

Distribution of Heavy Metals in the Environmental Samples of the Saemangeum Coastal Area, Korea

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Abstract—In order to understand the distribution of elements in the environmental samples, surface seawater and sediment samples collected from the Saemangeum region of western coast of Korea were analyzed for their concentrations of major and trace elements. Dissolved heavy metal concentrations except for Cd were highest in inner part of the Saemangeum dike and in the Geum River Estuary. Riverine discharge of dissolved metal in July was much higher than that in May, and high in the Mangyeong River and low in the Dongjin River. Dissolved Cd concentration was low in the river waters but high in saline waters, indicating desorption from SPMs with the increase of salinity (ionic strength increase). Because of land-based sources of pollutants, dissolved metals except Cd showed an inverse correlation with salinity. High metal concentrations and metal/Al ratios in the marine sediments were observed in the Geum River Estuary and in the southwestern part of the study area in August. In the surface sediments, TOC and mean grain size (Mz) are controlling factors of metal composition. However, TOC and Mz did not impose any significant effect on the abundance and distribution of Mn, Pb and As.

Keywords: Saemangeum region, environmental change, surface seawater, surface sediment, geochemical components, heavy metals

1. INTRODUCTION

The Yellow Sea is a semi-enclosed sea surrounded by the continent of China and the Korean Peninsula in the mid latitudes of the northwestern Pacific (Kim et al., 2000). The coastline of the Korean Peninsula is characterized by a long stretch of a Ria-type coast in the southern and western part, thus forming numerous inlets whose physiographic features significantly differ from each other (Hong et al., 1991). Because of high tide range, the tidal flats are largely developed along the west coast of Korea. Tidal ranges along the west coast reach a maximum of 10 m. High turbidity is developed by high tidal flow in the west coast. In some places the boundary between land and sea is in the form of abrupt and often spectacular cliffs but, elsewhere, the

boundary can take the form of a complex environment of intertidal sediments. These environments include shingle banks, sandy beaches, mud flats, saltmarsh and mangrove communities. In some cases one or other of these environments will occur, in others they will be associated with one another (Jickells and Rae, 1997). The wetlands along the western coast of the Korean Peninsula are among the largest in the world. They include almost a million hectares of tidal flats in South Korea alone, with North Korea also having many large tracts of coastal wetlands. However, significant environmental issues in the western coast of Korean Peninsula result from reclamation. In Korea, planned reclamation of estuaries, shallow bays, and intertidal mudflats threaten enormous areas of highly productive coastal habitats. Korea government has initiated unprecedented large-scale reclamation projects in its western coasts since 1962 to replace the crop lands lost to human encroachment as well as creating land for constructing industrial complexes and supporting coastal cities (Ahn *et al.*, 1998). Reclamation project of Asan, Shihwa and Saemangeum are examples of the representative land fill in Korea.

There are positive contributions to social and economic development in the reclamation projects of Korea. These massive projects have created land that can be used for constructing residential houses, industrial and agricultural estates, recreational facilities, parks, and the expansion of airport and seaport facilities. However, some negative impacts on the coastal environment were inevitable including the loss of natural coastlines and coastal habitats, adverse effects on fisheries and a decrease in the self-purification capacity of mudflats (Kim and Kahng, 2000). Large-scale reclamation projects have removed mudflats, reducing the natural pollution adsorption capabilities while increasing the pollution loads. Dike construction in estuaries has produced several artificial lakes in the western coast of Korea; however, most of the fresh and brackish lakes have suffered from severe eutrophication and various chemicals pollution because of extreme pollutant discharge. In particular, degradation of water quality in an artificial estuarine lake, Lake Shihwa, becomes a hot issue in Korea. The lake Shihwa is suffering from water and sediment contaminations such as nutrients, COD and heavy metals during the desalination after the dike establishment of 1994. It was a serious environmental threat (Jung *et al.*, 1997; Kim and Khang, 2000). In recent, the importance of tidal flat has been recognized while the tidal flats have been seriously destroyed due to reclamation and landfills.

Despite social concerns, a huge reclamation project in the Saemangeum area of the mid west coast of Korea is proceeding. This project has a total area of 401 km² (land—283 km², lake—118 km²). The Saemangeum dyke of 33 km was completely closed on 21 April 2006. In Korea, this region was one of the most important coastal areas for shellfish aquaculture and fisheries. This area receives a huge amount of fresh water, suspended matter and pollutants from Mangyeong and Dongjin Rivers.

Heavy metals (HMs) are often used without any strict definition. Natural metallic elements (NMEs) are characterized by a high specific gravity exceeding 5, and they are generally known as heavy metals. HMs are present in all environmental compartments, but generally in very small quantities (ppt–ppm): trace metal elements, trace elements, trace metals. Sediments consist as a function of either repository or source. They may have been eroded at the bottom of the sea in a form of sediment or

act as a source where NMEs radiate from the sediments. The amount of heavy metals in the water can be altered by bioaccumulation and biomagnifications; where organism uptakes and the metal contents magnify throughout foodweb. HMs are composed of essential elements such as Fe, Co, Cu and Zn and toxic elements known as Pb, Cd and Hg. And of course the large amount of essential elements can be defined as toxic elements from the biological perspective view. It can not be decomposed or destroyed in environment, meaning they are one of conservative pollutants. The input of metal contaminants into the aquatic system has various sources; and metal sources are divided into two major groups. Firstly, natural sources, which include erosion of ore-bearing rocks, wind-blown dust, volcanic activity and forest fires. Natural sources are based on inherently accommodated metals. Anthropogenic sources are produced from industrialization and urbanization processes; such as fossil fuel burning, waste incineration, industrial complex area, paint, mining etc (Wittmann et al., 1981). In the present, trace metal data of environmental samples such as water, sediment and biota are reliable for further study, however, in the past, trace metal data were unreliable due to lack of instruments and clean techniques for procedure and metal analysis (Windom, 1991; Coquery et al., 2000).

Cho et al. (2001) and Kim et al. (2004) reported the geochemical composition and the processes governing the distribution of elements in surface sediments from the Saemangeum tidal flat. Dissolved heavy metal concentrations in the Mangyeong River estuary where is one of the Saemangeum system were reported. However, some of the dissolved heavy metal data are unreliable (over estimation).

The objectives of this study are to obtain accurate metals in the Saemangeum area and to understand the distribution character of heavy metals in the surface seawater and sediments.

