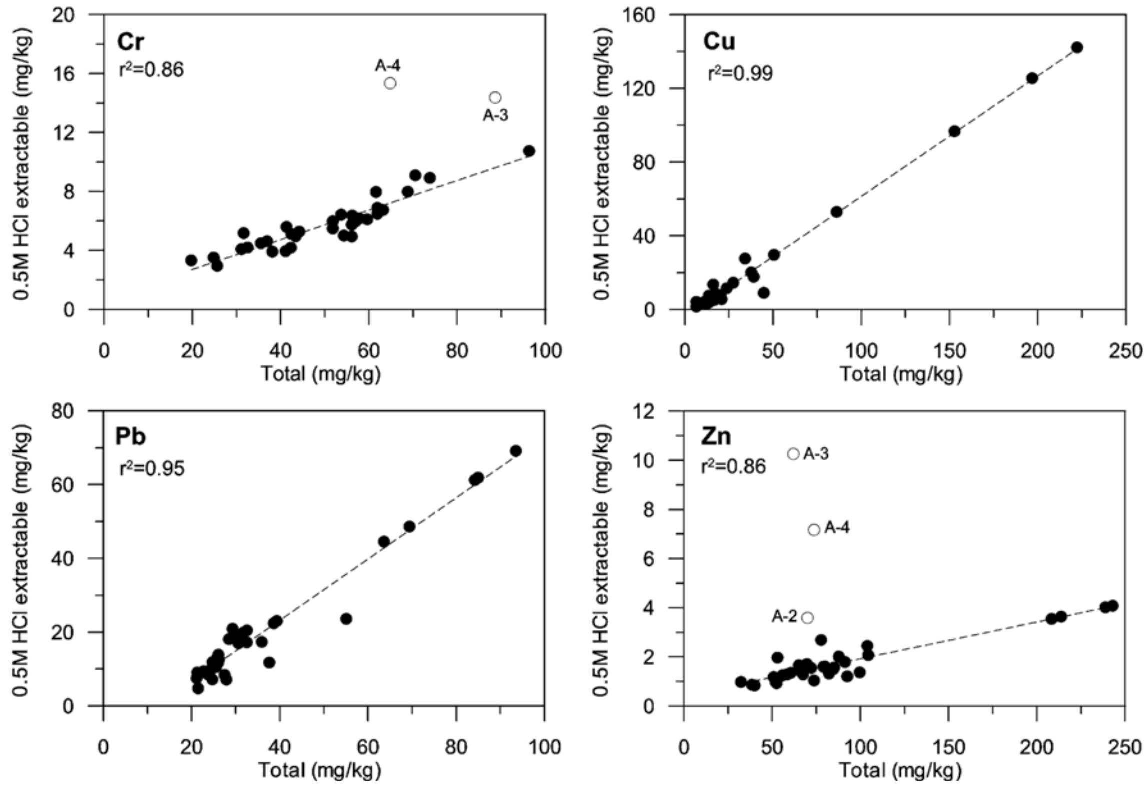

Fig. 2. Relationships between total and 0.5M HCl-extracted concentrations of heavy metals in the sediments.

for Zn. Nevertheless, significant positive correlations were found between the extractable metal concentrations and the total metal concentrations ($r > 0.9$, $p < 0.001$; Fig. 2), where outlier data points such as A-2, A-3 and A-4 were excluded before regression analyses for Cr and Zn. The excluded samples at Site A, especially A-3 and A-4 which were collected near an industrial complex (Fig. 1), showed significantly high extraction ratios of Cr and Zn in comparison with those collected from other sample points. The elevated extraction ratios of Cr and Zn observed from the excluded may possibly be due to plating and dyeing pollution from effluents containing soluble Cr and Zn from factories clustered in this industrial complex.

Extractable fractions of Cu and Pb accounted for a relatively large percentage of the total concentrations of 44.76 ± 15.64% and 50.48 ± 14.53%, respectively, while those of Cr and Zn were not over 15% of the total concentrations. Percentages of the extractable fractions of the metals were highly variable in terms of the sites as well as the metals. For example, the extractable fractions of Pb varied from 33.70 ± 5.46% at Site F to 72.45 ± 1.43% at Site I (Table 4). Compared with other sites, the highest extractable fractions were found at Site I for Cu and Pb, and at Site A for Cr and Zn. This is due to these being located near a copper mine and a coastal industrial complex, respectively. Dilute HCl leachates liberate adsorbed derital and non-detrital carbonate-bound metals and much of the Fe/Mn oxide and organic-associate metals, but minimizes the loss of residual silicate-bound metals (Sutherland and Tolosa, 2001; Sutherland, 2002; Sutherland et al., 2004). These results suggest that heavy metals have different associations with the sediment and/or different solubility during the HCl extraction, depending on the sites they were obtained from, which may be due to differences in the sources of metal pollution and the physical-chemical conditions favoring sediment contamination.

