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A Geo-statistical Assessment of Heavy Metal Pollution in the Soil Around a Ship Building Yard in Busan, Korea

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통계지표를 활용한 부산지역 조선소 주변 토양 내 중금속 오염조사 연구

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Abstract : *With the increase of metal usage in various industries, metal pollution and ecological toxicity in the environmental system have become a significant concern. A geo-statistical index has been widely used to determine contamination level with normalization through a background value. In this study, geo-statistical indexes such as an enrichment factor, accumulation index, and potential ecological risk index were used to assess metal pollution in soil at locations associated with shipbuilding manufacturing industries. Metal contamination, especially of Cu and Pb, was observed in some samples located closer to manufacturing sites. Enrichment factor and accumulation (IGEO) values were indicative of concerning levels of soil contamination in specific samples, and the soil contamination could be induced by anthropogenic sources. In further study, after more detailed sampling for soil and potential pollution sources, high interpretation techniques such as Pb isotope analysis and X-ray analysis will be needed to investigate source identification.*

Key Words : *Metal contamination, Geo-statistical index, Manufacturing industries, Anthropogenic source, Marine environment*

요 약 : 다양한 산업분야에서 중금속의 사용이 증가할수록, 중금속으로 인한 환경오염과 생물학적 위해성에 대한 우려의 목소리가 커지고 있다. 통계 지수는 배경농도 값과의 비교를 통해 중금속 오염농도를 정규화 시킴으로써 토양 오염의 정도를 수치화하고, 단계 별로 오염 정도를 판단 할 수 있어 많이 사용된다. 본 연구에서는 농축인자(Enrichment factor, EF), 축적 계수(accumulation index), 잠재적 생물학적 위험 지표(potential ecological risk index)등을 이용하여 중공업 근처 토양 내 중금속 오염가능성을 평가하였다. 연구결과, 중금속의 오염 정도는 정부 가이드라인에 비하여 낮은 수준이었으나, 특정 위치에서 아연, 구리, 납 등의 중금속 오염이 관찰 되었다. 농축인자, 축적계수, 생물학적 위험 지표를 통해 일부 토양 내 중금속 오염이 우려할 수준이며, 주변에 존재하는 인위적 오염원에 의한 오염가능성이 있음을 확인하였다. 연구대상지의 추가 시료채취 및 추정되는 오염원의 시료 확보 후, 동위원소 분석 및 x-ray 기반 분석을 통해 오염원 추적연구가 필요할 것으로 판단된다.

핵심용어 : 중금속 오염, 통계적 해석, 중공업, 인위적 오염원, 해양환경

1. Introduction

Manufacturing heavy industries, which are located in the coastal area or near a harbor, are one of the major anthropogenic sources for heavy metal pollution in environment systems such as soil, sediment, and seawater (Yang et al., 2017; Gottesfeld et al., 2018; Kulkarni et al., 2018). Exposed metals released from the industrial process are a great concern about potential biological toxicity as well as metal contamination (Gough et al., 1994; Govil and Narayana, 1999; Acero et al., 2003; Krishna and Govil, 2007; Han et al., 2016).

The harmful heavy metals generated during the process for

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Table 1. Analytical quality assurance for metal concentrations

	Al (%)	Ca (%)	Fe (%)	Mg (%)	Mn (%)	K (%)	Ti (%)	Cd (mg/kg)	Cu (mg/kg)	Pb (mg/kg)	Ni (mg/kg)	Zn (mg/kg)
Certification value	6.44	1.25	3.38	0.85	1.01	2.11	0.28	21.8	2950	5532	14.3	6953
Measured value	6.47	1.25	3.34	0.84	1.00	2.16	0.25	21.4	3020	5595	16.2	7006
STD	0.02	0.04	0.05	0	0.03	0.08	0.01	1.13	0.75	40.1	0.85	30.1
RSD (%)	0.31	3.20	1.50	0.00	3.00	3.70	4.00	4.81	0.02	0.72	4.63	0.43

building or repairing the ship are transferred into sediment and soils through the atmosphere, or released into the coastal area along with the rain. The study on contamination of coastal sediment or toxicity of benthic organisms (shellfishes) induced by manufacturing process in the industrial complex area has been variously carried out (Pekey, 2006; Chen et al., 2007). Untreated wastewater discharged from the neighboring industrial areas (Chen et al., 2007) and metal dust (Pekey, 2006) were significant sources of metal contamination in sediments. However, it has been relatively insufficient to investigate heavy metal pollution in the residential area near the shipbuilding manufacturing industries. Since soil contamination due to anthropogenic processes has been reported to a large extent (Wang et al., 2003; Sharma et al., 2006, 2007; Gottesfeld et al., 2018; Liu et al., 2018), it should be investigate heavy metal contamination that can be attributed to the marine industry not only in the marine environment but also in the vicinity of the heavy industry.

