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The State of Marine Pollution in the Waters adjacent to Shipyards in Korea - 3. Evaluation of the Pollution of Heavy Metals in Offshore Surface Sediments around Major Shipyards in Summer 2010

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Abstract : *In order to evaluate the pollution of heavy metals in offshore surface sediments around shipyards in Korea, surface sediment samples were collected at eleven stations around four major shipyards located in the southeastern coast of Korea in summer 2010 and nine kinds of heavy metals such as copper(Cu), zinc(Zn), cadmium(Cd), lead(Pb), chrome(Cr), arsenic(As), mercury(Hg), iron(Fe) and aluminum(Al) in sediments were analyzed. The concentrations of Cu at all sampling stations were in the range of 47.10～414.96 ㎎/㎏ and exceeded TEL(Threshold Effects Level) 20.6 ㎎-Cu/㎏ of Korean marine environmental standards for offshore sediments and ERL(Effect Range-Low) 34.0 ㎎-Cu/㎏. The concentrations of Cu at seven stations around four shipyards were 65.18～414.96 ㎎/㎏ and exceeded PEL(Probable Effects Level) 64.4 ㎎-Cu/㎏ of Korean marine environmental standards for offshore sediments. The concentration of Cu at one station around B-shipyard was 414.96 ㎎/㎏ and exceeded ERM(Effect Range-Median) 270.0 ㎎-Cu/㎏. The concentrations of Zn at all stations were in the range of 135.09～388.79 ㎎/㎏ which exceeded ERL 150.0 ㎎-Zn/㎏. The concentrations of Zn at seven stations around four shipyards were 157.57～388.79 ㎎/㎏ and exceeded PEL 157.0 ㎎-Zn/㎏. The concentration of Zn at one station around B-shipyard was 388.79 ㎎/㎏ and was approaching ERM 410.0 ㎎-Zn/㎏. The concentrations of Cd at all stations were in the range of 0.11～0.54 ㎎/㎏ and were below TEL 0.75 ㎎-Cd/㎏ and ERL 1.2 ㎎-Cd/㎏. The concentrations of Pb at all stations were in the range of 18.04～105.62 ㎎/㎏. The concentrations of Pb at two stations around B-shipyard were 73.87～105.62 ㎎/㎏ which exceeded TEL 44.0 ㎎-Pb/㎏ and ERL 46.7 ㎎-Pb/㎏, and were below PEL 119.0 ㎎-Pb/㎏ and ERM 218.0 ㎎-Pb/㎏. The concentrations of Cr at all stations were in the range of 51.26～85.39 ㎎/㎏. The concentration of Cr at one station around B-shipyard was 85.39 ㎎/㎏ and exceeded ERL 81.0 ㎎-Cr/㎏. The concentrations of As at all stations were in the range of 8.70～22.15 ㎎/㎏ which exceeded ERL 8.2㎎-As/㎏ and were below ERM 70.0 ㎎-As/㎏. The concentrations of As at eight stations around A-shipyard, B-shipyard and D-shipyard were 14.93～22.15 ㎎/㎏ which exceeded TEL 14.5 ㎎-As/㎏ and were below PEL 75.5 ㎎-As/㎏. The concentrations of Hg at all stations were in the range of 0.02～0.35 ㎎/㎏. The concentrations of Hg at three stations around A-shipyard were 0.11～0.13 ㎎/㎏ which were almost equal to TEL 0.11 ㎎-Hg/㎏. Those at two stations around B-shipyard were 0.27～0.35 ㎎/㎏ which exceeded TEL 0.11 ㎎-Hg/㎏ and ERL 0.15 ㎎-Hg/㎏, and were below PEL 0.62 ㎎-Hg/㎏ and ERM 0.71㎎-Hg/㎏. The concentrations of Fe and Al at all stations were in the range of 2.90～3.66 % and 3.12～6.80 %, respectively. These results imply that heavy metals such as copper, zinc, lead, arsenic and mercury were likely to be transferred to marine environment from shipyards, especially from B-shipyard.*

Key Words : *Shipyard, Sediment, Heavy metals, Copper(Cu), Zinc(Zn), Cadmium(Cd), Lead(Pb), Chrome(Cr), Arsenic(As), Mercury(Hg), Iron(Fe), Aluminum(Al)*

1. Introduction

The shipbuilding is a capital-labor-technology intensive industry and is strongly linked to other industries. It has made a great contribution to the national economy development of Korea (KOSHIPA, 2015). As of the end of the year 2009, 178 shipyards of large, medium or small size were located along the southern, eastern and western coasts of the Korean peninsula(Kim, 2010a; Kim, 2010b). Because lots of shipyard works are usually done in the open air in the processes of building, repair and breakup of ships, plenty of various pollutants from shipyards may be discharged into the sea, especially in rainy season. While a large amount of seawater would come in docks such as dry dock and floating dock and be drained away, coastal waters adjacent to shipyards are likely to be polluted. Pollution incidents in shipyards may lead to marine pollution around the shipyards(Kim and Han, 2014).

Marine pollutants have influence on the qualities of seawater and sediments, while settling and depositing on the sea bottom, and being released from the sediments or being suspended again in seawater(Kim, 2007). If various types of heavy metals and noxious chemical substances exist above certain levels of criteria in seawater and in sediments, they may exert an unfavorable

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influence on marine lives and ultimately on human health through the processes of bioaccumulation and biomagnification. Therefore, the current state of marine pollution in the waters adjacent to shipyards of Korea should be surveyed and elucidated for the prevention of pollution from shipyards(Kim and Han, 2014; Kim, 2015).

While water quality is changeable temporally and spatially and shows the short-term condition of water environment, sediment quality can scarcely change temporally and spatially for a short period and may indicate the history of water quality and the long-term environment of sediments(Kawai, 1993; Kim, 2007).

The pollution state of sediments on the sea bottom may usually indicate the degree and the extent of long-term marine pollution(Kim et al., 2012; Kim and Um, 2013). Some indices such as EF(Enrichment Factor), CER(Concentration Enrichment Ratio), I_{geo}(Index of Geoaccumulation) and MPI(Metal Pollution Index), which can be obtained using heavy metals in sediments, have been used for the evaluation of sediment pollution(Kim et al., 2008a; Kim et al., 2012; Kim et al., 2013; Kim and Um, 2013; Kim and Jang, 2014). To evaluate the degree of sediment pollution, the concentrations of heavy metals in sediments can be compared with ERL(Effect Range Low) and ERM(Effect Range Median) proposed by U.S. National Oceanic and Atmospheric Administration(Kim et al., 2008a; Kim et al., 2012; Kim et al., 2013; Kim and Jang, 2014). The heavy metals of sediments can also be compared with SQGs(Sediment Quality Guidelines) established by respective nations for evaluating the effects of trace metals on marine lives(Hwang et al., 2010; Hwang et al., 2011; Kim et al., 2012).