3.3. Metal Pollution Levels in Sediment

Evaluating the degree of sediment contamination in coastal areas often involves comparing heavy metal concentration with sediment quality guidelines (SQG) or quantifying the accumulation factor (PI) with respect to background values. Numerical SQGs have been developed during the past decades to protect animals, living in or near sediments, from the deleterious effects of sediment-bound contaminants, or to evaluate the spatial pattern of sediment contamination (Long et al., 1995; Environment Canada, 1998; Australian and New Zealand Environment and Conservation Council and Agriculture and Resource Management Council of Australia and New Zealand, 2000; China State Bureau of Quality and Technical Supervision, 2002). In this study, we use these SQGs to assess the degree of heavy metal contamination in tidal flat sediments because there is no available guideline and background information for

Table 5. Sediment pollution index and combined pollution index (mean ± sd) calculated using heavy metal concentrations determined and the recommended values from ISQG

Site	As	Cd	Cr	Cu	Pb	Zn	CPI
A	0.96 ± 0.39	0.62 ± 0.61	1.21 ± 0.32	3.00 ± 3.29	1.35 ± 0.62	0.56 ± 0.04	1.28 ± 0.77
B	0.73 ± 0.23	0.18 ± 0.07	1.18 ± 0.50	0.72 ± 0.31	0.84 ± 0.13	0.47 ± 0.11	0.69 ± 0.19
C	1.09 ± 0.24	0.29 ± 0.05	1.09 ± 0.10	2.08 ± 0.62	1.19 ± 0.11	0.69 ± 0.05	1.07 ± 0.05
D	0.90 ± 0.16	0.19 ± 0.03	0.88 ± 0.11	0.83 ± 0.16	0.93 ± 0.21	0.50 ± 0.09	0.70 ± 0.07
E	0.84 ± 0.06	0.12 ± 0.02	0.89 ± 0.12	1.09 ± 0.50	0.86 ± 0.07	0.53 ± 0.07	0.72 ± 0.11
F	0.62 ± 0.14	0.10 ± 0.04	0.59 ± 0.17	0.45 ± 0.08	0.83 ± 0.06	0.37 ± 0.08	0.49 ± 0.08
G	1.33 ± 0.18	0.18 ± 0.03	1.26 ± 0.07	0.94 ± 0.06	1.00 ± 0.05	0.77 ± 0.05	0.92 ± 0.06
H	0.87 ± 0.19	0.14 ± 0.04	1.04 ± 0.34	0.72 ± 0.26	0.93 ± 0.08	0.61 ± 0.19	0.72 ± 0.17
I	2.98 ± 0.18	0.29 ± 0.20	0.61 ± 0.09	8.79 ± 2.76	2.75 ± 0.29	1.82 ± 0.12	2.88 ± 0.55

heavy metals in tidal flat sediments in Korea.

The comparison of results from this study with those of SQGs (Table 3) shows that the concentrations of most heavy metals in tidal flat sediments in the western and southern coasts of Korea exceed the ISQG, the threshold set by Environment Canada (1998), but generally meet other SQGs. As shown in Table 3, there are, however, significant variations in heavy metal concentrations, with some exceeding the effects-range-low (ERL) guideline (for example, As at Sites G and I; Cu at Sites A, C, and I; Ni at Sites A, B, C, G, and H; Pb and Zn at Site I). The concentrations equal to or greater than ERL, but lower than effects-rang-median (ERM), represent a probable effect range in which adverse biological effects occasionally occur (Long et al., 1995).

The PI values, calculated according to the ISQG of heavy metals, vary greatly in different metals (Table 5). The PI values for Cr in sediments varied from 0.38 to 1.84, with about 47% of the analyzed samples presenting high PI of >1, which indicated moderate contamination. Arsenic, Cu and Pb presented low to high contamination levels, with much variability in PI values: 0.43–3.26 for As, 0.35–11.88 for Cu and 0.70–3.10 for Pb. High PI values (PI > 1) were found in 44% of the samples for As and Cu, 47% for Cr, and 41% for Pb. These data indicate that As, Cr, Cu and Pb contamination is relatively high in coastal areas of Korea. By contrast, Cd and Zn exhibited lower PI values in all of the sites except for Site I, which is moderately contaminated with Zn (PI ranging from 1.68 to 1.96), indicating that concentrations of Cd and Zn in most of the analyzed samples did not appear to reach pollution threshold levels.