Statistical analysis for metal contamination and biological toxicity is a useful technique to qualify degree of contamination (Li et al., 2017; Yang et al., 2017). There are many indexs such as enrichment factor, geo-accumulation index, potential ecological risk index (PERI), and contamination factor (CF) including spatial distribution, multivariate statistical analysis, principal component analysis (PCA) and correlation analysis. Many researchers have expressed the degree of contamination for heavy metals as numerical value instead of the concentration (Ghrefat and Yusuf, 2006; Li et al., 2017; Yang et al., 2017; Kulkarni et al., 2018), so that level of contamination is easily understood.

In this study, the assessment of heavy metal contamination in field soils was conducted by applying the geo-statistical index. To investigate the degree of contamination level, metal concentration in soils were estimated and measured using inductively coupled plasma-optical emission spectrometer (ICP-OES). And then, geo-statistical index such as enrichment factor (EF), geo-accumulation index, and potential ecological risk index (PERI) was calculated. Metal correlation using statistical method was investigated to identify the potential anthropogenic source.

2. Materials and methods

2.1 Sample collection and analytical procedures

We focused on manufacturing industry which develops large-scale engines, modules for plant facilities, and manufactures stationary floating facility and shipbuilding for various types. Eleven soil samples (5-20 cm depth) were collected using soil auger with a 300 m of manufacturing industries (latitudes of 35°683’N-35°5.715’N and longitudes of 129°2.994’E-129°3.124’E). The position of samples for each sample is summarized in Fig. 1. Collected samples were stored in a polyethylene bag at room temperature and air-dried for 75 hr. Prior to the analysis, the soil samples were sieved through a No. 100 sieve.

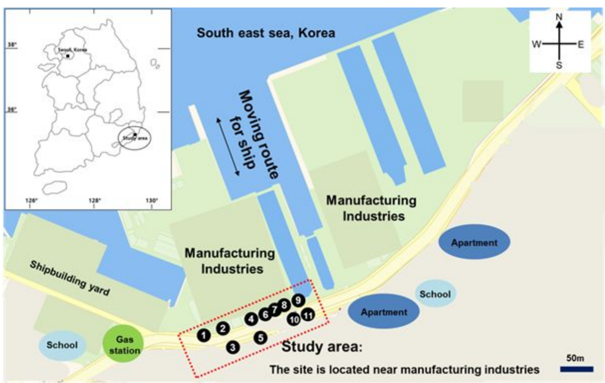

Fig. 1. Sample location in this study.

To estimate the metal concentration in a soil sample, the samples were analyzed using the proposed method by Jeon et al. (2017). Briefly, the soil was digested with hydrochloric acid (HCl) and nitric acid (HNO_3) at 105℃ for 2h. And separation of supernatant in digested solution was conducted using a centrifuge at 4000 rpm for 20 min, and the supernatant was diluted properly. Metal concentration was analyzed using inductively coupled plasma-optical emission spectrometer (ICP-OES). To ensure the quality of treatment method, the same standard reference proposed by Jeon et al. (2017) was analyzed and metal concentrations varied by less than ±5% of the values provided in the SRM. Details of the analytical quality assurance are summarized in Table 1. All samples were measured three times and average value was collected.