According to Notification No. 2013-186 of Ministry of Oceans and Fisheries in Korea, marine environmental criteria for the sediments on the sea bottom are divided into two parts for eight kinds of heavy metals in sediment, namely, TEL(Threshold Effects Level) for criterion of attention and PEL(Probable Effects Level) for criterion of control. TEL means the maximum concentration of heavy metal which has little negative effect on marine ecosystem. PEL means the minimum concentration of heavy metal that has a very high probability of negative effect on marine ecosystem.

Kim and Han(2014) analyzed the data of pollution incidents occurred in nationwide shipyards of Korea for recent 10 years. Kim(2015) elucidated the current state of pollution of heavy metals in seawater around major shipyards in Korea, because the potential for the discharge of various metals such as copper(Cu), zinc(Zn) and iron(Fe) from shipyards into their surrounding waters always exists in process of shipbuilding. However, it is necessary to evaluate the pollution state of heavy metals in offshore sediments around shipyards in Korea.

In this study, for the purpose of evaluating the pollution state of heavy metals in surface sediments on the bottom of sea around major shipyards in Korea, surface sediment samples were collected at eleven sampling stations around four major shipyards which were located in southeastern area of Korea, and nine kinds of heavy metals in sediments were analyzed. In addition, the concentrations of heavy metals obtained through analysis were compared with criterion indices such as TEL, PEL, ERL and ERM for the evaluation of the pollution of heavy metals in surface sediments around four major shipyards.

The outcomes of this study are expected to contribute to drawing up plans for the prevention of offshore sediment pollution from nationwide shipyards of Korea.

2. Sampling and analysis

2.1 Collection of sediment samples

Sediment samples were collected from the surface (0～2 ㎝) of sea bottom at three sampling stations A1, A2 and A3 around A-shipyard in Ulsan, at two sampling stations B1 and B2 around B-shipyard in Busan, at three sampling stations C1, C2 and C3

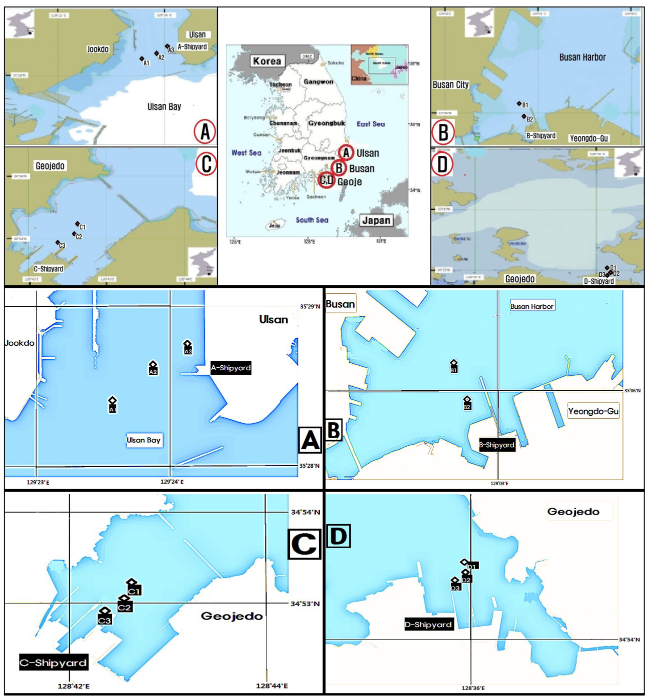

Fig. 1. Sampling stations around four major shipyards in Korea.

around C-shipyard in Geojedo, and at three sampling stations D1, D2 and D3 around D-shipyard in Geojedo for 5 days from August 2 to August 6, 2010, using grab sampler on board the ship(Fig. 1). Sediment samples were stored in the refrigerator on board the ship, transported to the laboratory with an icebox and stored at -20℃ in the freezer until the beginning of analysis.

2.2 Method of analysis

Sediment sample was ground to fine powder after freeze-drying. 0.5～1.0 g of sediment powder was decomposed in a Pyrex conical beaker or Teflon beaker using a mixed acid ($HNO_3 : HClO_4 : HF = 2:1:2$). For the complete evaporation of HF, 2 mL of HNO_3 was added to the completely decomposed sediment powder in the beaker. After adding 20 mL of 2 % HNO_3 solution, the above sediment powder in the beaker was kept warm on hot plate to keep the completely decomposed elements in dissolved condition. 100 mL of the above sample solution was analyzed for copper(Cu), zinc(Zn), cadmium(Cd), lead(Pb), chrome(Cr), arsenic(As), iron(Fe) and aluminum(Al) by Inductively Coupled Plasma Mass Spectrometer (ICP-MS, Perkin Elmer: Elan 6000). Mercury(Hg) was analyzed by Automatic Mercury Analyzer (AMA-254, Milestone). Analytical results of metal concentration in the SRM(MESS-2, PACS-2 (marine sediment)) showed the recovery rates of metals to be 94～98 % and the reliability of analysis to be satisfied.

3. Results and discussion

The analysis results of heavy metals in sediments were shown in Table 1, and the comparisons of analysis results with environmental standards of heavy metals in offshore sediments around four major shipyards were illustrated in Table 2.

Table 1. Analysis results of heavy metals in sediment samples collected around four major shipyards in Korea

Ship yard	St.	Position Lat.(N)	Position Long.(E)	Cu	Zn	Cd	Pb	Cr	As	Hg	Fe	Al
				(㎎/㎏-dw						)	(%)	
A	A1	35°28'25"	129°23'33"	47.10	157.57	0.15	36.86	71.44	14.93	0.13	3.47	6.80
	A2	35°28'38"	129°23'51"	48.05	156.59	0.45	36.23	69.99	18.00	0.11	3.49	6.80
	A3	35°28'46"	129°24'07"	65.18	177.53	0.14	36.82	69.88	21.20	0.11	3.48	6.00
B	B1	35°06'06"	129°02'47"	210.59	256.22	0.36	105.62	64.97	15.67	0.27	2.90	4.90
	B2	35°05'58"	129°02'51"	414.96	388.79	0.53	73.87	85.39	22.15	0.35	3.64	5.84
C	C1	34°53'14"	128°42'36"	70.89	185.15	0.13	29.44	71.15	13.70	0.05	3.66	5.83
	C2	34°53'04"	128°42'34"	73.30	166.02	0.11	24.65	69.27	14.24	0.04	3.63	5.30
	C3	34°52'56"	128°42'22"	93.04	136.14	0.12	18.04	54.85	8.70	0.02	3.13	3.68
D	D1	34°55'03"	128°35'56"	57.68	142.70	0.44	20.34	51.26	15.56	0.03	3.12	3.12
	D2	34°54'57"	128°35'53"	130.30	227.53	0.54	29.75	57.46	19.12	0.05	3.57	4.08
	D3	34°54'50"	128°35'50"	52.61	135.09	0.53	22.38	56.45	15.68	0.03	3.18	3.91

