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Article

Trace Metals in Surface Seawaters and Mussels around the Dokdo, Korea

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Abstract : Trace metals were investigated for the surface seawaters and mussels collected in adjacent sea to the Dokdo during 1999-2000. This study reports the temporal and spatial distributions of trace metals in seawaters and mussels of the Dokdo coastal areas. Clean technique for trace metal analyses was employed in all manipulations including the sampling and pretreatment procedures. The concentrations of dissolved Cu, Cd and Pb in the surface seawaters were similar to those of the previous data in the East Sea. Contents of particulate Al, Fe, Mn and Co were influenced by input of detrital materials from the Dokdo. The average EFs of particulate metals were to the order of Cd > Zn > Pb > Cu > Ni > Mn > Co > Fe, and the high values in Cd and Zn may be closely related to the preferential uptake of biogenic particles. With the exception of Cd, all metal contents in the mussels were in the same ranges with those from other world coastal areas. The Cd accumulation factor of mussel/seawater in this study was abnormally much higher than previous data from the Korean coasts.

Key words : trace metal, the East Sea (Sea of Japan), Dokdo, seawater, mussel, clean technique.

1. Introduction

The East Sea (Sea of Japan) is a semi-enclosed marginal sea of the North-West Pacific Ocean. The water of this sea is in communication with the East China Sea through the Korea/Tushima Strait, with the main North Pacific Ocean to the east through the Tsugaru Strait, and with the Okhotsk Sea through the Soya Strait and Tartarsky Strait. The surface area is about 1,007,600 km^2 and the mean water depth is 1,685 m. There are three major basins: the Japan basin in the northern half of the sea which is almost 4,000 m deep, and the Ulleung Basin and Yamato Basin to the south which are about 2,200 m deep. The Dokdo is located in the eastern margin of the Ulleung Basin. It consists of 89 rocky islands including the two large islands, Dongdo and Seodo (Hahn 2000).

In general, the main pathways of trace metals into the sea are rivers and atmosphere (Förstner and Wittmann 1981; Duce *et al.* 1991; Gao *et al.* 1992). Since some trace metals are persistently accumulated in sediments and biota in the marine environment, they comprise an essential class of the critical pollutants to be monitored. *Mytilus* species have been widely used as biological indicators to monitor heavy metal pollution (Phillips 1976; Goldberg *et al.* 1978; NAS 1980).

Knowledge on the concentration and distributions of trace metals in marine environment is required in order to understand the biogeochemical behaviors of trave metals and also to assess the fate of anthropogenic inputs. In trace metal analysis, control of contamination during sampling, sample storage and analysis is also necessary.

Employing clean techniques, KORDI (1995a, 1997) reported the dissolved trace metal concentrations in the surface seawaters of the East Sea. In the central East Sea, vertical distributions of dissolved Cu and Ni using a Go-flo sampler were reported (Yang, 1997). On the other hand the assessment of the heavy metal concentrations in the mussels collected from the coastal area of Korea was studied by many scientists (KORDI 1988, 1989, 1990; Choi *et al.* 1992).

This paper reports on the results of trace metal analysis

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in the seawater and mussel collected in adjacent sea to the Dokdo.

2. Materials and methods

Sampling

Surface seawaters (5 stations; Sts. A1-A17) were collected in the adjacent seas to the Dokdo during 2 cruises undertaken by R/V *Onnuri* (Sep. 1999) and *Eardo* (May 2000) of KORDI, and mussels were taken at St. B1 in July 2000 (Fig. 1).

Seawater samples were taken from two PE bottles of 1L hung on a PC plate using nylon rope in the bow of slowly moving ship (< 2 knots per hour) just before arrival to the observation station (Boyle *et al.* 1981). To avoid contamination in the sampling procedure, a vinyl glove was worn, and stopped bottles were enveloped with zip-lock PE bag after recovery. After collection, the samples were filtered through a vacuum filtration system and 0.4 μm PC membrane filters, pre-cleaned and pre-weighed, in a Class-100 laminar flow clean bench on board.

Mussel samples (*Mytilus coruscus*) were hand-collected by scuba diver at 5 m deep of St. B1. Three individuals with *ca.* 10 cm shell length among many samples for the biological study were selected for trace metal analysis and frozen with dry ice in the field. They were not depurated. After transportation to the laboratory, each mussel sample was shucked with a PC knife, weighed and homogenized in a PC blender.