Most heavy metal contamination in the sediments is a mixture of contaminants rather than contamination from one particular metal. The concept of a combined pollution index (CPI) has thus been introduced in many studies to identify multi-element contamination (Chon et al., 1998; Lee, 2003). The CPI of all analyzed samples varied from 0.39 to 3.40. The mean values of CPI, determined from each site, are summarized in Table 5 and spatial distributions are presented in Figure 3. As shown in Figure 3, only sites in the vicinity of the industrial complex (Sites A and C) and the mine district (Site I) had a CPI of >1.0, and were thus considered as moderately contaminated. The coastal industrial complexes and metal mines adjacent to the coast are thus assumed to be the sources of heavy metal contamination in the sediments sampled. The other sites have a CPI of <1.0, which signifies no significant contamination, although some metals tended to be locally enriched (for example, As and Cr at Site G).

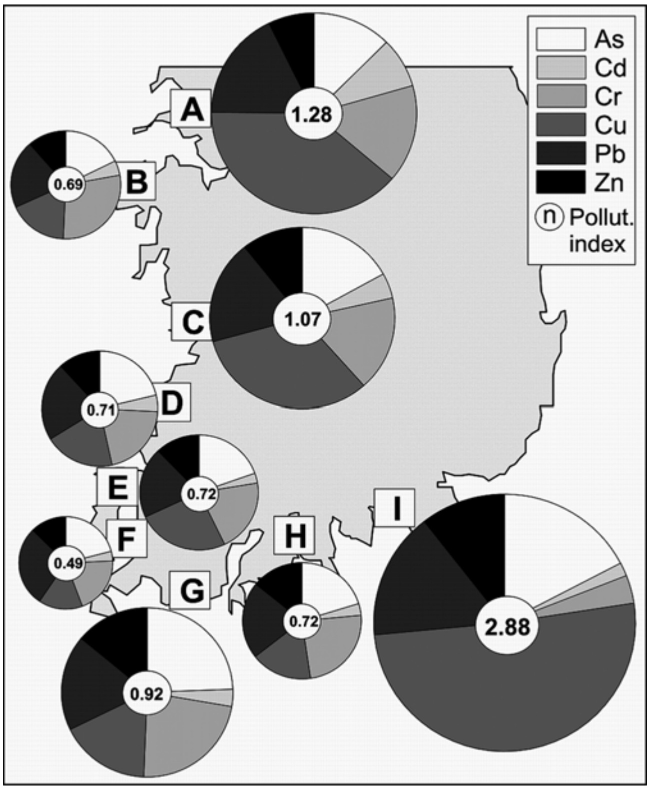

Fig. 3. Spatial distribution of Combined Pollution Index (CPI) in studied sites. The number and pies in circle represent average CPI and contribution fraction of each metal to CPI, respectively.

3.4. Metal Concentrations in Crab

Several studies have examined the usefulness of crustaceans as bioindicators of general metal pollution in aquatic environments (Phillips, 1977; Davies et al., 1981; Rainbow, 1985). The nature of these studies varies according to objectives so they have varying results and recommendations. Some studies have focused on the accumulation of heavy metals in specific organs of the crustacean species studied (e.g., hepatopancreas, gills, gonads, muscles). Measuring the levels of heavy metals in each organ in isolation has certain advantages: it avoids confusion with gut content analysis results (Chapman, 1985), increases understanding of the target tissues and metabolism for toxicants, and provides insights into potential toxicities of biota and how this relates to human health via the food chain. In this study, however, the metal concentrations were determined using whole crabs, since the species used in this study is small (carapace width of adult ≤ 20 mm) and is generally consumed whole without discarding any part.