Table 2. Comparisons between marine environmental standards and analysis values of heavy metals in sediments collected at sampling stations around four major shipyards

Heavy metals	Environmental standards(㎎/㎏) Korea TEL (PEL)	Environmental standards(㎎/㎏) NOAA ERL (ERM)	Ship yard	Mean value analyzed (㎎/㎏-dw)	Range of values analyzed (㎎/㎏-dw)
Cu (㎎/㎏)	20.6	34.0	A	53.44	47.10 ~ 65.18
			B	312.78	210.59 ~ 414.96
	(64.4)	(270)	C	79.08	70.89 ~ 93.04
			D	80.20	52.61 ~ 130.30
Zn (㎎/㎏)	68.4	150.0	A	163.90	156.59 ~ 177.53
			B	322.51	256.22 ~ 388.79
	(157)	(410)	C	162.44	136.14 ~ 185.15
			D	168.44	135.09 ~ 227.53
Cd (㎎/㎏)	0.75	1.20	A	0.25	0.14 ~ 0.45
			B	0.45	0.36 ~ 0.53
	(2.72)	(9.6)	C	0.12	0.11 ~ 0.13
			D	0.50	0.44 ~ 0.54
Pb (㎎/㎏)	44.0	46.7	A	36.64	36.23 ~ 36.86
			B	98.75	73.87 ~105.62
	(119)	(218)	C	24.04	18.04 ~ 29.44
			D	24.16	20.34 ~ 29.75
Cr (㎎/㎏)	116.0	81.0	A	70.44	69.88 ~ 71.44
			B	75.18	64.97 ~ 85.39
	(181)	(370)	C	65.09	54.85 ~ 71.15
			D	55.06	51.26 ~ 57.46
As (㎎/㎏)	14.5	8.2	A	18.04	14.93 ~ 21.20
			B	18.91	15.67 ~ 22.15
	(75.5)	(70.0)	C	12.21	8.70 ~ 14.24
			D	16.79	15.56 ~ 19.12
Hg (㎎/㎏)	0.11	0.15	A	0.12	0.11 ~ 0.13
			B	0.31	0.27 ~ 0.35
	(0.62)	(0.71)	C	0.04	0.02 ~ 0.05
			D	0.04	0.03 ~ 0.05
Fe (%)	-	-	A	3.48%	3.48 ~ 3.49%
			B	3.27%	2.90 ~ 3.64%
			C	3.47%	3.13 ~ 3.66%
			D	3.29%	3.12 ~ 3.57%
Al (%)	-	-	A	6.53%	6.00 ~ 6.80%
			B	5.37%	4.90 ~ 5.84%
			C	4.93%	3.68 ~ 5.83%
			D	3.70%	3.12 ~ 4.08%

TEL(Threshold Effects Level) means, as a criterion of attention, the maximum concentration of heavy metal which is expected to have little negative effect on marine ecosystem.

PEL(Probable Effects Level) means, as a criterion of control, the minimum concentration of heavy metal that has a very high probability of negative effect on marine ecosystem.

ERL(Effect Range-Low) means the concentration of heavy metal which may have influence on 10% of benthos statistically.

ERM(Effect Range-Median) means the concentration of heavy metal which may have influence on 50% of benthos statistically.

Remark: The concentration of Li was found to be below 33.1㎎/㎏.

3.1 Copper(Cu)

The concentrations of copper in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 2.

The concentration of Cu in sediment was in the range of 47.10 ㎎/㎏ at station A1 around A-shipyard to 414.96 ㎎/㎏ at station B2 around B-shipyard. The Cu concentrations of 47.10～414.96 ㎎/㎏ around four shipyards were much higher than 11.9～26.6 ㎎/㎏ of Cu on tidal flat sediments in Julpo Bay of Korea(Kim et al., 2008a), 22.0～39.5 ㎎/㎏ of Cu in sediments at Gaduk Channel in Jinhae Bay of Korea(Kim et al., 2008b), 9.2～46.2 ㎎/㎏ of Cu in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012), 3.4～21.30 ㎎/㎏ of Cu in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013), 1.3～31.12 ㎎/㎏ of Cu in the sediment of Gwangyang, Mokpo and Shinan of Korea(Kim and Um, 2013) and 4.53～24.57 ㎎/㎏ of Cu in the bottom sediments of Cheonsu Bay of Korea(Kim and Jang, 2014).

The average concentrations of Cu in sediments were 53.44 ㎎/㎏ at three stations A1, A2 and A3 around A-shipyard in Ulsan, 312.78 ㎎/㎏ at two stations B1 and B2 around B-shipyard in Busan, 79.08 ㎎/㎏ at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 80.20 ㎎/㎏ at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Cu in sediment around shipyard, B-shipyard ranked the first, D-shipyard the second, C-shipyard the third, and A-shipyard the fourth. The average concentration of Cu in sediment around B-shipyard was 3.9～5.9 times higher than those around A-shipyard, C-shipyard and D-shipyard. Kim(2015) showed that the average Cu concentration in seawater around B-shipyard was 1.3～2.0 times higher than those in seawater around A-shipyard, C-shipyard and D-shipyard, and B-shipyard was the highest of all shipyards with regard to the average Cu concentration in seawater. In the average concentrations of Cu around the shipyards, B-shipyard ranked the first in both sediment and seawater.

The range of Cu concentrations in sediment at all stations was 47.10～414.96 ㎎/㎏ and exceeded ERL 34.0 ㎎-Cu/㎏. But the Cu concentration at station B2 around B-shipyard was 414.96 ㎎/㎏ which was above ERM 270.0 ㎎-Cu/㎏. For the concentration criteria of heavy metals in sediments as the sediment quality guidelines, U.S. National Oceanic and Atmospheric Administration or NOAA(1991) proposed ERL(Effect Range-Low) and ERM(Effect Range-Median) which may have an adverse effect on 10 % and 50 %, respectively, of benthos statistically(Long et al., 1995; Kim et al., 2008; Kim and Jang, 2014).