Analytical methods

Six dissolved trace metals of Co, Ni, Zn, Cu, Cd and Pb were extracted by a solvent extraction method using a APDC-DDTC-freon (Danielsson *et al.* 1978) and determined with a GFAAS (Perkin Elmer model, 1100B). Particulate metals were determined with an ICP/MS (VG PQ II+) after dissolution in Teflon bomb (Savillex #561B) using ultrapur grade HNO_3 and HF (Windom and Smith 1991).

Wet mussel samples (*ca.* 2 g) for metal analysis were digested with a microwave digestion system (CEM model, MDS-2100) after addition of Merck ultrapur HNO_3 and H_2O_2 (Mccarthy and Ellis 1991). Sample solutions were analyzed for metals using an ICP/MS and metal contents of mussels were converted to a dry weight basis after freeze-drying (Labconco model, Freezone 6).

To avoid airborne contamination, all procedure of pretreatment was carried out in a Class-100 laminar flow clean bench. Accuracy of the analytical procedures was assessed by SRMs such as CASS-3 for dissolved metals in seawater, MESS-2 for particulate, and SRM-2976 for mussel. The total analytical errors are within 10% of the certified value.

3. Results and discussion

Concentrations of dissolved and particulate metals in September 1999 and May 2000 are shown in Table 1.

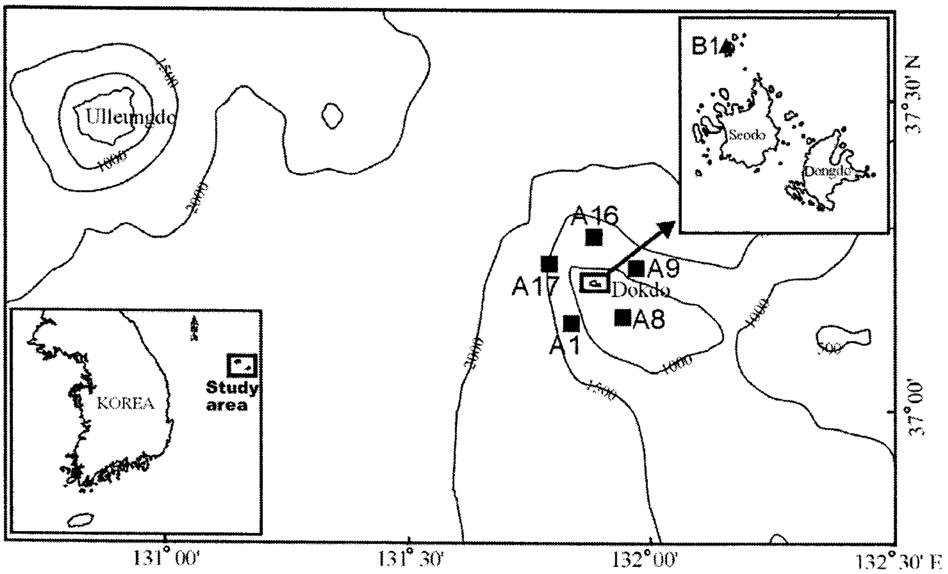

Fig. 1. A location map showing the sampling sites.

Table 1. Dissolved and particulate metals in the surface seawater near the Dokdo.

Sampling time	St.	SPM	Al_P	Fe_P	Mn_P	Co_D	Co_P	Ni_D	Ni_P	Zn_D	Zn_P	Cu_D	Cu_P	Cd_D	Cd_P	Pb_D	Pb_P
		mg/*l*	%	%	µg/g	µg/*l*	µg/g	µg/*l*	µg/g	µg/*l*	µg/g	µg/*l*	µg/g	µg/*l*	µg/g	µg/*l*	µg/g
Sep. 1999	A1	0.26	0.36	0.55	225	0.005	0.51	0.13	6.4	0.21	332	0.14	14.5	0.009	0.13	0.025	8.1
	A8	0.36	0.27	0.09	50	0.005	0.53	0.17	7.0	0.33	17.0	0.17	10.1	0.008	0.05	0.028	4.0
	A9	0.22	0.87	0.22	164	0.006	0.77	0.12	9.6	0.40	42.5	0.12	14.4	0.006	0.57	0.026	10.3
	A16	0.22	2.78	2.32	382	0.004	7.21	0.12	28.5	0.13	115	0.11	34.8	0.007	0.24	0.032	19.9
	A17	0.42	0.62	0.17	79	0.007	0.66	0.19	8.5	0.37	172	0.38	13.8	0.008	0.13	0.031	5.9
May 2000	A1	0.22	1.41	0.51	125	0.002	1.35	0.16	14.0	0.07	61.4	0.08	17.0	0.008	1.17	0.035	18.8
	A8	0.14	1.08	0.38	83	0.004	2.33	0.20	17.5	0.08	80.2	0.11	17.9	0.008	1.56	0.026	18.5
	A9	0.24	4.40	1.11	164	0.004	3.20	0.20	9.6	0.07	29.1	0.10	14.3	0.01	0.44	0.035	12.2
	A16	0.16	1.13	0.41	109	0.003	0.86	0.16	11.6	0.07	25.4	0.10	11.9	0.009	0.92	0.043	16.5
	A17	0.32	1.55	0.46	100	0.006	0.89	1.32*	7.31	0.17	20.8	0.39	8.90	0.008	0.26	0.032	7.6