The concentration of heavy metals in marine organisms may be influenced by the size and gender of the individual. Other investigators have found significant correlations between the metal content and the size of fish (Monteiro and Lopes, 1990; Canli and Atli, 2003; Kojadinovic et al., 2007). However, our results indicate that the metal content is not significantly correlated with the size of the crab ($p > 0.05$) as shown in Table 6. We also found that, due to molting, there was no significant correlation between metal content and the size of crab. Crabs are known to accumulate high levels of metals in their exoskeleton and depurate them through molting (Keteles and Fleeger, 2001; Reinecke et al., 2003; Bergey and Weis, 2007; Reichmuth et al., 2010). In contrast, significant differences were found between the mean concentration of some metals in male and female crabs (Table 6). Most non-essential metals, such as As, Cd and Pb, were significantly higher in females than in males ($p = 0.010$, 0.024 and 0.035, respectively). Chromium and Ni also tended to be slightly higher in females than in males, although this finding is not statistically significant ($p = 0.060$ and 0.153, respectively). On the contrary, essential metals such as Cu and Zn did not show any significant difference with gender ($p = 0.656$ and 0.587, respectively). Several studies have indicated that decapod crustaceans can regulate essential trace metals (Arumugam and Ravindranath, 1983; Rainbow, 1985; Bryan et al., 1986; Rainbow and White, 1989).

Bulk metal concentrations in crab are summarized in Table 7. The crab samples analyzed were limited to male individuals with carapace widths of 12–14 mm, so as to minimize a possible bias due to differences in the size and gender of the crab and to facilitate comparison of subsamples. For comparative purposes, Table 7 lists the US Food

Table 6. Relationships between heavy metal concentration and size and gender of crab

	Influence of size (N = 12)		Influence of gender (N = 10)	
	Correlation	Significance	Difference	Significance
As	r = 0.239	p = 0.454	F > M (t = 4.56)	p = 0.010
Cd	r = 0.246	p = 0.440	F > M (t = 3.54)	p = 0.024
Cr	r = 0.247 (–)	p = 0.439	F > M (t= 2.60)	p = 0.060
Cu	r = 0.118 (–)	p = 0.716	F ≥ M (t = 0.48)	p = 0.656
Ni	r = 0.105 (–)	p = 0.746	F > M (t = 1.76)	p = 0.153
Pb	r = 0.361	p = 0.249	F > M (t = 3.13)	p = 0.035
Zn	r = 0.338	p = 0.282	F ≥ M (t = 0.59)	p = 0.587

Table 7. Heavy metal concentrations (mean ± sd, mg kg^{-1} dry wt.) in crabs from the coastal areas of Korea

Site	As	Cd	Cr	Cu	Ni	Pb	Zn
A	1.16 ± 0.04	0.35 ± 0.31	1.43 ± 0.42	52.07 ± 15.16	2.80 ± 0.05	1.68 ± 0.44	57.36 ± 7.50
B	1.41 ± 0.04	0.10 ± 0.05	3.78 ± 2.24	49.72 ± 4.14	2.73 ± 0.60	1.63 ± 0.50	51.34 ± 1.94
C	1.79 ± 0.75	0.20 ± 0.05	4.11 ± 1.40	53.50 ± 6.18	2.49 ± 0.66	2.47 ± 0.50	43.19 ± 4.80
D	1.97 ± 0.87	0.19 ± 0.09	3.15 ± 0.42	57.46 ± 3.20	2.06 ± 0.33	2.64 ± 0.54	40.08 ± 2.37
E	1.25 ± 0.28	0.09 ± 0.02	2.43 ± 1.02	58.16 ± 16.71	1.71 ± 0.44	2.05 ± 1.20	62.92 ± 27.35
F	0.88 ± 0.37	0.16 ± 0.05	2.08 ± 0.76	39.50 ± 12.84	1.80 ± 0.22	2.33 ± 1.49	55.54 ± 5.31
G	0.89 ± 0.11	0.15 ± 0.07	2.46 ± 0.50	37.01 ± 9.28	2.12 ± 0.33	1.53 ± 0.39	53.50 ± 3.14
H	0.83 ± 0.43	0.11 ± 0.11	1.89 ± 0.77	51.73 ± 13.74	1.76 ± 0.27	1.02 ± 0.30	56.65 ± 16.68
I	0.36 ± 0.38	0.23 ± 0.12	0.51 ± 0.13	88.60 ± 7.61	1.03 ± 0.67	3.24 ± 1.21	64.61 ± 8.93
USFDA[a]	76	3	12	–	70	1.5	–
World[b]	–	–	0.69	70.0	–	1.0	80.0

[a]'Levels of Concern' established for human intake of crustaceans, all concentrations are in mg kg^{-1} wet weight (USFDA, 1993); [b]Mean concentrations of metals reported for decapod crustaceans in other world areas, all concentrations are in mg kg^{-1} dry weight of edible tissue (Bryan, 1976) ; "–" means not available.