The range of Cu concentrations in sediment at four stations A1, A2, D1 and D3 around A-shipyard and D-shipyard was 47.10～57.68 ㎎/㎏ and exceeded TEL(Threshold Effects Level) 20.6 ㎎-Cu/㎏ of Korean marine environmental standards for offshore sediments. The range of Cu concentrations in sediments at seven stations A3, B1, B2, C1, C2, C3 and D2 around four shipyards was 65.18～414.96 ㎎/㎏ and exceeded PEL(Probable Effects Level) 64.4 ㎎-Cu/㎏ of Korean marine environmental standards for offshore sediments(Fig. 2). This implies that lots of copper was used in anti-fouling paint for ships' hulls(Clark, 1986) and this use inevitably resulted in copper being transferred to the marine environment from shipyards(Kim, 2015), especially from B-shipyard.

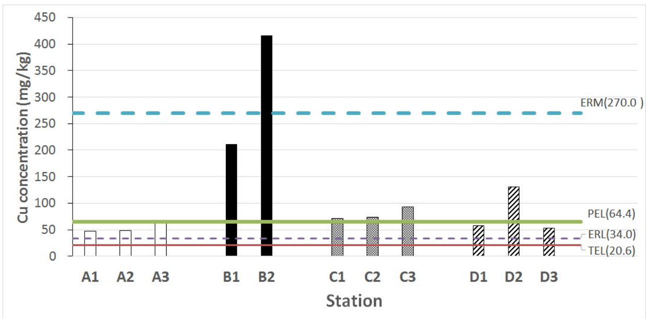

Fig. 2. Comparison of Cu in offshore sediments among stations around four major shipyards.

3.2 Zinc(Zn)

The concentrations of zinc in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 3.

The concentration of Zn in sediment was in the range of 135.09 ㎎/㎏ at station D3 around D-shipyard to 388.79 ㎎/㎏ at station B2 around B-shipyard. The Zn concentrations of 135.09～388.79 ㎎/㎏ around four shipyards were higher than 74.8～130.5 ㎎/㎏ of Zn on tidal flat sediments in Julpo Bay of Korea(Kim et al., 2008a), 122.0～159.0 ㎎/㎏ of Zn in sediments at Gaduk Channel in Jinhae Bay of Korea(Kim et al., 2008b), 94.0～134.0 ㎎/㎏ of Zn in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012), 34～138 ㎎/㎏ of Zn in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013) and 13.53～124.80 ㎎/㎏ of Zn in the sediment of Gwangyang, Mokpo and Shinan of Korea(Kim and Um, 2013).

The average concentrations of Zn in sediments were 163.90 ㎎/㎏ at three stations A1, A2 and A3 around A-shipyard in Ulsan, 322.51 ㎎/㎏ at two stations B1 and B2 around B-shipyard in Busan, 162.44 ㎎/㎏ at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 168.44 ㎎/㎏ at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Zn in sediment around shipyard, B-shipyard ranked the first, D-shipyard the second, A-shipyard the third, and C-shipyard the fourth. The average concentration of Zn in sediment around B-shipyard was 1.9～2.0 times higher than those around A-shipyard, C-shipyard and D-shipyard. According to Kim(2015), the average Zn concentration in seawater around B-shipyard was 1.0～2.5 times higher than those in seawater around A-shipyard, C-shipyard and D-shipyard, and B-shipyard was the highest of all shipyards with regard to the average Zn concentration in seawater. In the average concentrations of Zn around the shipyards, B-shipyard ranked the first in both sediment and seawater.

The range of Zn concentrations in sediment at three stations C3, D1 and D3 around C-shipyard and D-shipyard was 135.09～142.70 ㎎/㎏ which was below ERL 150.0 ㎎-Zn/㎏ but was approaching ERL 150.0 ㎎-Zn/㎏. The range of Zn concentrations at eight stations A1, A2, A3, B1, B2, C1, C2 and D2 around four shipyards was 156.59～388.79 ㎎/㎏ which was above ERL 150.0 ㎎-Zn/㎏ and below ERM 410.0 ㎎-Zn/㎏. The Zn concentration in sediment at station B2 around B-shipyard was 388.79 ㎎/㎏ and was approaching ERM 410.0 ㎎-Zn/㎏. The range of Zn concentrations in sediments at four stations A2, C3, D1 and D3 around A-shipyard, C-shipyard and D-shipyard was 135.09～156.59 ㎎/㎏ and exceeded TEL 68.40 ㎎-Zn/㎏. The range of Zn concentrations in sediments at seven stations A1, A3, B1, B2, C1, C2 and D2 around four shipyards was 157.57～388.79 ㎎/㎏ and exceeded PEL 157.0 ㎎-Zn/㎏(Fig. 3). This suggests that lots of zinc was used in galvanized coatings of metals for ships' hulls(Clark, 1986) and zinc wastes were likely to be discharged to sea from shipyards(Kim, 2015), especially from B-shipyard.

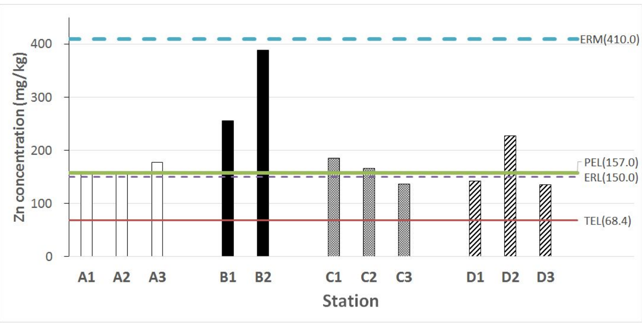

Fig. 3. Comparison of Zn in offshore sediments among stations around four major shipyards.

3.3 Cadmium(Cd)

The concentrations of cadmium in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 4.

The concentration of Cd in sediment was in the range of 0.11 ㎎/㎏ at station C2 around C-shipyard to 0.54 ㎎/㎏ at station D2 around D-shipyard. The Cd concentrations of 0.11～0.54 ㎎/㎏ around four shipyards were higher than 0.05～0.36 ㎎/㎏ of Cd on tidal flat sediments in Julpo Bay of Korea(Kim et al., 2008a), 0.038～0.244 ㎎/㎏ of Cd in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012), 0.028～0.101 ㎎/㎏ of Cd in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013) and ND～0.314 ㎎/㎏ of Cd in the sediment of Gwangyang, Mokpo and Shinan of Korea(Kim and Um, 2013), and were lower than 0.01～1.15 ㎎/㎏ of Cd in the bottom sediments of Cheonsu Bay of Korea(Kim and Jang, 2014).