*suspected of Ni contamination.

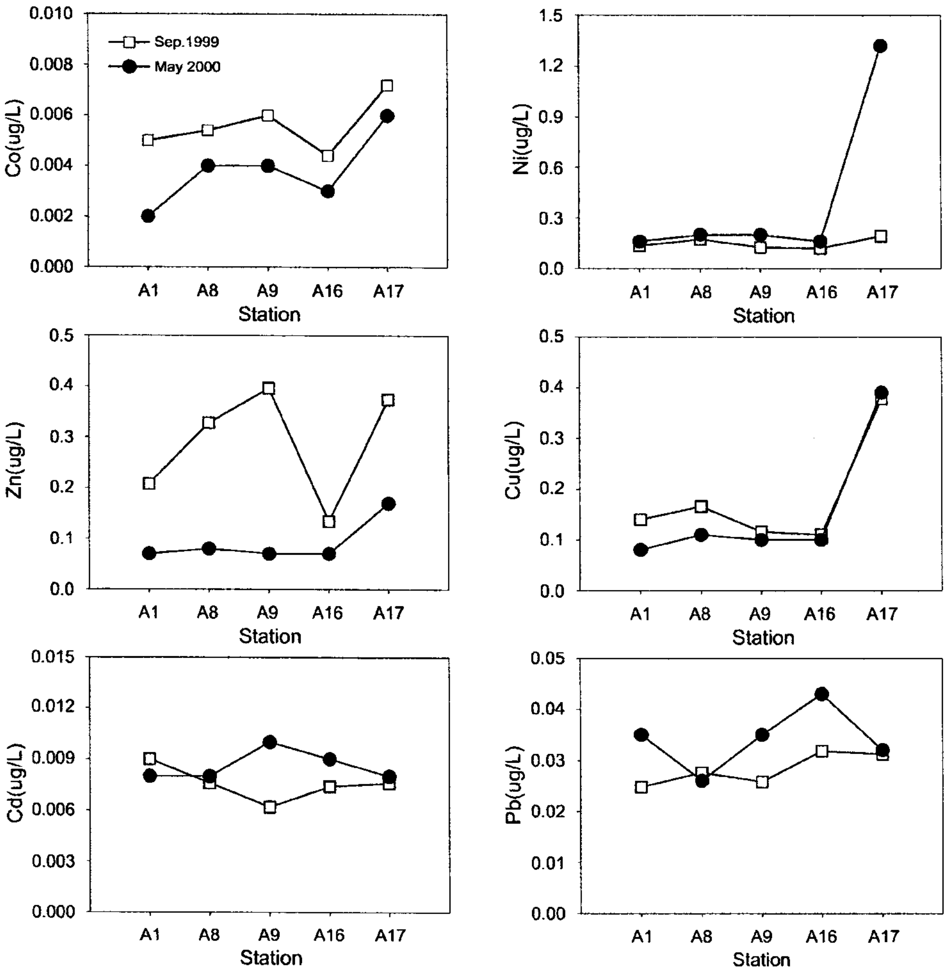

Fig. 2. Distributions of dissolved trace metals in the surface seawater.