and Drug Administration (USFDA) 'Level of Concern' for human intake of crustaceans (1993) and the mean concentrations of metals reported for decapod crustaceans in other parts of the world (Bryan, 1976). Bulk metal concentrations in the crabs were highly variable: As 0.36–2.84, Cd 0.01–0.83, Cr 0.37–6.03, Cu 28.01–98.60, Ni 0.02–3.33, Pb 0.56–4.11 and Zn 38.02–85.11 mg kg^{-1} dry weight. With the exception of Pb, all metal concentrations in crabs were well below the USFDA recommended levels for contamination or human consumption, even though our concentrations are determined using dry weight. In the case of Pb, the mean concentration of Cr was also significantly higher than that of decapod crustaceans in other parts of the world. It is difficult to compare this result with others since bulk concentrations in crab are rarely investigated. The high Pb and Cr in this study may be due to our digestion method, which included the exoskeleton. The concentration sequence of the metals was usually Zn>Cu>Cr, Ni, Pb>As>Cd except Sites C, D and I (where Cu>Zn). The relative concentration levels of Cr, Ni and Pb also varied according to sites. For example, relative concentrations at Sites B, C, G and H were Cr>Ni>Pb, while at Site A, they were Ni>Pb>Cr, and at Site I were Pb>Ni>Cr. The sequence associated with each site was almost similar to that found in the sediments, indicating that metal levels in the crabs reflect those in the sediments of a particular locality.

3.5. Bioaccumulation Factor

The bioaccumulation factor (BAF) for each metal in the crabs, which was linked to results of heavy metal concentration estimates in sediments where the crabs were collected, is summarized in Table 8. BAFs higher than 1.0 were found only for Cd, Cu and Zn, indicating potential biomagnification. Highly variable BAF ranges were also found for different metals: 0.14–5.25 for Cd, 0.35–11.18 for Cu, and 0.25–2.13 for Zn. The BAFs for Cu were higher than 1.0 in all crabs studied except those from Site I. As mentioned above, Site I was highly polluted by an abandoned Cu mine, which is characterized by the highest concentrations of Cu and Zn in both the sediments and the crab. The low BAFs were also characteristic of Site I. It is also noteworthy that the concentrations of Cu and Zn in crabs tended to converge to a narrow range (40–60 mg/kg) as shown in Figure 4. For decapod crustaceans, Cu and Zn are essential for hemocyanin and enzymatic activity, thus, these metals are regulated to specific concentrations by decapod crustaceans (Bryan, 1964, 1968, 1971; White and Rainbow, 1982, 1984; Arumugam and Ravindranath, 1983; Rainbow, 1985; Bryan et al., 1986; Nugegoda and Rainbow, 1989; Rainbow and White, 1989), but once these thresholds are reached, the regulatory process becomes saturated and accumulation begins (Engel and Brouwer, 1987; Rainbow, 1985, 2002; Wang and Rainbow, 2008). This could explain the high concentrations of Cu and Zn in the crabs and their low BAFs observed at Site I. BAFs for Cd were also higher than 1.0 in more than 50% of the crabs studied. Non-essential metals like Cd are not regulated and accumulation can occur at all concentrations (Brouwer and Lee, 2007; Rainbow, 1985). Bondgaard and Bjerregaard (2005) found that molting of the crab *C. maenas* did not reduce Cd levels, which may also explain the high BAF for Cd observed in this study. The other metals displayed very low BAFs, which indicates no biomagnifications. The mean BAFs were found to follow the following order: Cu (3.62 ± 2.73) > Cd (1.28 ± 1.07) > Zn (0.78 ± 0.46) > As (0.17 ± 0.09) > Ni (0.13 ± 0.09) > Pb (0.06 ± 0.03) Cr (0.05 ± 0.03).

3.6. Comparison of Metal Concentrations in Sediments and Crabs

Crabs are well known scavengers in tidal flats. They have a wide diet, consuming detritus, sediment, algae, and plant material as well as other crabs and fish (Reichmuth et al., 2009), from which they accumulate metals in different forms and concentrations. The concentrations of metals accumulated in individuals of this population may be more representative of what is present in the sediments rather than what is present in other organisms they would normally consume (Reichmuth et al., 2010). The highly vari-

Table 8. Bioaccumulation factors (mean ± sd) relative to total heavy metal concentrations in the sediments