The average concentrations of Cd in sediments were 0.25 ㎎/㎏ at three stations A1, A2 and A3 around A-shipyard in Ulsan, 0.45 ㎎/㎏ at two stations B1 and B2 around B-shipyard in Busan, 0.12 ㎎/㎏ at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 0.50 ㎎/㎏ at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Cd in sediment around shipyard, D-shipyard ranked the first, B-shipyard the second, A-shipyard the third, and C-shipyard the fourth. The average concentration of Cd in sediment around D-shipyard was 1.1～4.2 times higher than those around A-shipyard, B-shipyard and C-shipyard. Kim(2015) showed that the average Cd concentration in seawater around A-shipyard was 1.4～2.2 times higher than those in seawater around B-shipyard, C-shipyard and D-shipyard. In the average concentrations of Cd around the shipyards, D-shipyard ranked the first in sediment, whereas A-shipyard ranked the first in seawater. According to Kim(2015), the range of Cd concentrations in seawater at all stations around four major shipyards was 0.007～0.031 ㎍/L which was lower than Korean environmental standards of 10 ㎍-Cd/L for the protection of human health and 19 ㎍-Cd/L for short-term protection of marine ecosystem.

The range of Cd concentrations in sediments at all stations around four shipyards was 0.11～0.54 ㎎/㎏ which was below TEL 0.75 ㎎-Cd/㎏ and ERL 1.20 ㎎-Cd/㎏(Fig. 4). This implies that the pollution of Cd in offshore sediment and in seawater around shipyards was not serious.

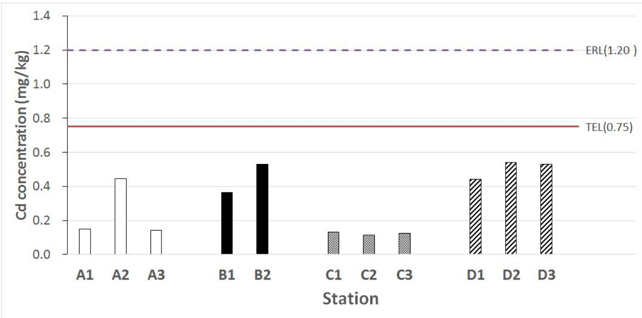

Fig. 4. Comparison of Cd in offshore sediments among stations around four major shipyards.

3.4 Lead(Pb)

The concentrations of lead in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 5.

The concentration of Pb in sediment was in the range of 18.04 ㎎/㎏ at station C3 around C-shipyard to 105.62 ㎎/㎏ at station B1 around B-shipyard. The Pb concentrations of 36.23～36.86 ㎎/㎏ around A-shipyard and 73.87～105.62 ㎎/㎏ around B-shipyard were higher than 25.9～30.7 ㎎/㎏ of Pb in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012), and those of 36.23～36.86 ㎎/㎏ around A-shipyard were similar to 29.7～37.5 ㎎/㎏ of Pb in sediments at Gaduk Channel in Jinhae Bay of Korea(Kim et al., 2008b). However, those of 18.04～29.75 ㎎/㎏ around C-shipyard and D-shipyard were similar to 16.6～24.6 ㎎/㎏ of Pb on tidal flat sediments in Julpo Bay of Korea(Kim et al., 2008a), 15.6～26.1 ㎎/㎏ of Pb in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013), 0.91～29.73 ㎎/㎏ of Pb in the sediment of Gwangyang, Mokpo and Shinan of Korea(Kim and Um, 2013) and 14.50～28.37 ㎎/㎏ of Pb in the bottom sediments of Cheonsu Bay of Korea(Kim and Jang, 2014).

The average concentrations of Pb in sediments were 36.64 ㎎/㎏ at three stations A1, A2 and A3 around A-shipyard in Ulsan, 98.75 ㎎/㎏ at two stations B1 and B2 around B-shipyard in Busan, 24.04 ㎎/㎏ at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 24.16 ㎎/㎏ at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Pb in sediment around shipyard, B-shipyard ranked the first, A-shipyard the second, D-shipyard the third, and C-shipyard the fourth. The average concentration of Pb in sediment around B-shipyard was 2.7～4.1 times higher than those around A-shipyard, C-shipyard and D-shipyard. The average Pb concentration in seawater around D-shipyard was 3.8～18.0 times higher than those in seawater around A-shipyard, B-shipyard and C-shipyard(Kim, 2015). In the average concentrations of Pb around shipyards, B-shipyard ranked the first in sediment, whereas D-shipyard ranked the first in seawater. On the other hand, the Pb concentrations in seawater at all stations around four major shipyards were 0.004～0.334 ㎍/L and were lower than Korean environmental standards of 50 ㎍-Pb/L for the protection of human health and of 7.6 ㎍-Pb/L for short-term protection of marine ecosystem(Kim, 2015).

The range of Pb concentrations in sediments at nine stations A1, A2, A3, C1, C2, C3, D1, D2 and D3 around A-shipyard, C-shipyard and D-shipyard was 18.04～36.86 ㎎/㎏ and was below TEL 44.0 ㎎-Pb/㎏ and ERL 46.7 ㎎-Pb/㎏. The range of Pb concentrations in sediments at two stations B1 and B2 around B-shipyard was 73.87～105.62 ㎎/㎏ which exceeded TEL 44.0 ㎎-Pb/㎏ and ERL 46.7 ㎎-Pb/㎏, but was below PEL 119.0 ㎎-Pb/㎏ and ERM 218.0 ㎎-Pb/㎏(Fig. 5). This suggests that the pollution of Pb in offshore sediment and in seawater around shipyards was not serious except in sediment around B-shipyard.

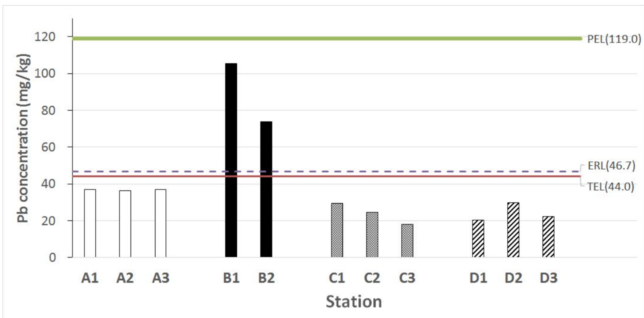

Fig. 5. Comparison of Pb in offshore sediments among stations around four major shipyards.

3.5 Chrome(Cr)

The concentrations of chrome(Cr) in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 6.

The concentration of Cr in sediment was in the range of 51.26 ㎎/㎏ at station D1 around D-shipyard to 85.39 ㎎/㎏ at station B2 around B-shipyard. The Cr concentrations of 51.26～

85.39 ㎎/㎏ around four shipyards were similar to 73.2～83.2 ㎎/㎏ of Cr in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012) and 22.7～93.7 ㎎/㎏ of Cr in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013), and were higher than 43.8～64.0 ㎎/㎏ of Cr on tidal flat sediments in Julpo Bay of Korea(Kim et al., 2008a) and 21.47～65.68 ㎎/㎏ of Cr in the bottom sediments of Cheonsu Bay of Korea(Kim and Jang, 2014).