2. MATERIALS AND METHODS

Sampling

To obtain high accuracy of the experiment, each procedure has been undergone thoroughly. In May 2006, seawater samples were collected from 18 different sites including Mangyeong and Dongjin River Estuaries and surface sediments were collected from 16 different sites. In July 2006, seawater samples were collected from 22 sites. In August 2006, sediment samples were collected from 19 sites (Fig. 1). To minimize the contamination due to metal emission from the small boat to the sea, the boat traveled at low speed and minimized the wave movements, and a long pole; holding the sampling bottle is used to collect seawater samples from the moving craft (Boyle et al., 1981). Those samples were filtered through a vacuum filtration system and 0.4 μm PC membrane filters (Whatman Co.), pre-cleaned and pre-weighed, in a Class-100 laminar flow clean bench. To analyze Hg, samples were placed in the acid rinsed 300 mL glass bottle and BrCl solution was added and stored.

The surface sediment samples were collected by a van Veen Grab sampler and top 1 cm layer was taken by a PE spoon, and put into the acid-washed PE jars. The collected samples were immediately frozen with dry ice. Subsamples for grain size analysis were stored in room temperature. In laboratory, these sediment samples for

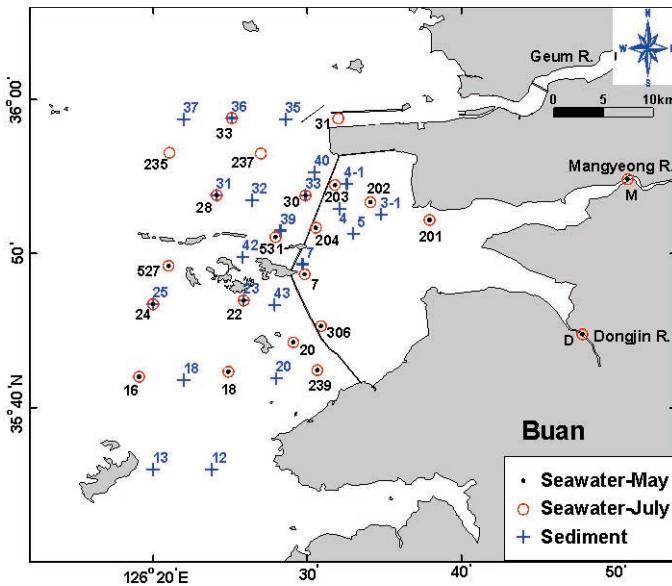


Fig. 1. A map of the study area and sampling sites.

chemical analysis were dried by freeze-dryer (Labconco Freezezone 6), and grounded in an automatic agate mortar (Fritsch Corp. Pulverisette 6).

Analysis

Eight dissolved heavy metals of Fe, Mn, Co, Ni, Zn, Cu, Cd and Pb in the seawater were extracted by a solvent extraction method using a APDC-DDTC-freon (HCFC 131) (Danielsson *et al.*, 1978) and determined with a ICP/MS (Thermo Elemental X-7). Instrument drift and matrix effects during measurement were corrected by using the internal standard of Rh. Hg was determined by using a CVAFS (Tekran 2500).

Sediment samples were size-analyzed by wet sieving (Carver, 1971) and a Sedigraph 5100 after removing organic matter and calcium carbonate. Total organic carbon (TOC) in the sediment samples were analyzed. TOC contents were determined by a C/S analyzer (LECO-SC 444) after eliminating inorganic carbon with 10% HCl. For determination of metals except Hg, approximately 50 mg of dried sediment material was weighed and completely digested in acid-cleaned Teflon bomb (Savillex #561R) by using 2 mL of hydrofluoric acid, 2.5 mL of hydrochloric acid (Merck Suprapur in both cases) and 1 mL of nitric acid (Merck Ultrapur). For digestion process, the bombs were heated at 170°C for 24 hrs in a clean room. After heating, the sample digests were dried and the residues were dissolved with 1% HNO₃ (Windom *et al.*, 1989). Metals were analyzed by ICP/MS (Thermo Elemental X-7). Hg was analyzed by cold vapor atomic fluorescence spectrometry (CVAFS, Tekran

2500) after digestion with 5 mL of 1:1 HNO₃ and HCl on a hot plate for 2 hrs using a Teflon vessel (Saville #561R) (Smith, 1993).

Accuracy of the analytical procedures was assessed by CRM such as CASS-3 for dissolved metals in seawater and the collection percentage was ranged between 89.6% (Mn) and 100% (Co). Reproducibility and accuracy of metal analysis data for sediment were checked by using marine sediment SRM (MESS-2 of NRC, Canada) as a reference. Recoveries of all the metals are ranged from 95.4% for Fe to 108% for Cd. The results indicate good agreement between the certified and the analytical values.

3. RESULTS AND DISCUSSION

Dissolved metals in surface water

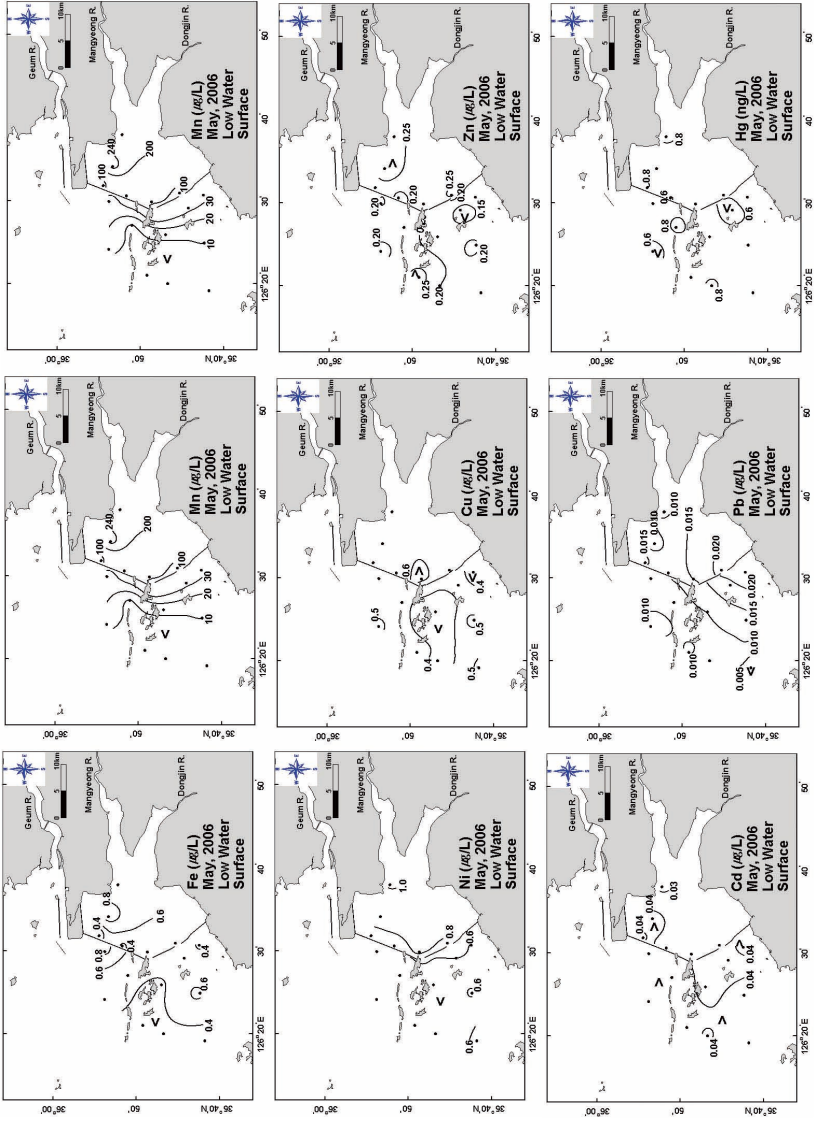
The concentration of dissolved heavy metals (DMs) and acids soluble Hg in the surface seawaters were measured in May and July 2006, ranging from: 0.25–8.87 µg/L for Fe, 1.28–247 µg/L for Mn, 0.018–0.297 µg/L for Co, 0.29–4.19 µg/L for Ni, 0.23–1.52 µg/L for Cu, 0.06–1.66 µg/L for Zn, 0.003–0.048 µg/L for Cd, 0.007–0.053 µg/L for Pb and 0.09–25.8 ng/L for Hg. The average concentrations of Fe, Mn, Co, Ni, Cu, Zn and Cd in May 2006 were higher than those in July 2006 (Table 1).