Site	As	Cd	Cr	Cu	Ni	Pb	Zn
A	0.21 ± 0.02	0.83 ± 0.31	0.03 ± 0.01	4.01 ± 0.94	0.14 ± 0.02	0.08 ± 0.02	0.81 ± 0.12
B	0.32 ± 0.12	0.74 ± 0.12	0.08 ± 0.03	5.86 ± 2.39	0.18 ± 0.02	0.06 ± 0.01	1.05 ± 0.34
C	0.13 ± 0.02	0.95 ± 0.10	0.09 ± 0.01	2.31 ± 0.64	0.14 ± 0.02	0.07 ± 0.01	0.76 ± 0.22
D	0.28 ± 0.08	1.47 ± 0.61	0.06 ± 0.00	3.91 ± 0.52	0.10 ± 0.00	0.11 ± 0.01	0.60 ± 0.07
E	0.20 ± 0.03	0.98 ± 0.16	0.05 ± 0.02	4.16 ± 2.21	0.09 ± 0.03	0.08 ± 0.05	0.88 ± 0.31
F	0.16 ± 0.05	2.61 ± 1.55	0.06 ± 0.02	5.50 ± 1.60	0.16 ± 0.09	0.06 ± 0.04	1.32 ± 0.32
G	0.09 ± 0.01	1.24 ± 0.56	0.04 ± 0.01	2.09 ± 0.61	0.07 ± 0.01	0.05 ± 0.01	0.54 ± 0.05
H	0.13 ± 0.04	1.30 ± 1.62	0.04 ± 0.01	4.96 ± 3.60	0.08 ± 0.03	0.04 ± 0.01	0.94 ± 0.68
I	0.03 ± 0.01	1.28 ± 0.88	0.02 ± 0.01	0.61 ± 0.24	0.01 ± 0.00	0.04 ± 0.01	0.28 ± 0.02
Total mean	0.17 ± 0.09	1.28 ± 1.07	0.05 ± 0.03	3.62 ± 2.73	0.13 ± 0.09	0.06 ± 0.03	0.78 ± 0.46

able diet of crabs makes this group of animals highly suitable as bioindicators for systems where pollution sources are diverse and variable.

The levels of Cd, Cu, Ni and Pb in the crabs are significantly positively correlated ($p \leq 0.05$) with those in the sediments. Corresponding correlation coefficients derived to describe these relationships are provided in Figure 4, although the correlations of Cu and Pb are greatly influenced by Site I, which is highly polluted by metals (As, Cu, Pb and Zn) from a nearby copper mine. Considering only samples from Site I, the Cu and Pb of crabs are positively correlated with Cu and Pb in the sediments ($p = 0.025$ and 0.015, respectively). These positive correlations between metal concentrations in the surface sediments and that found in the crabs, indicate that some of the metals held in the sediment may become available to the crabs. In contrast, statistically significant relationships were not observed for As, Cr and Zn. The acid-extractable fractions of Cr and Zn (<15%) were much lower than those of Cu and Pb (≥44%). This difference may be attributed to different relationships between sediment and crab for different heavy metals.

Another interesting result was the inverse relationship between BAF and exposure concentration. Statistically significant ($p \leq 0.05$) inverse relationships between BAFs and sediment exposure concentrations were observed for As, Cu, Ni and Zn (Fig. 5). Evidence for some inverse rela-