The average concentrations of Cr in sediments were 70.44 ㎎/㎏ at three stations A1, A2 and A3 around A-shipyard in Ulsan, 75.18 ㎎/㎏ at two stations B1 and B2 around B-shipyard in Busan, 65.09 ㎎/㎏ at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 55.06 ㎎/㎏ at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Cr in sediment around shipyard, B-shipyard ranked the first, A-shipyard the second, C-shipyard the third, and D-shipyard the fourth. The average concentration of Cr in sediment around B-shipyard was 1.1～1.4 times higher than those around A-shipyard, C-shipyard and D-shipyard.

The range of Cr concentrations in sediments at all stations around four shipyards was 51.26～85.39 ㎎/㎏ and was below TEL 116.0 ㎎-Cr/㎏ and ERL 81.0 ㎎-Cr/㎏ with the exception of 85.39 ㎎/㎏ at station B2 around B-shipyard(Fig. 6). This means that the pollution of Cr in offshore sediment around shipyards was not serious except around B-shipyard.

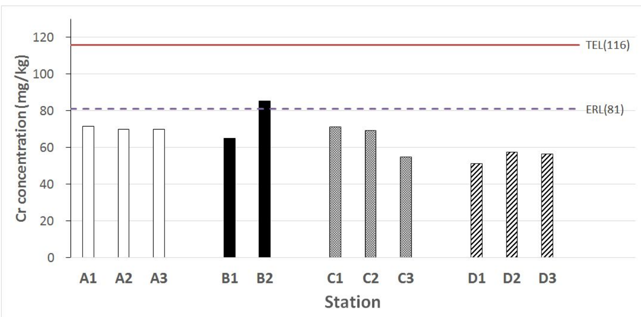

Fig. 6. Comparison of Cr in offshore sediments among stations around four major shipyards.

3.6 Arsenic(As)

The concentrations of arsenic in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 7.

The concentration of As in sediment was in the range of 8.70 ㎎/㎏ at station C3 around C-shipyard to 22.15 ㎎/㎏ at station B2 around B-shipyard. The As concentrations of 8.70～22.15 ㎎/㎏ around four shipyards were higher than 6.6～13.0 ㎎/㎏ of As in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012), 2.7～7.7 ㎎/㎏ of As in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013) and 3.53～8.12 ㎎/㎏ of As in the bottom sediments of Cheonsu Bay of Korea(Kim and Jang, 2014).

The average concentrations of As in sediments were 18.04 ㎎/㎏ at three stations A1, A2 and A3 around A-shipyard in Ulsan, 18.91 ㎎/㎏ at two stations B1 and B2 around B-shipyard in Busan, 12.21 ㎎/㎏ at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 16.79 ㎎/㎏ at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of As in sediment around shipyard, B-shipyard ranked the first, A-shipyard the second, D-shipyard the third, and C-shipyard the fourth. The average concentration of As in sediment around B-shipyard was 1.0～1.5 times higher than those around A-shipyard, C-shipyard and D-shipyard.

The range of As concentrations in sediments at eight sampling stations A1, A2, A3, B1, B2, D1, D2, and D3 around A-shipyard, B-shipyard and D-shipyard was 14.93～22.15 ㎎/㎏ which exceeded TEL 14.5 ㎎-As/㎏ and ERL 8.2 ㎎-As/㎏, but was below PEL 75.5 ㎎-As/㎏ and ERM 70.0 ㎎-As/㎏. The range of As concentrations in sediments at three sampling stations C1, C2 and C3 around C-shipyard was 8.70～14.24 ㎎/㎏ which was below TEL 14.5 ㎎-As/㎏, but was above ERL 8.2 ㎎-As/㎏. The range of As concentrations in sediments at all stations around four shipyards was 8.70～22.15 ㎎/㎏ which was above ERL 8.2 ㎎-As/㎏ and was below ERM 70.0 ㎎-As/㎏(Fig. 7). This indicates that the works of shipyards or the discharges from shipyards had a little effect on the pollution of As in offshore sediment around shipyards.

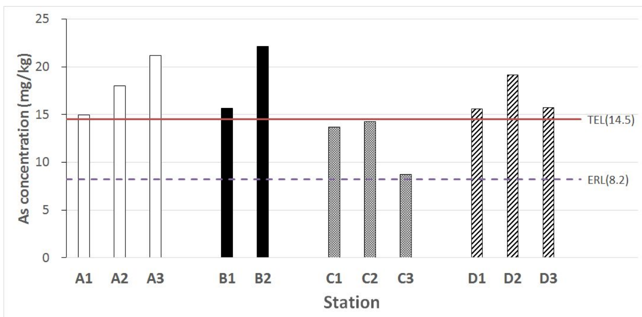

Fig. 7. Comparison of As in offshore sediments among stations around four major shipyards.

3.7 Mercury(Hg)

The concentrations of mercury in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 8.

The concentration of Hg in sediment was in the range of 0.02 ㎎/㎏ at station C3 around C-shipyard to 0.35 ㎎/㎏ at station B2 around B-shipyard. The Hg concentrations of 0.02～0.35 ㎎/㎏ around four shipyards were higher than 0.0217～0.0698 ㎎/㎏ of Hg in sediments at Gaduk Channel in Jinhae Bay of Korea(Kim et al., 2008b), 0.01294～0.03354 ㎎/㎏ of Hg in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012) and 0.006～0.014 ㎎/㎏ of Hg in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013), and were similar to 0.01～0.36 ㎎/㎏ of Hg in the bottom sediments of Cheonsu Bay of Korea(Kim and Jang, 2014).

The average concentrations of Hg in sediments were 0.12 ㎎/㎏ at three stations A1, A2 and A3 around A-shipyard in Ulsan, 0.31 ㎎/㎏ at two stations B1 and B2 around B-shipyard in Busan, 0.04 ㎎/㎏ at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 0.04 ㎎/㎏ at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Hg in sediment around shipyard, B-shipyard ranked the first, A-shipyard the second, and C-shipyard shared the third place with D-shipyard. The average concentration of Hg in sediment around B-shipyard was 2.6～7.8 times higher than those around A-shipyard, C-shipyard and D-shipyard. Kim(2015) showed that the average Hg concentration of 0.004 ㎍/L in seawater around A-shipyard was 2.0 times than the average Hg concentration of 0.002 ㎍/L in seawater around B-shipyard, C-shipyard and D-shipyard. In the average concentrations of Hg around shipyards, B-shipyard ranked the first in sediment, whereas A-shipyard ranked the first in seawater. On the other hand, the Hg concentrations in seawater at all stations were 0.002～0.008 ㎍/L and were lower than Korean environmental standards of 0.5 ㎍-Hg/L for the protection of human health and 1.8 ㎍-Hg/L for short-term protection of marine ecosystem(Kim, 2015).