In May 2006, the highest concentrations of six metals (Fe, Ni, Cu, Zn, Pb and Hg) were found in the Mangyeong or Dongjin River Estuary. However, two elements (Fe and Hg) in July were the highest concentrations in these estuaries. Spatial distributions of 6 dissolved metals (Co, Ni, Cu, Zn, Cd and Pb) in the surface seawaters during the study period showed in Fig. 2. For Ni and Co, decreasing trend of concentrations was shown as the sampling site is further apart from the estuary. Concentration of Cu and Zn are distributed evenly in the Saemangeum area. There is a subtle distinction for Cd; however, it is defined to have increase concentration of Cd as the

Table 1. Summary of dissolved metals concentration range and average in the surface seawaters.

Sampling Time	Fe	Mn	Co	Ni	(µg/L)				Hg* (ng/L)
					Cu	Zn	Cd	Pb	
May, 2006	0.29–8.87 (1.42)	1.28–247 (62.6)	0.018–0.297 (0.130)	0.41–4.19 (0.83)	0.31–1.52 (0.57)	0.14–1.66 (0.29)	0.002–0.048 (0.037)	0.007–0.053 (0.015)	0.49–5.11 (1.04)
July, 2006	0.25–5.91 (1.07)	15.0–232 (61.7)	0.045–0.147 (0.091)	0.29–0.86 (0.53)	0.23–1.18 (0.49)	0.06–0.95 (0.19)	0.003–0.045 (0.024)	0.010–0.051 (0.026)	0.09–25.8 (2.32)

*Acid soluble phase



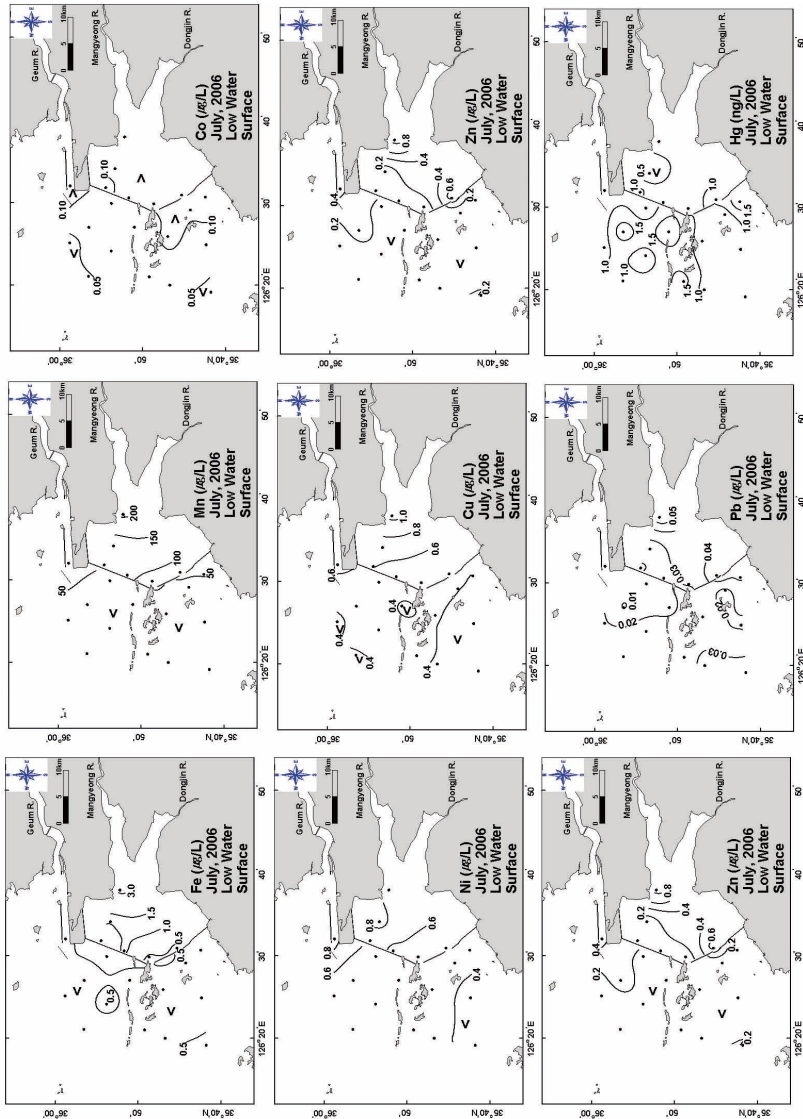


Fig. 2. Distribution of dissolved metals in the surface seawaters of the study area.