Dissolved trace metals in surface seawaters

Concentrations for dissolved Co ranged from 0.002 to 0.007 µg/*l* (avg. 0.005 µg/*l*), for Ni from 0.012 to 1.32 µg/*l* (avg. 0.28 µg/*l*), for Zn from 0.07 to 0.40 µg/*l* (avg. 0.19

Table 2. Comparisons of dissolved metals in this study area and other regions. (unit : $\mu g/l$)

	Co	Ni	Zn	Cu	Cd	Pb	References
Central East Sea	-	(0.274)	-	(0.368)			(1)
South Western East Sea	-	-	0.03-0.13 (0.08)	0.05-0.15 (0.10)	0.004-0.010 (0.006)	0.014-0.042 (0.023)	(2)
Yellow Sea	0.003-0.012 (0.006)	0.26-0.69 (0.45)	0.09-0.51 (0.19)	0.26-0.69 (0.45)	0.010-0.024 (0.016)	0.010-0.024 (0.016)	(3)
North Western Pacific	-	(0.157)	-	(0.043)	(0.012)	-	(4)
Dokdo	0.002-0.007 (0.005)	0.012-1.32 (0.28)	0.07-0.40 (0.19)	0.08-0.39 (0.17)	0.006-0.010 (0.008)	0.025-0.043 (0.031)	This Study

Figures in parenthesis are the averaged values. (1) Yang (1997), (2) KORDI (1997), (3) Kim *et al.* (2001), (4) Murozumi and Nakamura (1981).

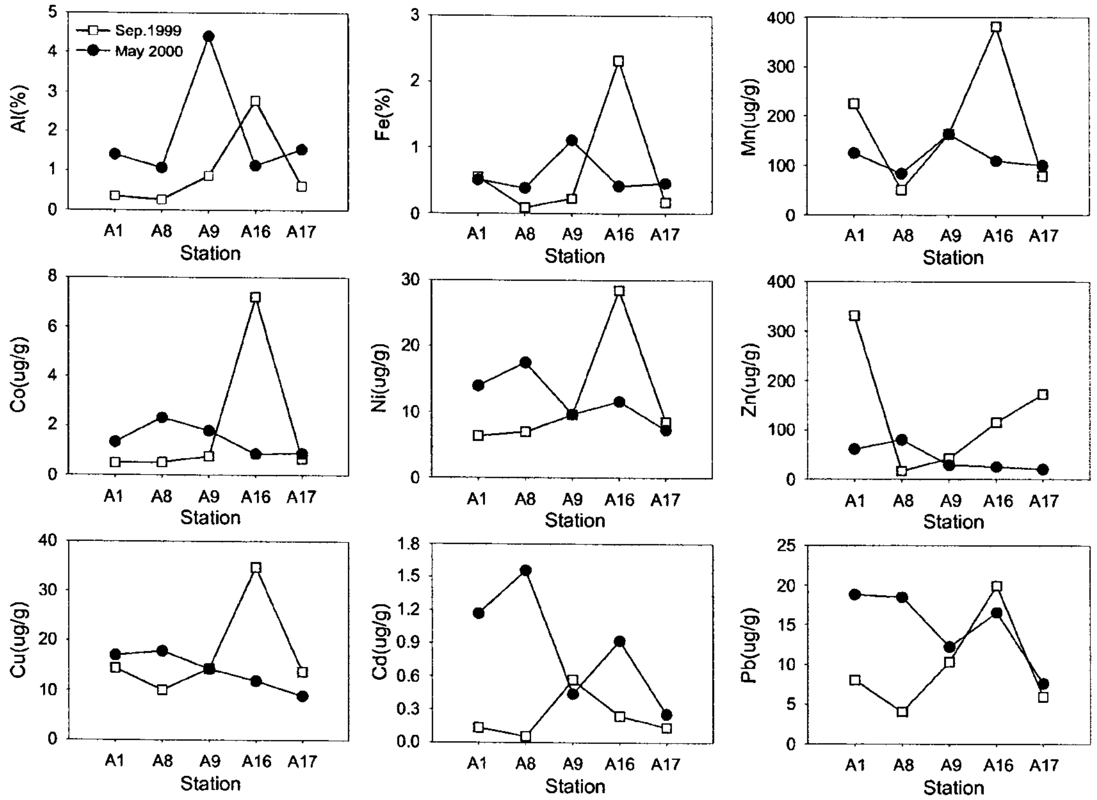

Fig. 3. Distributions of particulate metals in the surface seawarter.