2.2 Enrichment factor (EF)

Many researchers have suggested qualifying the extent of heavy metal contamination in the soil. For the estimation of anthropogenic sources, the enrichment factor was often calculated in the soil (Ghrefat and Yusuf, 2006; Chen et al., 2007; Çevik et al., 2009). The metal concentration in soil was normalized to metal concentration of background value, and widely Al (Chen et al., 2007) and Fe (Ghrefat and Yusuf, 2006) for normalization were used. The upper continental crust value (Taylor and McLennan, 1995) was used to background concentration of metal (Upper crust value; Al: 8.04 wt%, Fe: 3.5 wt%, Pb: 20 µg/g, Zn: 71 µg/g, Cd: 98 µg/g, Cu: 25 µg/g, Cr: 85 µg/g). The enrichment factor (EF) is defined as follows:

$$EF_{Fe} = \frac{(\frac{C}{Fe})sample}{(\frac{C}{Fe})background}, \quad EF_{Al} = \frac{(\frac{C}{Al})sample}{(\frac{C}{Al})background} \qquad (1)$$

Where, $(C/Fe \text{ or } C/Al)_{sample}$ $(C/Fe \text{ or } C/Al)_{background}$ represents the relative toxic metal concentration in the sample and geochemical background.

The numerical results are indicated by different pollution level and EF is classified the five levels (Sutherland et al., 2000); EF < 2, minimal; 2 < EF < 5, moderate; 5 < EF < 20, significant; 20 < EF < 40, very high; and EF > 40, extremely high.

2.3 Index of geo-accumulation (I_{GEO})

I_{GEO} enables and can be calculated by comparing background concentration as below equation.

$$I_{\geq O} = \ln \frac{C_n}{1.5B_n} \qquad (2)$$

Where, C_n is the estimated concentration of each element in soil, B_n is the background concentration as geochemical value. The constant 1.5 was considered to the natural variation in the contents of a given substance in the environment and to the effect of small anthropogenic source.

The geo-accumulation index is representative value to compare relative soil contamination and consists of seven grades (Muller, 1969; Yaqin et al., 2008) (Table 2). The second grade may be 10 fold greater than the background value, and sixth grade means extremely contamination with hundredfold greater than background (Teng et al., 2002).

Table 2. Six grade of the geoaccumulation index

Grade	Value	Soil quality
0	$I_{GEO} \ll 0$	Practically uncontaminated
1	$0 < I_{GEO} < 1$	Uncontaminated to moderately contaminated
2	$1 < I_{GEO} < 2$	Moderately contaminated
3	$2 < I_{GEO} < 3$	Moderately to heavily contaminated
4	$3 < I_{GEO} < 4$	Heavily contaminated
5	$4 < I_{GEO} < 5$	Heavily to extremely contaminated
6	$5 < I_{GEO}$	Extremely contaminated

2.4 Ecological toxicity

Biological toxicity can be expressed potential ecological risk index (PERI) proposed by Hakanson in 1980. PERI consists of three modules and is calculated as follow (Jiang et al., 2014); a degree of contamination (CD), toxic-response factor (TR) and potential ecological risk factor (ER).

$$\begin{aligned} EC_f^i &= C_D^i / C_R^i \\ E_R^i &= T_R^i \times C_f^i \\ RI &= \Sigma_{i-1}^m E_R^i \end{aligned} \qquad (3)$$

Where, C_D^i is a concentration of heavy metals in each sample, C_R^i is the background value as base line, C_f^i is the contamination level of a single element, E_R^i is the biological hazard of a single element, RI is the overall biological hazard, T_R^i is biological toxicity weight of a single element with 1for zinc (Zn), 2 for chromium (Cr), 30 for cadmium (Cd), 5 for copper (Cu) and lead (Pb). The meanings of the values calculated by the above equations are shown in Table 3.

Table 3. Potential ecological risk index (PERI) as biological toxicity

$E_{R}i$	Pollution degree	RI	Risk level	Risk degree
$E_{R}i$ <30	Slight	RI <40	A	Slight
30≤ $E_{R}i$ <60	Medium	40≤ RI <80	B	Medium
60≤ $E_{R}i$ <120	Strong	80≤ RI <160	C	Strong
120≤$E_{R}i$ <240	Very strong	160≤ RI <320	D	Very strong
240≤ $E_{R}i$	Extremely strong	320≤RI	-	