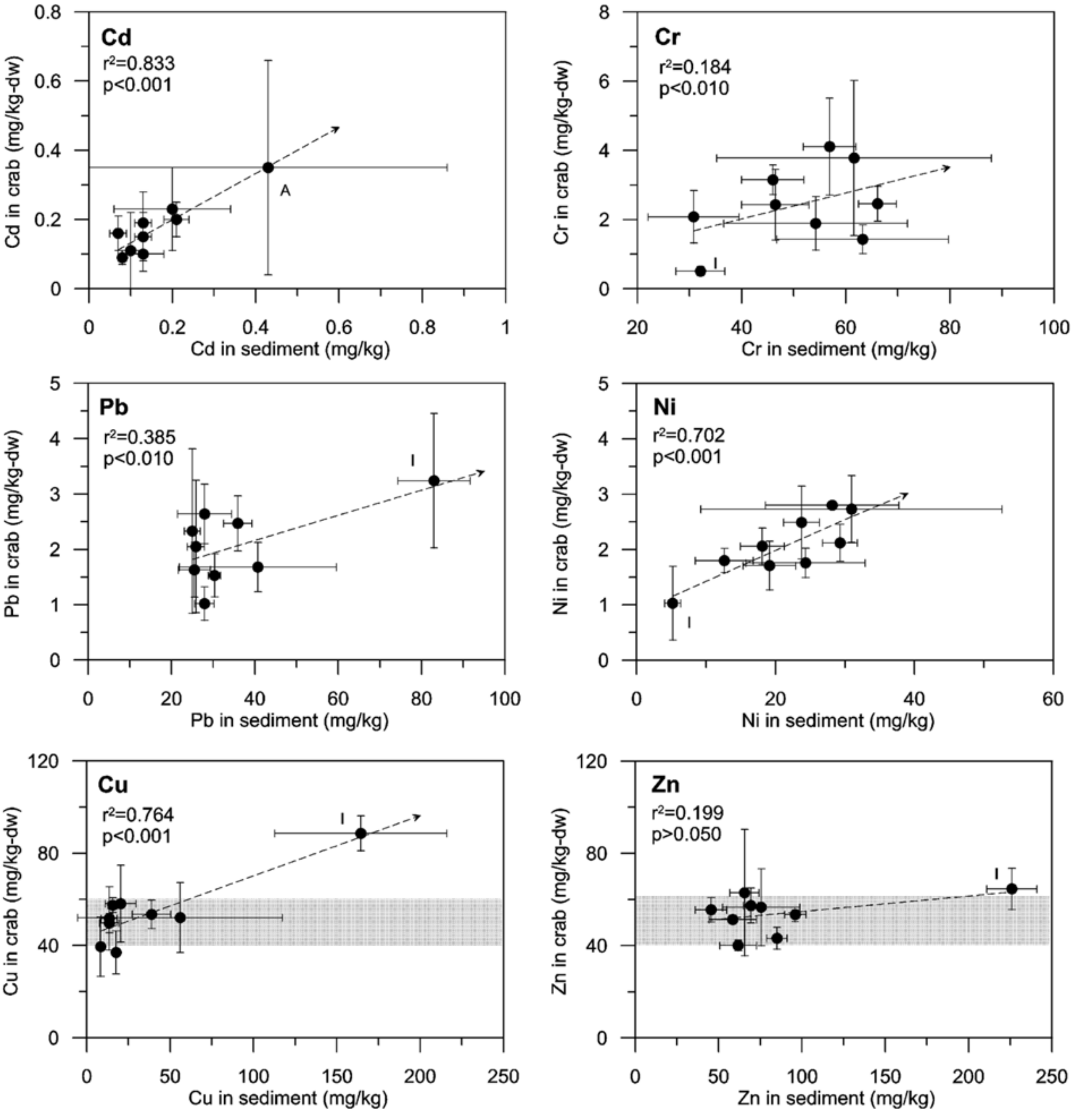

Fig. 4. Relationships between heavy metal contents in crabs and sediments.