$\mu g/l$), for Cu from 0.08 to 0.39 $\mu g/l$ (avg. 0.17 $\mu g/l$), for Cd from 0.006 to 0.010 $\mu g/l$ (avg. 0.008 $\mu g/l$) and for Pb from 0.025 to 0.043 $\mu g/l$ (avg. 0.031 $\mu g/l$). In September 1999 and May 2000, the highest concentrations of trace metals except for Cd and Pb were found at St. A9 and St. A17. In May 2000, the highest concentrations of some metals including Ni are suspected to be contaminated. So we need more detailed study to clarify these facts. The concentrations of Co, Ni, Cu, Cd and Pb were similar in both seasons, but on the other hand, Zn concentrations in September 1999 were higher than those in May 2000 (Fig. 2).

In Table 2, dissolved trace metal concentrations from this study were compared with those of other seas. The concentrations of Cu, Cd and Pb in this study were similar to those of the KORDI survey. Although the metal concentrations may vary with space and time, concentrations of metals except for Pb were in the same or lower level compared to those the Yellow Sea and the North Western Pacific Ocean.

Particulate metals in surface seawaters

The surface distributions of particulate metals in the study are presented in Fig. 3. The particulate metal contents for Al ranged from 0.27 to 4.40% (avg. 1.45%), for Fe from 0.09 to 2.32% (avg. 0.62%), for Mn from 50.0 to 382 μg/g (avg. 148 μg/g), for Co from 0.51 to 7.21 μg/g (avg. 1.70 μg/g), for Ni from 6.37 to 28.5 μg/g (avg. 12.0 μg/g), for Zn from 17.0 to 332 μg/g (avg. 89.6 μg/g), for Cu from 8.90 to 34.9 μg/g (avg. 15.8 μg/g), for Cd from 0.05 to 1.56 μg/g (avg. 0.55 μg/g) and for Pb from 4.0 to 19.9 μg/g (avg. 12.2 μg/g). In September 1999, the distributions of all the particulate metals except for Zn and Cd showed very similar to one another and the highest contents were found at St. A16. However, the distributions of particulate metal contents in May 2000 were more complicate compared with those obtained from the first cruise. The average contents of Fe, Mn, Co, Zn and Cu in September 1999 were higher than those in May 2000 (Fig. 3).

The particulate metal/total metal ratios for Cu, Ni, Zn, Cd, Pb and Co ranged from 0.2% (Co at St. 5 in May 2000) to 29.6% (Zn at St. A1 in Sep. 1999). The average PM/TM ratios of Cu, Ni, Zn, Cd, Pb and Co during 2 cruises were 2.80%, 1.67%, 11.2%, 1.35%, 8.06% and 7.78%, respectively. According to KORDI (1995b), the average PM/TM ratios for Cu, Ni, Zn, Cd, Pb and Co in the west coast of Korea were 29.5%, 43.9%, 79.2%, 7.8%, 89.6% and 91.6%, respectively. The difference of ratios between two areas might be closely related to the SPM concentrations.

Although there are 5 stations during each cruise, the correlation coefficients among metals are presented in Table 3. There are somewhat different variations with elements and seasons. However, the correlation coefficients among Al, Fe, Mn and Co in the results of both cruises are relatively high. This may be due to input of detrital particles from the Dokdo and show a characteristics in coastal suspended solid (MOMAF 1999).

Enrichment factor (EF) might provide the assessment for contamination or accumulation of metals in SPM, sediment and aerosol. EF was calculated as value that particulate metal content was normalized to the crust abundance relative to the content of Al. That is defined as follows;

$$EF_m = (M_s/Al_s)/(M_c/Al_c)$$

where M_s and Al_s are the particulate content of element M and Al, and M_c and Al_c are content of element M and Al

Table 3. Correlation coefficients(r) among particulate metal contents.