3. Results and discussion

3.1 A degree of metal contamination in soils

The individual results obtained of each metal are presented in Table 4 and Fig. 2. The metal concentration had a range of 21.2-85.3 mg/kg, 18.7-44.2 mg/kg, 47.1-335.6 mg/kg, 34.7-338.0 mg/kg, and 0.2-0.8 mg/kg for Pb, Cr, Zn, Cu, and Cd. Overall, no high level of contamination in soil was observed comparing background values in upper crust (Pb: 20 mg/kg, Zn: 71 mg/kg, Cd: 98 mg/kg, Cu: 25 mg/kg, Cr: 85 mg/kg) (Taylor and McLennan, 1995) and government guidelines. But concern levels of pollutants in specific soil showed with giving rise to concern anthropogenic pollution. Especially, metal concentration in specific points such as S-4, S-5, and S-10 was significantly higher than in the other position and it could be assumed that metal particles scattered from heavy industry facility were deposited and accumulated into soil. S-4 and S-5 were classified into area near and far from heavy industry on the basis of roads, and metal concentration in their samples was extremely different according to sample position. The difference of linear distance from the manufacturing industry to the sample is only about 10 meters, but the difference of heavy metal concentration is more than double. It can be seen that the possibility of the dispersion and deposition of heavy metal particles increase as closer from manufacturing industry, and it could be assumed that heavy metal industry is one of the anthropogenic sources for soil contamination. And, characteristically, the concentration of heavy metals in S-3 and S-10 were higher than other samples such as S-2, S-6, S-7, S-8, and S-9, which indicated metal particle scattered from heavy industry could be deposited to the other side of roads. The linear distance from heavy industry is approximately 50-200 m, so it is possibility to disperse metal particles from anthropogenic sources.

Heavy metal contamination can be found in many industries area and disposal dumping, which can release high concentration of heavy metals (Govil and Narayana, 1999). These elements can be leached into ground water or sea water, and later the plant that absorbed heavy metal may affect human health (Gough et al., 1994; Acero et al., 2003; Krishna and Govil, 2007). Although metal contamination in this study was not severe, consideration and monitoring of potential pollution hazards in the future around manufacturing industries are needed.

Table 4. Metal concentration in soils

Sample name	Pb (mg/kg)	Cr (mg/kg)	Zn. (mg/kg)	Cu (mg/kg)	Cd (mg/kg)
S-1	34.3	34.5	51.1	66.6	0.4
S-2	61.2	28.4	17.4	34.7	0.5
S-3	39.7	28.5	154.4	192.2	0.8
S-4	85.3	44.2	335.6	338.0	0.7
S-5	71.7	29.3	299.9	250.9	0.5
S-6	25.9	19.5	54.0	64.2	0.2
S-7	21.2	23.0	63.7	83.2	0.3
S-8	60.4	25.7	112.8	130.9	0.2
S-9	28.9	29.7	47.1	50.3	0.3
S-10	79.1	25.7	176.7	220.8	0.8
S-11	64.6	18.7	80.4	114.2	0.4

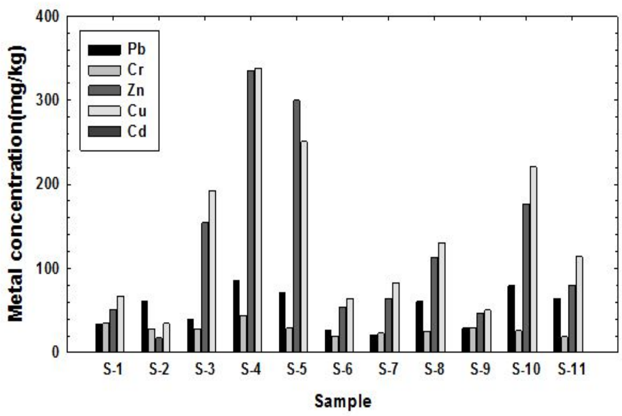

Fig. 2. Metal concentration in soils.