The concentrations of Hg in sediments at three stations A1, A2 and A3 around A-shipyard were 0.11～0.13 ㎎/㎏ which were almost equal to TEL 0.11 ㎎-Hg/㎏. Those at two stations B1 and B2 around B-shipyard were 0.27～0.35 ㎎/㎏ which were above TEL 0.11 ㎎-Hg/㎏ and ERL 0.15 ㎎-Hg/㎏, and were below PEL 0.62 ㎎-Hg/㎏ and ERM 0.71 ㎎-Hg/㎏. However, the range of Hg concentrations in sediments at six stations C1, C2, C3, D1, D2 and D3 around C-shipyard and D-shipyard was 0.02～0.05 ㎎/㎏ which was below TEL 0.11 ㎎-Hg/㎏ and ERL 0.15 ㎎-Hg/㎏(Fig. 8). This implies that the works of B-shipyard or the discharges from B-shipyard had a little influence on the pollution of Hg in offshore sediment around B-shipyard.

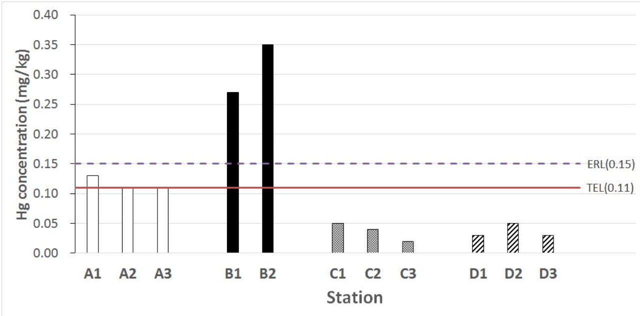

Fig. 8. Comparison of Hg in offshore sediments among stations around four major shipyards.

3.8 Iron(Fe)

The concentrations of iron in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 9.

The concentration of Fe in sediment was in the range of 2.90 % at station B1 around B-shipyard to 3.66 % at station C1 around C-shipyard. The Fe concentrations of 2.90～3.66 % around four shipyards were similar to 2.29～3.61 % of Fe on tidal flat sediments in Julpo Bay of Korea(Kim et al., 2008a) and 1.238～4.181 % of Fe in the sediment of mariculture management area in Ongjin-gun of Korea(Kim et al., 2013), and were lower than 3.45～4.07 % of Fe in the surface sediment of the Gamak Bay of Korea(Kim et al., 2012).

The average concentrations of Fe in sediments were 3.48 % at three stations A1, A2 and A3 around A-shipyard in Ulsan, 3.27 % at two stations B1 and B2 around B-shipyard in Busan, 3.47 % at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 3.29 % at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Fe in sediment around shipyard, A-shipyard ranked the first, C-shipyard the second, D-shipyard the third, and B-shipyard the fourth. The average concentration of Fe in sediment around A-shipyard was 1.0～1.1 times higher than those around B-shipyard, C-shipyard and D-shipyard. The average concentrations of Fe in sediments around four major shipyards were 3.27～3.48 %. The average Fe concentration in seawater around A-shipyard was 1.3～2.2 times higher than those in seawater around B-shipyard, C-shipyard and D-shipyard, and A-shipyard was the highest of all shipyards with regard to the average Fe concentrations in seawater(Kim, 2015). In the average concentrations of Fe around the shipyards, A-shipyard ranked the first in both sediment and seawater.

There is not any criterion index such as ERL, ERM, PEL or TEL in the standards of iron(Fe) for offshore sediments.

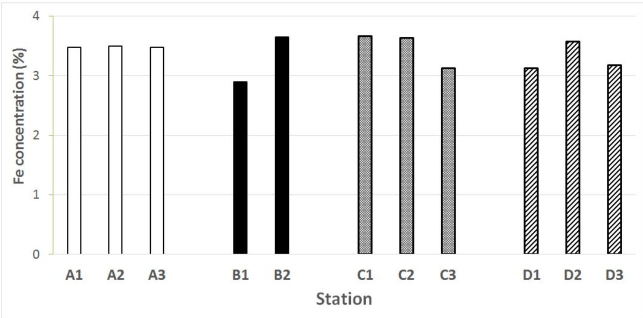

Fig. 9. Comparison of Fe in offshore sediments among stations around four major shipyards.

3.9 Aluminum(Al)

The concentrations of aluminum in sediments at eleven sampling stations around four major shipyards were compared one another, as shown in Fig. 10.

The concentration of Al in sediment was in the range of 3.12 % at station D1 around D-shipyard to 6.80 % at station A1 around A-shipyard. The Al concentrations of 3.12～6.80 % around four shipyards were much lower than 5.93～9.37 % of Al on tidal flat sediments in Julpo Bay of Korea(Kim et al., 2008a), 8.14～9.31 % of Al in sediments at Gaduk Channel in Jinhae Bay of Korea(Kim et al., 2008b) and 3.17～8.12 % of Al in the bottom sediments of Cheonsu Bay of Korea(Kim and Jang, 2014).

The average concentrations of Al in sediments were 6.53 % at three stations A1, A2 and A3 around A-shipyard in Ulsan, 5.37 % at two stations B1 and B2 around B-shipyard in Busan, 4.93 % at three stations C1, C2 and C3 around C-shipyard in Geojedo, and 3.70 % at three stations D1, D2 and D3 around D-shipyard in Geojedo, respectively. With regard to the average concentration of Al in sediment around shipyard, A-shipyard ranked the first, B-shipyard the second, C-shipyard the third, and D-shipyard the fourth. The average concentration of Al in sediment around A-shipyard was 1.2～1.8 times higher than those around B-shipyard, C-shipyard and D-shipyard. The average concentrations of Al in sediments at all stations around four major shipyards were 3.70～6.53 %.

There is not any criterion index such as ERL, ERM, PEL or TEL in the standards of aluminum(Al) for offshore sediments.

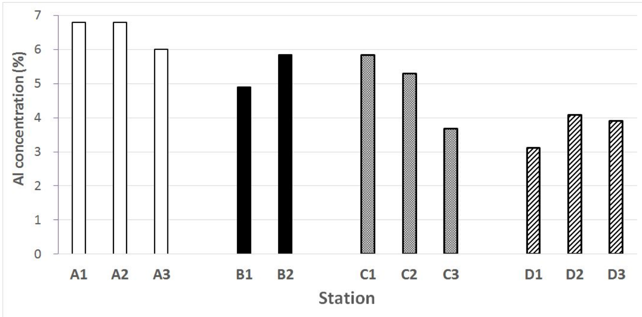

Fig. 10. Comparison of Al in offshore sediments among stations around four major shipyards.