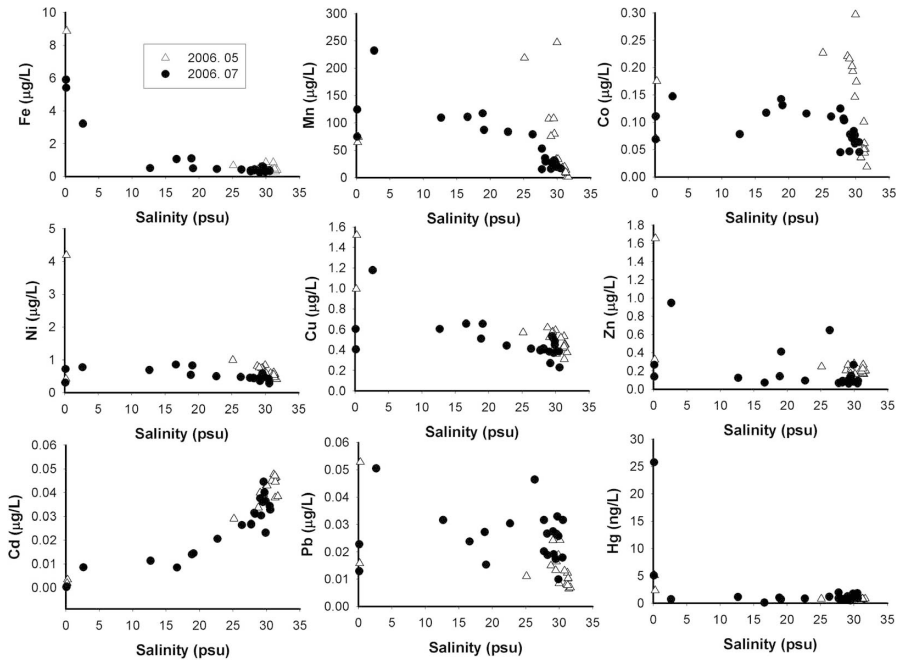


Fig. 3. Relationship between salinity and dissolved metals (Hg: acid-soluble phase) in the surface seawaters.

sampling site is further apart from the estuary. In July, concentration of Ni, Co, Cu and Zn at inner dike was higher than outer sea. And, relevantly high concentrations on exterior region of Geum River Estuary informed that they were from Geum River. Distribution of Cd has a similar trend to May. Pb shows a decreasing trend as the sampling sites were away from the estuary.

The relationship between salinity and DMs (Hg: acid-soluble phase) are illustrated to visualize the concentric alteration of DMs at different salinity. The salinity is strongly dependent on percentage of sodium chloride dissolved in the water. There are 3 main aspects which influence the salinity. Salinity is used to determine physical mixing rate and applied to adsorption and desorption mechanism and used in chemical reactions (Schubel and Kennedy, 1984; Millero *et al.*, 1987; Byrd *et al.*, 1990). Exponential relationship is illustrated by the Fe *vs.* salinity curve. For Mn, Co, Cu and Pb, negative linear relationship was informed. A positive linear relationship was observed for Cd, which allowed the study to understand the effect of desorption from SPM. Furthermore, the data distribution for May was very concentrated in small salinity range (Fig. 3).

The temporal variation from April 2002 to July 2006 for DM was studied. Co showed unstabilized pattern but definitely, increased. Ni was nearly unchanged throughout last four years. Cu, Zn and Pb were not stabilized. The variation of Cd

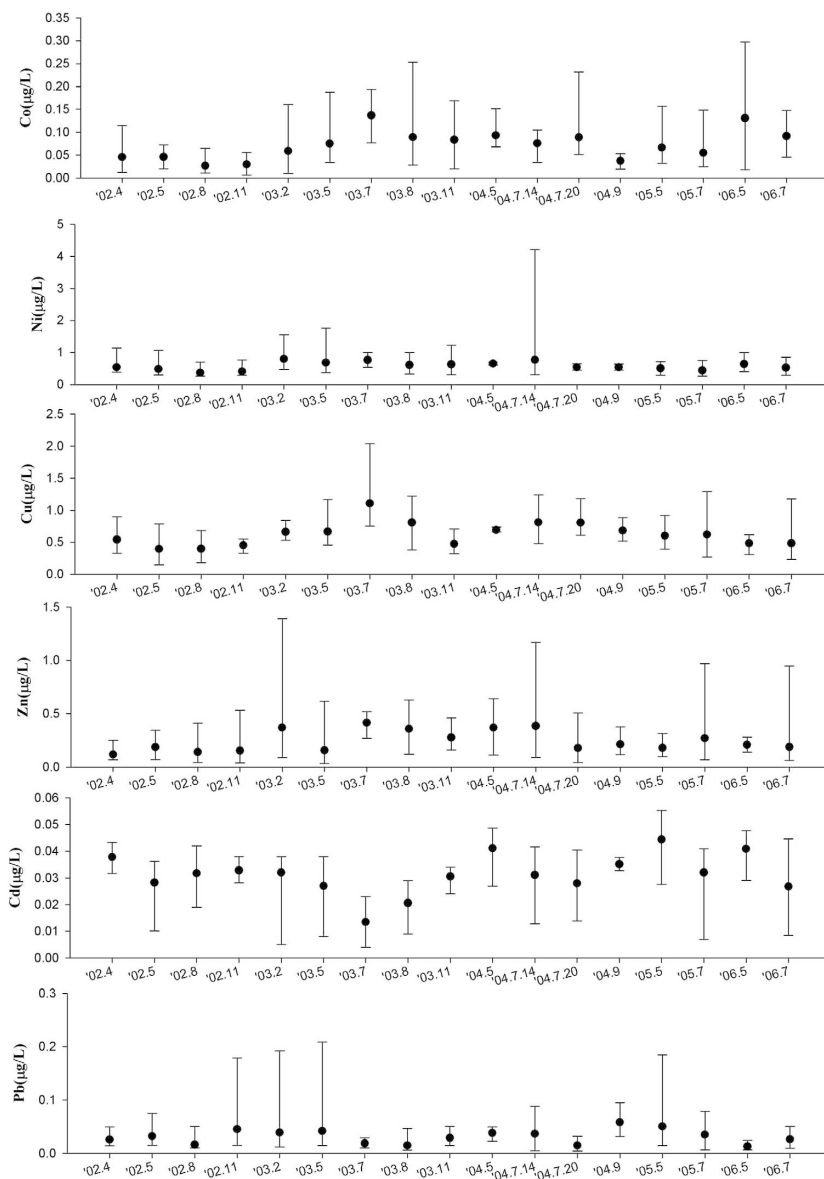


Fig. 4. Temporal variation of metal concentration in the surface seawaters except for the sites at the Mangyeong and Dongjin River Estuaries.

concentration was extremely unstable (Fig. 4). This is due to environmental facts such as weather, season, geographical variations and artificial facts such as reclamation process, industrialization, urbanization and etc.