3.2 Enrichment factor and geo-accumulation index

For the estimation of anthropogenic sources, Enrichment factor (EF) and geo-accumulation index were calculated based on metal concentration (Table 5 and Figs. 3-4). EF factor and geo-accumulation index effectively represent the degree of heavy metal present in the soil with comparing background value (Salomons and Förstner, 1984; Feng et al., 2004). An EF value of more than 5 indicated environmental concern about heavy metal

Table 5. Enrichment factor and geo-accumulation index at each metal

Metal	Index	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-11
Pb	EF_{Al}	1.8	3.2	2.1	4.4	3.7	1.3	1.1	3.1	1.5	4.1	3.3
	EF_{Fe}	4.2	7.5	4.9	10.4	8.8	3.2	2.6	7.4	3.5	9.7	7.9
	I_{GEO}	0.1	0.7	0.3	1.0	0.9	-0.1	-0.3	0.7	0.0	1.0	0.8
Zn	EF_{Al}	0.7	0.3	2.3	4.9	4.4	0.8	0.9	1.6	0.7	2.6	1.2
	EF_{Fe}	0.8	0.3	2.3	5.0	4.5	0.8	1.0	1.7	0.7	2.7	1.2
	I_{GEO}	-0.7	-1.8	0.4	1.1	1.0	-0.7	-0.5	0.1	-0.8	0.5	-0.3
Cu	EF_{Al}	2.8	1.4	8.0	14.0	10.4	2.7	3.4	5.4	2.1	9.1	4.7
	EF_{Fe}	2.8	1.5	8.2	14.4	10.7	2.7	3.6	5.6	2.1	9.4	4.9
	I_{GEO}	0.6	-0.1	1.6	2.2	1.9	0.5	0.8	1.2	0.3	1.8	1.1
Cd	EF_{Al}	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
	EF_{Fe}	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
	I_{GEO}	-5.8	-5.8	-5.3	-5.3	-5.7	-6.5	-6.2	-6.5	-6.3	-5.2	-5.8
As	EF_{Al}	4.3	0.5	5.5	5.7	4.9	5.9	6.1	3.3	6.0	5.9	4.1
	EF_{Fe}	4.5	0.5	5.7	5.9	5.0	6.1	6.2	3.4	6.2	6.1	4.2
	I_{GEO}	1.0	-1.1	1.3	1.3	1.1	1.3	1.4	0.8	1.3	1.3	1.0

contamination and also I_{GEO} index more than 1. The EF value was not severe level in overall samples and has a range with 1.1-4.4, 0.3-4.9, 1.4-14.0, 0.2-0.5 for Pb, Zn, Cu, and Cr based on Al (Based on Fe: 2.6-10.4 for Pb, 0.3-5.0 for Zn, 1.5-14.4 for Cu, 0.2.-0.6 for Cr) (Fig. 3). Contamination of Cd was not observed unlike other metals (EF < 0.1 based on Al and Fe). The relative values of Pb and Cu were higher than those of other metals. Especially, pollution level for S-4 and S-5 were significant so soil contamination in soil induces by anthropogenic source could be estimated with comparing other samples.

Geo-accumulation index (I_{GEO}) showed similar trend to the enrichment factor. I_{GEO} value had a range with -0.3-1.0, -1.8-1.1, -0.1-2.2, -1.0-1.8 for Pb, Zn, Cu, and Cr (Fig. 4). In specific soil samples, moderately contamination was observed for Pb, Zn, and Cu not Cd. Especially, in S-3-S-5 and S-10, copper contamination was severe (I_{GEO}: 1.6-2.2) and the value was more than two which indicated heavily contaminated condition. The I_{GEO} values of Pb in most soils fell in group 1(uncontaminated to moderately contaminated) except S-4 and S-10 (in group 2) (moderately contaminated). For the Zn metal I_{GEO} grade varied from group 0 (practically uncontaminated) to group 2 (moderately contaminated). And, for the Cu metal, overall I_{GEO} grad was high from group 0 (practically uncontaminated) to group 3 (moderately to heavily contaminated condition). Pollution of Cu and Pb could be due to anthropogenic sources including manufacturing industries.