4. Conclusion

The surface sediment samples were collected at eleven offshore sampling stations around four major shipyards located in the southeastern coast of Korea in summer 2010, and nine kinds of heavy metals in sediment samples were analyzed. The evaluation of the pollution of heavy metals in surface sediments around four major shipyards in Korea was summarized as follows.

1) The range of Cu concentrations in sediments at all sampling stations around four major shipyards was 47.10～414.96 ㎎/㎏ which exceeded ERL(Effect Range Low) 34.0 ㎎-Cu/㎏, and the concentration of Cu in sediment at one sampling station around B-shipyard was 414.96 ㎎/㎏ which exceeded ERM(Effect Range Median) 270.0 ㎎-Cu/㎏. The concentrations of Cu in sediments at all sampling stations exceeded TEL(Threshold Effects Level) 20.6 ㎎-Cu/㎏ of Korean marine environmental standards for offshore sediments and those at seven sampling stations were above PEL(Probable Effects Level) 64.4 ㎎-Cu/㎏ of Korean marine environmental standards for offshore sediments. The average concentration of Cu in sediment around B-shipyard was 312.78 ㎎/㎏ and was 3.9～5.9 times higher than those around A-shipyard, C-shipyard and D-shipyard. This implies that lots of copper used and lost in shipyards was transferred to the marine environment from shipyards, especially from B-shipyard.

2) The range of Zn concentrations in sediments at all stations around four shipyards was 135.09～388.79 ㎎/㎏ which exceeded TEL 68.40 ㎎-Zn/㎏. The average concentration of Zn in sediment

around B-shipyard was 322.51 ㎎/㎏ and was 1.9～2.0 times greater than those around A-shipyard, C-shipyard and D-shipyard. The concentrations of Zn in sediments at seven stations were 157.57～388.79 ㎎/㎏ which exceeded PEL 157.0 ㎎-Zn/㎏, and those at eight stations were 156.59～388.79 ㎎/㎏ which exceeded ERL 150.0 ㎎-Zn/㎏. Especially, the concentration of Zn in sediment at station B2 was 388.79 ㎎/㎏ and was approaching ERM 410.0 ㎎-Zn/㎏. This suggests that lots of zinc wastes were likely to be discharged to sea from shipyards, especially, from B-shipyard.

3) The range of Cd concentrations in sediments at all stations around four shipyards was 0.11～0.54 ㎎/㎏ which was below TEL 0.75 ㎎-Cd/㎏ and ERL 1.20 ㎎-Cd/㎏. The average concentration of Cd in sediment around D-shipyard was 0.50 ㎎/㎏ and was 1.1～4.2 times higher than those around A-shipyard, B-shipyard and C-shipyard.

4) The range of Pb concentrations in sediments at all stations around four shipyards was 18.04～105.62 ㎎/㎏. The concentrations of Pb in sediments at two stations around B-shipyard were 73.87～105.62 ㎎/㎏ which exceeded TEL 44.0 ㎎-Pb/㎏ and ERL 46.7 ㎎-Pb/㎏, but did not reach PEL 119.0 ㎎-Pb/㎏ or ERM 218.0 ㎎-Pb/㎏. The average concentration of Pb in sediment around B-shipyard was 98.75 ㎎/㎏ and was 2.7～4.1 times higher than those around A-shipyard, C-shipyard and D-shipyard. This means that the works of B-shipyard or the discharges from B-shipyard had a little influence on the pollution of Pb in offshore sediment around B-shipyard.

5) The range of Cr concentrations in sediments at all stations around four shipyards was 51.26～85.39 ㎎/㎏ which was below TEL 116.0 ㎎-Cr/㎏ and ERL 81.0 ㎎-Cr/㎏. However, the concentration of Cr at one station around B-shipyard was 85.39 ㎎/㎏ and exceeded ERL 81.0 ㎎-Cr/㎏. The average concentration of Cr in sediment around B-shipyard was 75.18 ㎎/㎏ and was 1.1～1.4 times higher than those around A-shipyard, C-shipyard and D-shipyard.

6) The range of As concentrations in sediments at all stations around four shipyards was 8.70～22.15 ㎎/㎏. The concentrations of As in sediments at eight stations around A-shipyard, B-shipyard and D-shipyard were 14.93～22.15 ㎎/㎏ which exceeded TEL 14.5 ㎎-As/㎏ and ERL 8.2 ㎎-As/㎏, but did not reach PEL 75.5 ㎎-As/㎏ and ERM 70.0 ㎎-As/㎏. Those at three stations around C-shipyard were 8.70～14.24 ㎎/㎏ which were below TEL 14.5 ㎎-As/㎏, but were above ERL 8.2 ㎎-As/㎏. The average concentration of As in sediment around B-shipyard was 18.91 ㎎/㎏ and was 1.0～1.5 times higher than those around A-shipyard, C-shipyard and D-shipyard. This indicates that the works of shipyards or the discharges from shipyards had a little effect on the pollution of As in offshore sediment around shipyards.

7) The range of Hg concentrations in sediments at all stations around four shipyards was 0.02～0.35 ㎎/㎏. The concentrations of Hg in sediments at three stations around A-shipyard were 0.11～0.13㎎/㎏ which were almost equal to TEL 0.11 ㎎-Hg/㎏. Those at two stations around B-shipyard were 0.27～0.35 ㎎/㎏ which were above TEL 0.11 ㎎-Hg/㎏ and ERL 0.15 ㎎-Hg/㎏, but below PEL 0.62 ㎎-Hg/㎏ and ERM 0.71 ㎎-Hg/㎏. Those at six stations around C-shipyard and D-shipyard were 0.02～0.05 ㎎/㎏ which were below TEL 0.11 ㎎-Hg/㎏ and ERL 0.15 ㎎-Hg/㎏. The average concentration of Hg in sediment around B-shipyard was 0.31 ㎎/㎏ and was 2.6～7.8 times higher than those around A-shipyard, C-shipyard and D-shipyard. This implies that the works of B-shipyard or the discharges from B-shipyard had a little influence on the pollution of Hg in offshore sediment around B-shipyard.

8) The range of Fe concentrations in sediments at all stations around four shipyards was 2.90～3.66 %. The average concentration of Fe in sediment around A-shipyard was 3.48 %, and was 1.0～1.1 times higher than those around B-shipyard, C-shipyard and D-shipyard.

9) The range of Al concentrations in sediments at all stations around four shipyards was 3.12～6.80 %. The average concentration of Al in sediment around A-shipyard was 6.53 %, and was 1.2～1.8 times higher than those around B-shipyard, C-shipyard and D-shipyard.

Acknowledgements

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