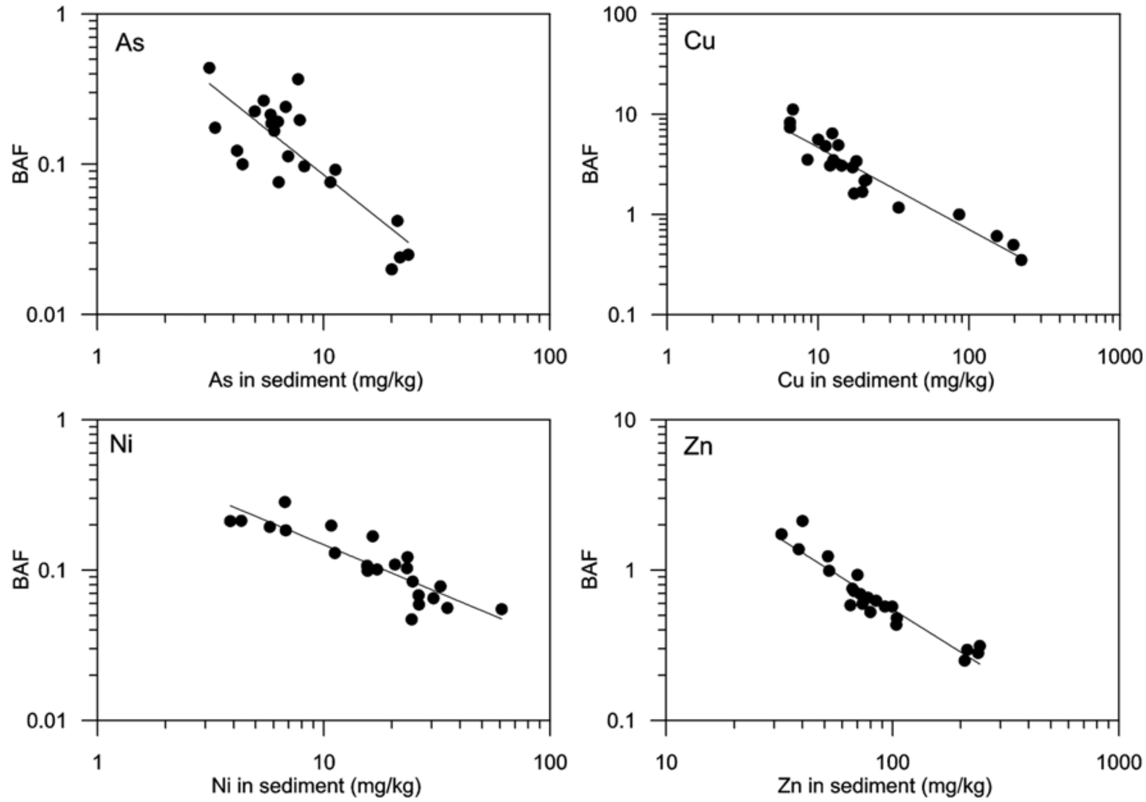

Fig. 5. Relationships between BAFs and exposure concentrations.

tionships between BAFs and exposure concentrations is generally supported in the literature (DeForest et al., 2007; Mcgeer et al., 2003). These inverse relationships between BAFs and exposure concentrations may be a response to multiple mechanisms, including active regulation (e.g., White and Rainbow, 1982) and saturation of uptake kinetics (e.g., Simkiss and Taylor, 1989) at elevated concentrations. In contrast, statistically significant inverse relationships were not observed for Cd ($p = 0.130$), Cr ($p = 0.851$) and Pb (0.066) (data not shown). Of these, Pb was just marginally insignificant. The absence of an inverse relationship between BAF and exposure concentration may be due to a proportional bioaccumulation of metal to its exposure concentration, or to a narrow range of sediment concentrations, which hinder evaluation of whether an inverse relationship may exist (DeForest et al., 2007).

The significance of the inverse relationship between BAFs and exposure concentration, and the inability of these factors to predict hazard potential, has been previously addressed by McGeer et al. (2003). More recently, DeForest et al. (2007) pointed out that the inverse relationships between BAFs and exposure concentrations have important implications for the use of metal bioaccumulation data in site-specific environmental evaluations, such as ecological and human health risk assessments. However, BAF alone cannot be used to express bioaccumulation without consideration of the exposure concentration. Thus consideration of results from a single BAF test is not indicative of the hazard posed by that metal because the highest factors are associated with the lowest exposure concentrations (i.e., the least toxic or hazardous).

4. CONCLUSION

Concentrations of heavy metals in tidal flat sediments varied considerably within and between sites due to variations of pollutant source input. There are significant variations in metal concentrations, with some exceeding the effects-range-low guideline, suggesting that adverse biological effects occasionally occur. The PI values varied greatly for different metals and indicated that As, Cr, Cu and Pb contamination is relatively high in the coastal area of Korea. The combined pollution index (CPI) ranged from 0.49 to 2.88, with the highest CPI in the abandoned metal mine area. Spatial distributions indicated that the abandoned metal mine and the industrial complex are the main sources of metal pollution. Bulk metal concentrations in the crabs showed no significant correlation with the size of crab. In contrast, significant differences were found between the mean concentrations of some metals in male and female crabs; most non-essential metals were significantly higher in females than in males, but essential metals such as Cu and Zn did not show any significant differences associated with gender. The bulk metal concentrations in the crabs were highly variable as indicated by the concentration ranges: As 0.36–2.84, Cd 0.01–0.83, Cr 0.37–6.03, Cu 28.01–98.60, Ni 0.02–3.33, Pb 0.56–4.11 and Zn 38.02–85.11 mg kg^{-1} dry weight. All metal concentrations in the crabs are well below the USFDA recommended levels for contamination or human consumption. The concentrations of most metals in crabs were significantly positively correlated to those in sediments, indicating the usefulness of crabs as bioindicators for metal pollution in tidal flats. The sequence of bioaccumulation factor (BAF) for different metals was in the order of Cu>Cd>Zn>As>Ni>Pb>Cr, and generally tended to be significantly inversely correlated to exposure concentrations, suggesting that the BAF alone is not indicative of the hazards posed by metals because the highest factors are associated with the lowest exposure concentrations.

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