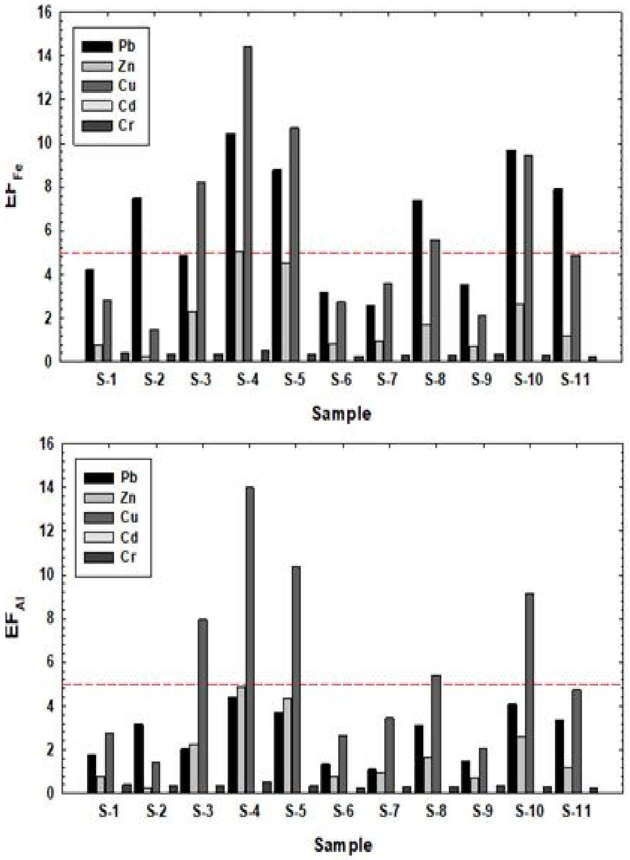

Fig. 3. Enrichment factor based on Fe and Al for each sample.

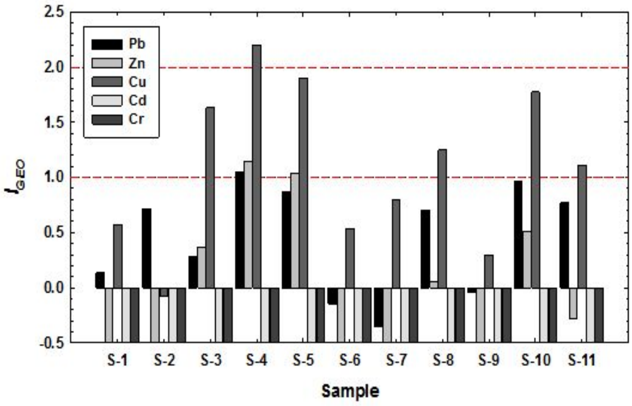

Fig. 4. Geo-accumulation index for each sample.

3.3 Metal correlation in soils

The concentration distribution of heavy metal in soil was similar in places where pollution level was high. Lead, zinc, and copper were commonly observed in contaminated soil, so the correlation was calculated using statistical methods to confirm the correlation of these metals. A relative correlation was expressed as a number from -1 to +1, with 1 indicating definitely positive and -1 indicating a negative relation. Also, to observe these results more clearly and easier, a heating map was used to express correlation degree using red, white and blue color (Table 6). There was a high correlation ($p < 0.05$) between Pb, Zn, and Cu in soils, and their correlation value was about 0.69 for Zn and 0.72 for Cu based on Pb. And the correlation of Cd between other metals was not significant.

The manufacturing industry in study area develops large-scale engines, modules for plant facilities, and manufactures stationary floating facility and shipbuilding for various types. Various metals are used in the related process, and the metals could be exposed and scattered to the atmosphere and then deposited. Al-Mg-Zn alloy was interested in shipbuilding industries because strength property was better than Al-Mg alloy (Dudzik and Czechowski, 2009). Metals pollution such as Cu, Zn, Cd, and Cr in samples was observed due to shipbuilding actives (Adamo et al., 2005) and also inorganic compounds oxides of Ti, Zn, Cu, Fe, Pb generally used in paint with trace elements (Jolly et al., 2006) for primers, anticorrosive and antifouling (EPA, 1997; EPA, 2000). Manufacturing industry, which had various processes, could be one of the important anthropogenic sources affecting soil contamination.

Table 6. Metal correlation in soil samples by statistical analysis

	Pb	Cr	Zn	Cu	Cd
Pb	1.00				
Cr	0.36	1.00			
Zn	0.69	0.56	1.00		
Cu	0.72	0.54	0.97	1.00	
Cd	0.60	0.44	0.60	0.72	1.00

※ Heating map shows more clearly the correlation between heavy metals. The red color means strongly positive relationship (+1) and the blue color means strongly negative relationship (-1). The white color is presented in no relationship between metals.