Sep. 1999

	Al	Fe	Mn	Co	Ni	Zn	Cu	Cd	Pb
Al	1.000								
Fe	0.950	1.000							
Mn	0.851	0.928	1.000						
Co	0.981	0.979	0.852	1.000					
Ni	0.994	0.962	0.836	0.995	1.000				
Zn	−0.150	0.074	0.270	−0.107	−0.160	1.000			
Cu	0.983	0.987	0.911	0.985	0.983	0.017	1.000		
Cd	0.240	0.036	0.229	0.070	0.148	−0.331	0.135	1.000	
Pb	0.965	0.944	0.942	0.934	0.944	−0.018	0.967	0.363	1.000

May 2000

	Al	Fe	Mn	Co	Ni	Zn	Cu	Cd	Pb
Al	1.000								
Fe	0.995								
Mn	0.891	0.923	1.000						
Co	0.760	0.763	0.538	1.000					
Ni	−0.430	−0.384	−0.410	0.206	1.000				
Zn	−0.364	−0.326	−0.389	0.269	0.938	1.000			
Cu	−0.026	0.035	0.023	0.512	0.883	0.897	1.000		
Cd	−0.541	−0.492	−0.477	0.064	0.988	0.901	0.824	1.000	
Pb	−0.383	−0.308	−0.179	0.073	0.898	0.756	0.832	0.922	1.000

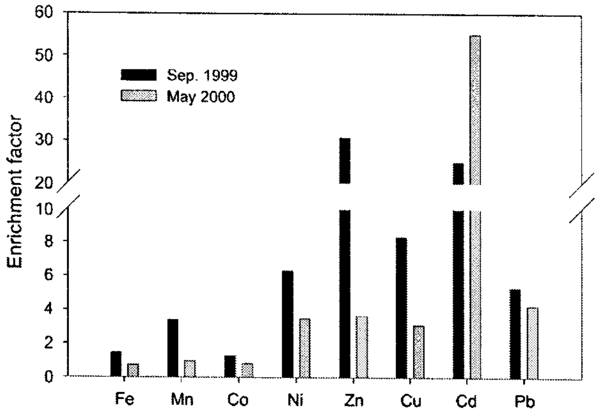

Fig. 4. Enrichment factors of particulate metals in the surface seawater.

in crust. The abundance of element in crust was used in the values of upper crust reported by Taylor and McLennan (1995). The distributions of average values of enrichment factors (EFs) in each season are presented in Fig. 4. In September 1999, the EFs of all metals studied are higher than 1. Especially, the average EFs of Zn and Cd are extremely high as much as 30.8 and 25.0, respectively. In May 2000, the EFs of Fe, Mn and Co are not greater than 1 but other metals are higher than 3. The EF of Cd is 55.1, the highest in this study. The average EFs of metals were to the order of Cd > Zn > Pb > Cu > Ni > Mn > Co > Fe. In the Yellow Sea, the high EFs of Cu, Cr, Cd and Pb were found in the area far away from the shore. Suspended particles in summer were the mixture of organic and inorganic particles (Kim *et al.* 2002). Organic particles which are comprised mainly of phytoplankton and zooplankton tend to scavenge or absorb preferentially trace metals such as Cu and Cd than Al in seawater (Collier and Edmond 1984). Due to organic particles, the high EFs values of Cd were found in the central part of the Yellow Sea (Kim *et al.* 2002). Therefore, the high EFs values of these metals in this study might be closely related to the preferential metal uptake by biogenic particles.

Trace metals in mussels

The range and average of trace metal contents in the mussels are presented in Fig. 5. The contents of Mn in the mussels ranged from 1.43 to 4.73 μg/g (avg. 3.04 μg/g), for Cr from 1.83 to 2.26 μg/g (avg. 1.98 μg/g), for Co from 0.78 to 1.61 μg/g (avg. 1.08 μg/g), for Ni from 2.34 to 2.53 μg/g (avg. 2.50 μg/g), for Zn from 67.3 to 139 μg/g (avg. 97.1 μg/g), for Cu from 4.20 to 6.81 μg/g (avg. 5.28 μg/g), for Cd from 26.9 to 35.9 μg/g (avg. 30.5 μg/g) and for Pb from 2.21 to 3.05 μg/g (avg. 2.65 μg/g). Table 4 shows comparison of trace metal contents in mussels from Korean and other coastal areas of the world oceans. With the exception of high Cd level, all metals were in the same range with those from other world coastal areas.