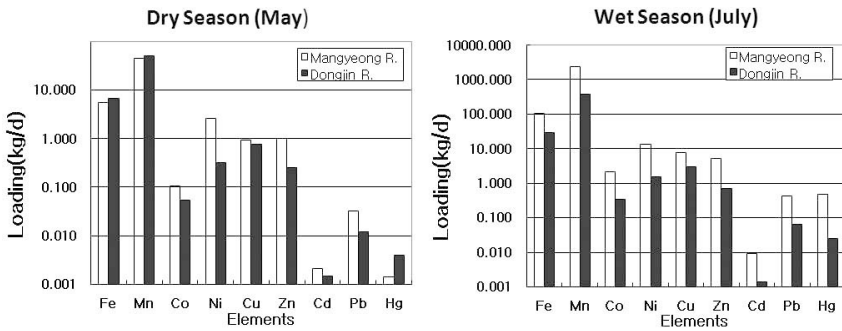


Fig. 5. Dissolved metal fluxes through the Mangyeong and Dongjin River Estuaries.

Concentrations of DMs in the surface seawater in 2006 are compared to Lena River Estuary which represents the natural condition, Bristol Channel and Severn Estuary which represents DMs concentrations in highly polluted region, and past research of Geum River Estuary, Shihwa lake and Saemangeum. A comparison of the DMs concentrations of 1999 Saemangeum research and this study shows that the concentrations of DMs are significantly decreased except the Co and Cd. Their concentrations are roughly half of the previous values. A trend of lower concentrations of DMs of this study is easily observed compare to Bristol Channel and Severn Estuary (Harper, 1991) and Shihwa lake (KORDI, 1999). The higher concentration in Saemangeum compare to Lena River Estuary (Martin *et al.*, 1993) informs that the Saemangeum is more polluted. Cu and Cd concentrations in this study are similar to Geum River Estuary (KORDI, 1996) (Table 2).

Figure 5 showed the dissolved metal flux from Mangyeong and Dongjin Rivers in different seasons. Understanding the influence of weather or season on DMs aided to analyze further research. May is considered as dry season and July is considered as wet season due to continuous rainy season in July. During the dry season, loading of DMs in Mangyeong River were much greater than Dongjin's except some metals including Pb, Mn and Hg. The effect of season; which is wet season, the loading is generally increased by one order. The trend of greater amount of DMs from Mangyeong River than Dongjin River remained the same.

Mean grain size, TOC and metals in surface sediment

Mean grain size, TOC and metal concentrations in the surface sediments of the Saemangeum area are summarized in Table 3.

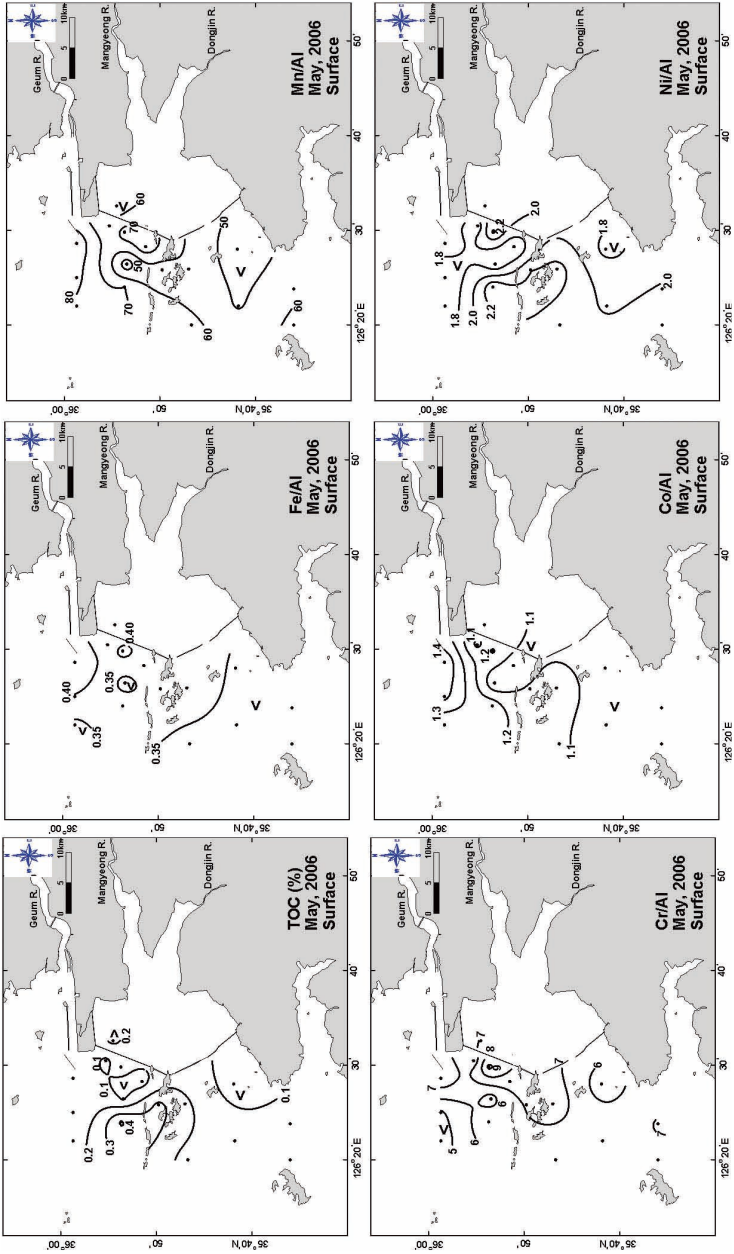
Mean grain size (Mz) in May and August ranged from 2.34–5.04 ϕ (Avg. 3.49 ϕ) and 0.58–6.59 ϕ (Avg. 3.42 ϕ), respectively. During two surveys the lowest Mz (0.58 ϕ) was observed at St. 23 nearby the Gogunsan Islands in August. However, Mz at St. 23 in May had relatively high value (5.04 ϕ). These variations may be caused by change of sedimentation condition according to season and potential difference in

Table 2. Comparison of dissolved metal concentrations in the Saemangeum coastal area and other regions.

Sampling Area	Co	Ni	Cu	Zn	Cd	Pb	Reference
Lena River Estuary	—	0.23–0.49	0.36–0.75	0.25–0.66	0.002–0.023	0.002–0.031	Martin et al. (1993)
Bristol Channel & Severn Estuary	—	—	1.7–4.7	—	0.011–0.140	0.020–10.0	Harper (1991)
Geum River Estuary	—	—	0.68–1.17	0.34–1.69	0.012–0.026	0.015–0.072	KORDI (1996)
Shihwa lake	0.020–0.499	1.19–8.11	1.36–7.32	0.53–11.0	0.014–0.077	0.010–0.093	KORDI (1999)
Saemangeum	0.032–0.188	0.32–1.99	0.50–1.93	0.10–1.89	0.004–0.026	0.10–0.177	KORDI (1999)
Saemangeum	0.018–0.297	0.29–0.99	0.23–1.18	0.06–0.95	0.009–0.048	0.007–0.051	This Study

Table 3. Range and mean of mean grain size, TOC and metals in the surface sediments.