3.4 Ecological toxicity

The potential ecological risk index (PERI) for Pb, Zn, Cu, and Cd was calculated and presented in Table 7 and Fig. 5. In Fig. 5, the RI which indicated the overall biological hazard was only presented at each sample. The RI value was in medium grade ($40 < RI < 80$) for S-3, S-5, S-8, and S-10. And, strong biological risk ($80 < RI < 160$) was only observed in S-4. Among the various metals, E_R indicated that the biological hazard of a single element of Cu was largest.

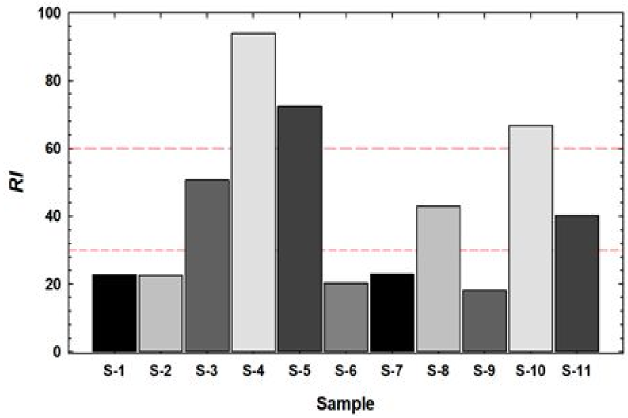

Fig. 5. Biological toxicity of heavy metals in soils.

Leung et al. (2017) reported that the level of heavy metal such as Pb, Zn, and Cd in sediment extremely exceeded comparing reference value and their bioaccumulation was also a concern level. The metals released from anthropogenic sources were present in the groundwater, sea water, and sediment and human health could be affected by plant absorbed metals (Acero et al., 2003; Krishna and Govil, 2007). Although the pollution of metal was localized in a specific point, consideration of potential biological risks and steady monitoring are needed.

Table 7. PERI at each metal in each sample

		S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-11
Pb	E_{cf}	1.7	3.1	2.0	4.3	3.6	1.3	1.1	3.0	1.4	4.0	3.2
	E^R	8.6	15.3	9.9	21.3	17.9	6.5	5.3	15.1	7.2	19.8	16.2
Zn	E_{cf}	0.7	0.2	2.2	4.7	4.2	0.8	0.9	1.6	0.7	2.5	1.1
	E^R	0.7	0.2	2.2	4.7	4.2	0.8	0.9	1.6	0.7	2.5	1.1
Cu	E_{cf}	2.7	1.4	7.7	13.5	10.0	2.6	3.3	5.2	2.0	8.8	4.6
	E^R	13.3	6.9	38.4	67.6	50.2	12.8	16.6	26.2	10.1	44.2	22.8
Cd	E_{cf}	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
	E^R	0.1	0.1	0.2	0.2	0.2	0.1	0.1	0.1	0.1	0.3	0.1
	RI	22.8	22.6	50.8	93.9	72.5	20.2	22.9	42.9	18.0	66.7	40.3

4. Conclusion

In this study, to estimate the effect of an anthropogenic source to soil contamination, statistical analysis methods such as the enrichment factor, geo-accumulation index, and ecological toxicity index were calculated. Various statistical methods were used because the pollution degree of metals can be estimated differently according to applied value. Metal contamination in soils was not severe but concern level of pollution for metals was observed in some points, especially Zn, Cu, and Pb. And also, strong biological risk ($80 < RI < 160$) and heavily contamination condition ($2 < I_{GEO}$) were resulted in specific sample which was located in front of manufacturing industry. The degree of metal contamination by various statistical methods was not severe, but it indicates that it is necessary to consider potential pollution by the heavy manufacturing industry and monitor it continuously.

The metals generated during the process for building or repairing the ship are transferred into sediment and soils through the atmosphere, or released into the coastal area along with the rain. However, it has been relatively insufficient to investigate heavy metal pollution in residential area near the shipbuilding manufacturing industries. In this study, metal contamination caused by the marine industry can be observed not only in the coastal but also in the soil, and it can be used as basic data for prevention against heavy metal pollution. In future work, after additional sampling for field and potential anthropogenic sources, researches as source identification for contamination, transportation mechanism of metals, and characteristic of metals in soil is needed using a variety of analytical methods such as isotope analysis and X-ray technique.

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