The Cd contents in the mussels (*ca.* 10 cm shell length) from the Dokdo, however, were over 20 times higher than those (5-8 cm shell length) from the other Korean coastal areas. Riget *et al.* (1996) reported 1.20-2.34 μg/g (avg. 1.75 μg/g) for Cd in mussels(3 cm shell length). When the slope of regression is positive, element contents increase with mussel size, implying that the element is accumulated faster than the mussels grow. Na, Cd, La, Ce and Eu all showed increasing contents with size. For Cd (which is one of the elements most frequently studied with respect to size-dependence of contents), significant positive regression coefficients were noted in 10 of 12 samples from the West Greenland (Riget *et al.* 1996). However, other studies have reported different results for the size dependence of Cd levels. Cossa *et al.* (1980) found a negative correlation of Cd contents with size, while Harris *et al.* (1979) noted a positive correlation. According to Ahn *et al.* (1996), kidney of *Laternula elliptica* from the Antarctica strongly accumulated particularly Cd and Zn. Positive correlations of Cd contents with those of several metals in the kidney, in particular with those of Zn, reflect, in part, binding to metallothionein-like proteins, namely detoxification of the accumulated heavy metals (Walsh 1990). Cd and Pb contents in mussel foot and in the gills depend on their size and age as well as on the metal levels in a particular environment and the exposure time to elevated metal concentrations in the experimental media

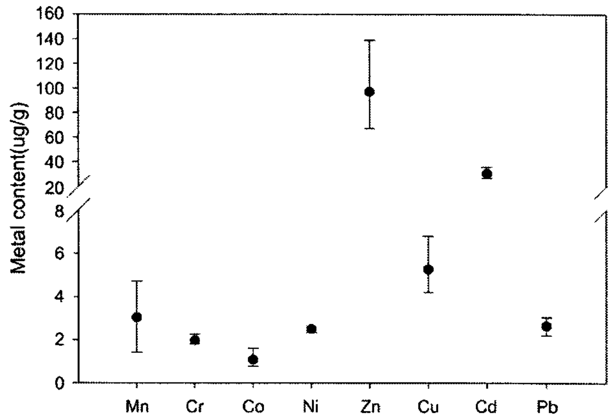

Fig. 5. Range and average of metal contents in the tissues of mussel from the Dokdo.

Table 4. Trace metal conctents in mussels from the Korean and other coastal waters of the world oceans.

(unit :μg/g dry wt.)

	Mn	Cr	Co	Ni	Zn	Cu	Cd	Pb	References
Korean coast	3.8-17.4 (8.7)	0.52-2.19 (1.04)	-	0.8-3.9 (1.6)	72-182 (105)	3.7-7.6 (5.6)	0.62-3.85 (1.26)	0.40-1.97 (1.00)	(1)
East coast, Korea	-	-			(121)	(5.35)	(0.79)	(0.88)	(2)
South coast, Korea					(108)	(5.42)	(0.93)	(0.70)	(2)
West coast, Korea					(90)	(6.62)	(2.30)	(4.44)	(2)
Narragansett Bay, east coast, USA	-	(0.9)	-	(3.36)	(92)	(11)	(1.26)	(2.86)	(3)
Nieuport, Belgium	-	-	-	-	(130)	(8.3)	(0.58)	(2.4)	(4)
Dokdo	(3.04)	(1.98)	(1.08)	(2.50)	(97.1)	(5.28)	(30.5)	(2.65)	This study

Figures in parenthesis are the averaged values. (1) KORDI (1990), (2) Lee *et al.* (1998), (3) NOAA (1987), (4) Meeus-Verdinne *et al.* (1983).

Table 5. Accumulation factors of trace metals in the mussels of the Dokdo and Korean coastal areas.

Element	This study: Dokdo	East*	South*	West*
Co	2.0×10^5	-	-	-
Ni	1.5×10^4	-	-	-
Zn	4.5×10^5	3.5×10^4	8.9×10^4	3.1×10^5
Cu	3.1×10^4	8.8×10^3	1.0×10^4	1.4×10^4
Cd	3.4×10^6	1.5×10^4	4.0×10^4	1.3×10^5
Pb	7.8×10^4	1.2×10^4	1.1×10^4	1.3×10^4

*data from Lee *et al.* (1998).

(Odžak *et al.* 1994). Thus, the high contents of Zn and Cd in this study may result from the preferential uptake.

In Table 5, the metal accumulation factors (AFs) of mussel/seawater from this study were compared with those of the Korean coastal areas. Based on the total and average metal concentrations of surface seawaters found in this study, the AFs was calculated. The AFs of Zn and Pb from this study were more than 5 times than those of the other coasts. Due to relatively low Cd concentration in seawater and high contents of Cd in mussels, the AF (3.4×10^6) of Cd was abnormally much higher than those of the other Korean coasts. This result may be influenced by a response ability on the environmental conditions and size-dependent differences in growth rate.

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