Sampling Time	Mz (ϕ)	TOC	Al		Fe	Mn	Cr	Co	Ni	Metals				Hg (ng/g)
			%							($\mu\text{g/g}$)				
May, 2006	2.34–	0.06–	4.48–	1.67–	221–	21.7–	5.08–	7.26–	2.99–	25.7–	3.18–	0.05–	18.6–	1.26–
	5.04	0.41	6.15	2.24	426	50.3	7.25	13.6	8.23	44.7	5.80	0.14	28.2	9.62
	(3.49)	(0.16)	(5.40)	(1.96)	(341)	(36.1)	(6.21)	(10.7)	(5.83)	(35.6)	(4.36)	(0.09)	(22.4)	(4.60)
August, 2006	0.58–	0.03–	3.28–	0.46–	211–	7.27–	2.16–	2.87–	1.75–	10.2–	1.73–	0.03–	17.9–	5.12–
	6.59	0.46	6.87	2.69	429	56.5	9.42	20.1	12.8	62.3	6.53	0.11	26.5	19.6
	(0.18)	(0.18)	(5.36)	(1.90)	(304)	(32.2)	(6.28)	(10.9)	(5.95)	(36.0)	(4.57)	(0.08)	(22.3)	(9.91)



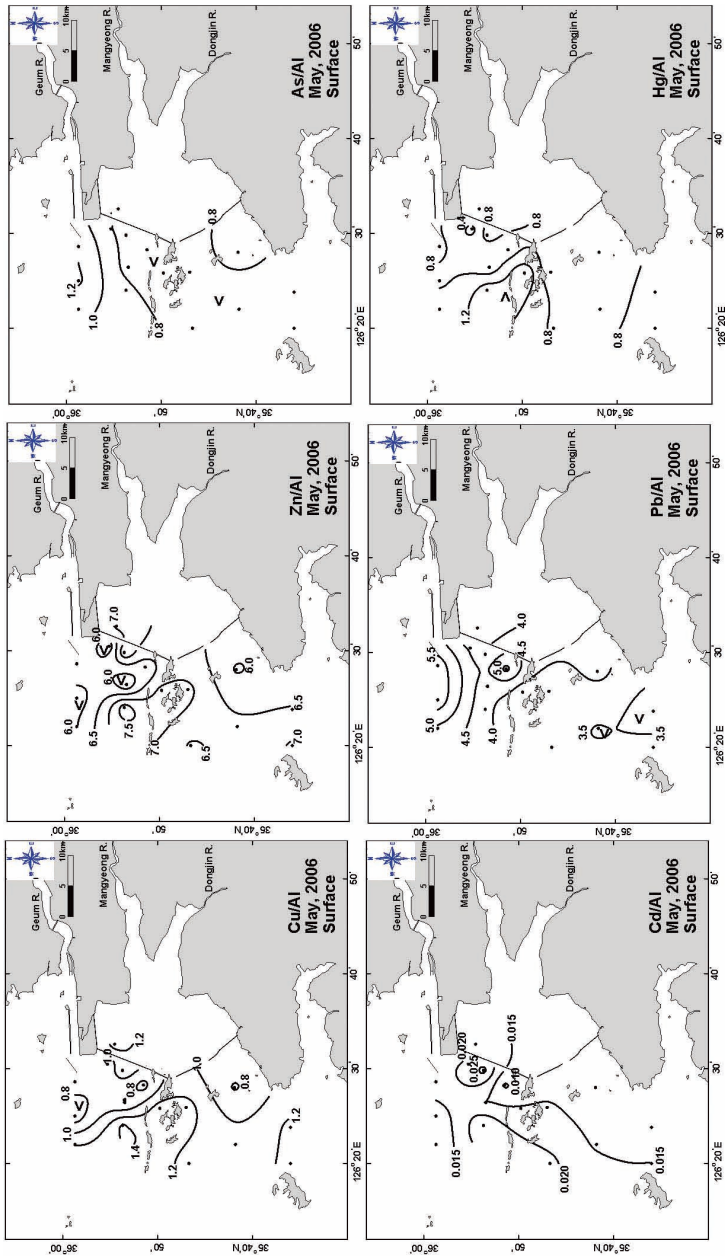
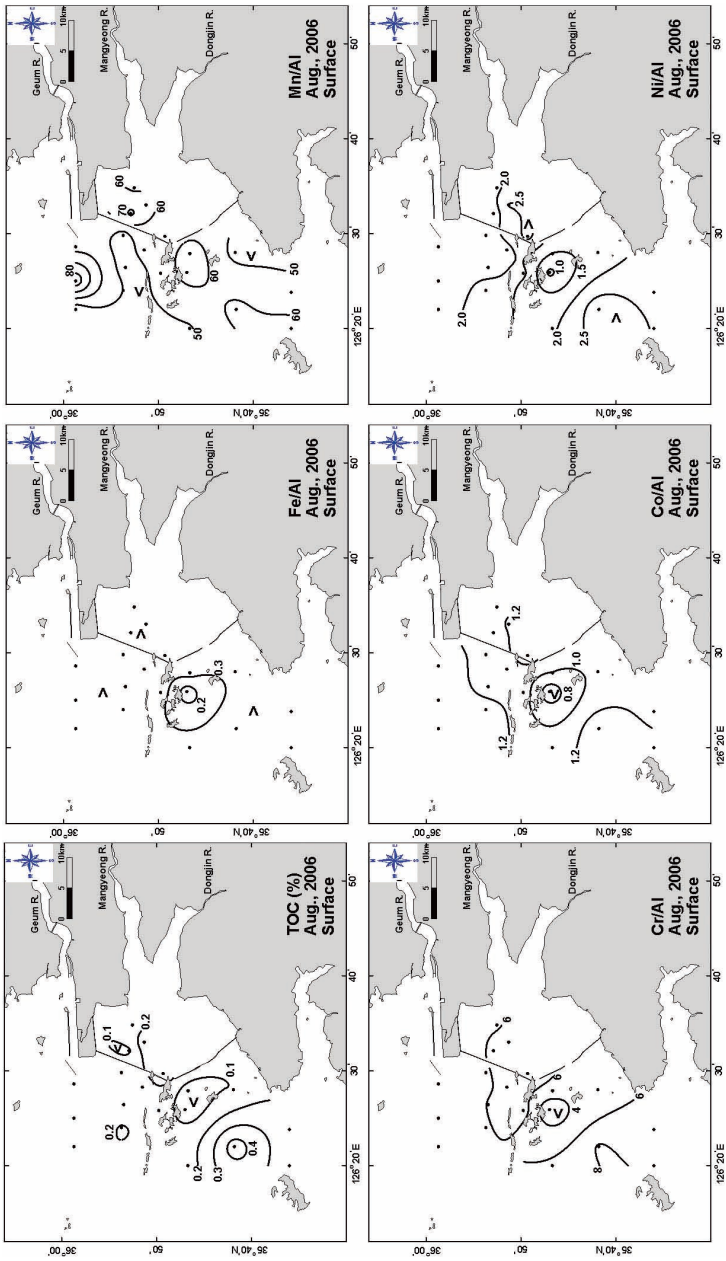


Fig. 6. Distribution of metals in the surface sediments of the study.



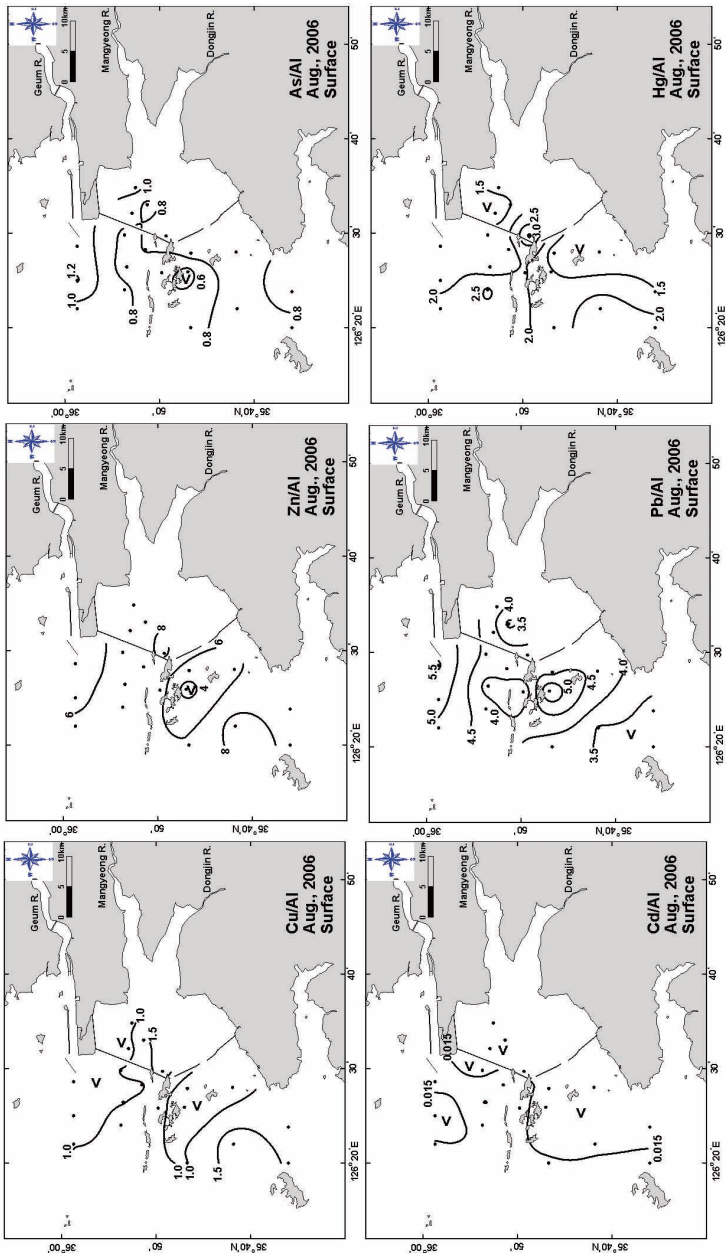


Fig. 6. (continued.)

sampling position. Relatively high Mz during all two surveys was found in the inner part of the dike but low in outer sea.

TOC in the sediments varied from 0.06–0.41% (Avg. 0.16%) in May and 0.03–0.46% (Avg. 0.18%) in August (Table 3). The maximum values were about 30 times higher than the minimum ones. The minimum content (0.03%) was found at St. 23 with the minimum Mz (0.58 ϕ) nearby Gogunsan Islands in August and the maximum (0.46%) at St. 18 with the maximum Mz (6.59 ϕ) in August (Fig. 6). High correlation coefficient ($R = 0.883$) between Mz and TOC in August was observed, but low one ($R = 0.436$) in May. Yang *et al.* (1998) reported close relationship between grain size and organic carbon in the surface sediments of the Yellow Sea. In general, the organic carbon content increases as grain size decreases (Bordovskiy, 1965). Biological productivity in the surface water, chemistry of the water column and sedimentation rate are the most important factors controlling the geographical variation of organic carbon contents (Cho *et al.*, 1999).

The concentrations of chemical elements in the surface sediments were measured in May and August 2006, ranging from: 0.03–0.46% for TOC, 3.28–6.87% for Al, 0.46–2.69% for Fe, 211–429 $\mu\text{g/g}$ for Mn, 7.27–56.5 $\mu\text{g/g}$ for Cr, 2.16–9.42 $\mu\text{g/g}$ for Co, 2.87–20.1 $\mu\text{g/g}$ for Ni, 1.75–12.8 $\mu\text{g/g}$ for Cu, 10.2–62.3 $\mu\text{g/g}$ for Zn, 1.73–6.53 $\mu\text{g/g}$ for As, 0.03–0.14 $\mu\text{g/g}$ for Cd, 17.9–28.2 $\mu\text{g/g}$ for Pb and 1.26–19.6 ng/g for Hg (Table 3).

Al informed the highest value at St. 4-1 in May and at St. 18 in August. In May, most metals including Fe, Cr, Cd, Mn, Co, As and Pb showed the highest concentrations in the sites nearby the Geum River Estuary and the northern part in the offsea of the dyke. In August, the highest concentrations of most metals except Cd, Pb and Hg were found at St. 7 in southern part of outer dyke. In these sediments, variations in chemical compositions are controlled by changes in the river-derived material supply as well as from the source area. Most of the nearshore marine sediments are composed of solid material brought to the sea mainly by the action of rivers (Chester, 1990). The chemical composition of marine sediments also varies primarily depending upon source rock types, climate, composition of constituents, diagenesis, grain size and organic carbon content (Zhao and Yan, 1993). Several kinds of refractory metals such as Al, Fe, Mg, Ti, Sc, Li and Cs have been used to normalize the grain size effect for metal concentrations in sediments (Schropp *et al.*, 1990; Soto-Jiménez and Páez-Osuna, 2001). Some elements such as Al, Cr, Co, Ni, Cu and Zn have reliable positive linear relationship to Mz. But Mn, As, Cd and Pb curves had no relevance. Irrelevance is caused by high metal concentrations from Geum River Estuary. On the other hand, relationships of TOC and metals generally consisted of positive linear relationship, however Mn, As and Pb were scattered (Fig. 7).

Distribution of metal/Al ratios in sediment are plotted in different sites in this study area. Since the metal to Al ratio (Metal/Al; % for Al and Fe or $\mu\text{g/g}$ for others) is used generally, our study used this ratio to compensate grain size effect for metal concentrations. In May, the high ratio of Mn, Co, As, Cd and Pb were found at Geum River Estuary. Most of elements showed relatively low ratio in southern area of outer dyke (Fig. 6). In August, the high ratio of Mn, As and Pb were found at Geum River Estuary. Low ratios were reported at Gogunsan Islands region.

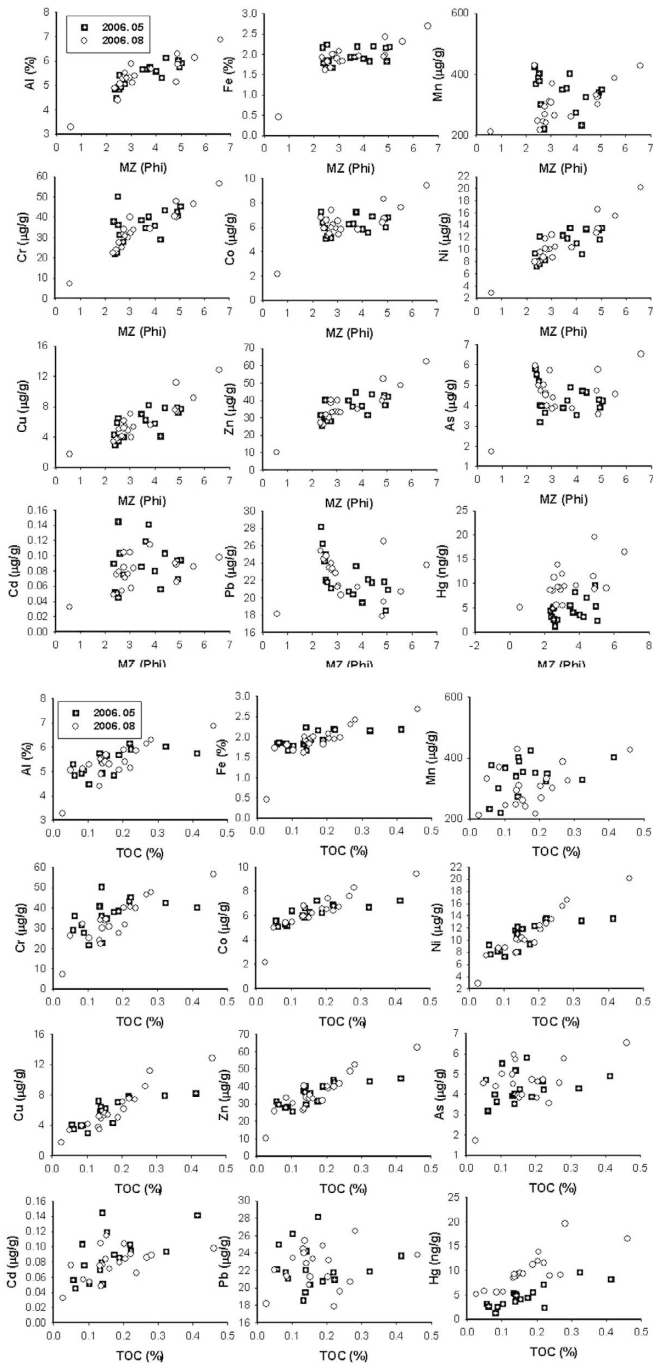


Fig. 7. Plots of Mz vs. metal and TOC vs. metal in the surface sediments of the study area.

Table 4. The average metal concentration and range for each area studied (unit in %, **ng/g, µg/g).

Elements	NOAA SQCs		KORDI (2003)	Cho et al. (2001)	This Study
	ERL	ERM	Sea sediment in 2002	Intertidal F.S. in 1993	Sea sediment in 2006
Al*	—	—	4.93–6.60 (6.05)	4.85–7.90 (6.58)	3.28–6.87 (5.38)
Fe*	—	—	—	1.86–3.88 (2.68)	0.46–2.69 (1.93)
Mn	—	—	299–680 (425)	308–1155 (535)	211–429 (323)
Cr	81	370	23.9–41.6 (35.7)	25–77 (49)	7.27–56.5 (34.7)
Co	—	—	5.86–10.1 (7.11)	6–14 (9)	2.16–9.42 (6.25)
Ni	20.9	51.6	8.94–16.4 (12.7)	10–36 (21)	2.87–20.1 (10.8)
Cu	34	270	4.63–12.2 (7.31)	4–24 (12)	1.75–12.8 (5.89)
Zn	150	410	33.6–58.9 (46.8)	21–71 (43)	10.2–62.3 (35.8)
Cd	1.2	9.6	0.03–0.06 (0.04)	—	0.03–0.14 (0.09)
Pb	46.7	218	18.5–28.5 (21.9)	16–53 (24)	17.9–28.2 (22.4)
As	8.2	70	3.78–7.14 (4.91)	—	1.73–6.53 (4.47)
Hg**	150	710	3.45–18.2 (8.14)	—	1.26–19.6 (7.26)

Sediments act as storage for substances, and if the storage function overtakes its role, that is overloaded, they cause the problems such as pollution affecting preservation. Thus, adequate control is required to manage the level of sediments accumulating. There is no marine sediment quality guideline in Korea, therefore our data were compared to US NOAA guidelines. ERL (Effect range low) and ERM (Effect range median) guidelines for marine sediment were proposed by US NOAA (Long et al., 1995). The ERL and ERM are the 10th and 50th percentiles, respectively, on an ordered list of concentrations in sediment found in the literature that co-occur with any biological effect. All metal concentrations of this study are below ERL. Our metal values in the Saemangeum coastal area in 2006 were compared to those measured in the past for this area (intertidal flat sediment and sea sediment). Our values are quite close to those of the sea sediments in 2002 (KORDI, 2003) but lower than those of intertidal flat sediments collected in 1993 (Cho et al., 2001) (Table 4).

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