Characteristics of tailings from the closed metal mines as potential contamination source in South Korea

Kwang-Koo Kim \cdot Kyoung-Woong Kim \cdot Ju-Yong Kim \cdot In S. Kim Young-Wook Cheong \cdot Jeong-Sik Min

Abstract There are about 1,500 closed metal mines in South Korea. Most of the tailings have been left without any management in these mines and have become the main source of heavy metal contamination of agricultural soils and crops in the mining areas. In this study, to predict the potential impact of tailings on nearby environments, the characteristics such as pH, particle size distribution, loss on ignition, cation exchange capacity, and the concentration and speciation of heavy metals in the tailings collected from ten closed metal mines in South Korea were investigated. Based on these characteristics, the relative metal-binding capacity, pollution index, and danger index were calculated so as to enumerate the priorities for remediation. The relative metal-binding capacities based on pH, clay, and organic content were very weak (<1) in the tailings from the Daduck, Duckum, and Imchun mines. The concentrations of Cd, Cu, Pb, and Zn exceeded the tolerable levels to the degree of being phytotoxically excessive in all the tailings, and the pollution indices ranged from 5.45 to 58.58. The results of the sequential extraction analysis showed that a large proportion of heavy metals existed in the form of a residual fraction in most tailings. The concentrations of the mobile phase of these heavy metals, however, were relatively high in the tailings from the

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K.-K. Kim · K.-W. Kim () · J.-Y. Kim · I.S. Kim Advanced Environmental Monitoring Research Center (ADEMRC), Kwangju Institute of Science and Technology, Oryong-dong, Puk-gu, Kwangju 500-712, South Korea E-mail: kwkim@kjist.ac.kr Tel.: +82-62-9702442 Fax: +82-62-9702434

Y.-W. Cheong · J.-S. Min Korea Institute of Geology, Mining and Materials (KIGAM), Taejon 305-350, South Korea Daduck, Dalsung, Imchun, and Duckum mines. Considering the results of the relative metal-binding capacity, pollution index, and danger index, the priority of remediation for these mines from most urgent to less urgent were Daduck, Imchun, Dalsung and Sujum, and then Goobong.

Keywords Tailings · Heavy metals · Relative metalbinding capacity · Pollution index · Danger index

Introduction

Heavy metals make up a large group of elements with an atomic density greater than 6 g/cm³ (Phipps 1981), and are known as the potential sources of contamination in soils, streams, and groundwater. It has recently been estimated that about 0.5, 20, 240, 250, and 310 million tonnes of Cd, Ni, Pb, Zn, and Cu, respectively, have been mined and ultimately deposited in surface and subsurface soils (Nriagu 1984). According to certain changes in the physical and chemical properties in the lithosphere, heavy metals in tailings can be transported to, dispersed to, and accumulated in plants and animals, and can then be passed up the food chain to human beings as a final consumer. Heavy metals in tailings exist in various forms, and those species may have an influence on the mobility and bioavailability. If heavy metals exist in a form that is exchangeable or adsorbed to the surface of clay, organic matter, or oxides with weak bonding strength, they can be easily moved and dispersed into the ecosystem. Metals bound with organic ligands or held within a crystal lattice, however, are not easily separated and moved (Fletcher 1981; Thomson and Wood 1982). To monitor the longterm impact of heavy metals on the nearby environment, it is necessary to examine the speciation as well as total concentrations of these metals.

There are about 1,500 closed metal mines in South Korea, and piles of tailings have been left without any treatment system in the vicinity of the mining areas. The tailings contain several kinds of heavy metals, some of which are toxic contaminants in the soil that may cause adverse effect on the ecosystem around the metal mines (Jung and Thornton 1996; Min and others 1997; Kim and others 1998).

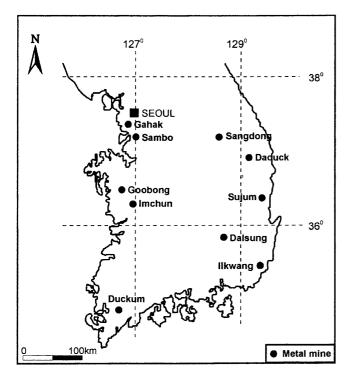


Fig. 1
Sampling locations of ten metalliferous mines in Korea

To control the hazards presented by these closed metal mines, preventive facilities should have already been constructed, but most of the closed metal mines in Korea have not yet been remediated. The degree of impact from these unremediated mines has not been determined, and the recent heavy metal contamination of agricultural soils and crops in the mining areas is one of most serious environmental problems in South Korea. A detailed investigation on the tailings as a source of contamination in metal mining areas needs to be carried out, the results of

which should include a priority-based schedule for remediation. The objectives of this study were to investigate the characteristics of tailings as a potential contamination source, and to evaluate the priority of remediation for metal mining areas in Korea. The results of this study may also be useful in selecting remediation technologies for the contaminated mining areas.

Materials and methods

Tailing samples were collected from ten closed metal mines: Daduck, Dalsung, Duckum, Gahak, Goobong, Ilkwang, Imchun, Sambo, Sangdong, and Sujum (Fig. 1). The target mining metals, bedrock type, and heavy metal concentrations in the bedrock are listed in Table 1 (Thornton 1983; John 1997). About 5 kg of one composite tailing sample was composed of 80 subsamples collected by hand auger at surface, 15, 30, and 45 cm depth of tailing piles. The tailing samples were air-dried, disaggregated, and sieved to a –10 mesh (less than 2 mm).

To determine the pH of the tailings, 25 mL of deionized water was added to 5 g of a tailing sample, stirred intermittently for 1 h and then allowed stand for 0.5 h. The pH was measured with a pH meter that had been calibrated with the standard solution of 4.01 and 7.00. The hydrometer method was used for the particle size analysis (Day 1965). To determine the loss on ignition, crucibles containing 10 g of a sample were dried at 105 °C, placed in a muffle furnace and ignited at 400 °C for 24 h. The samples were cooled to 105 °C, and the percentage lost on ignition was calculated from the dry weight.

To determine the total concentrations of the heavy metals, 1 mL of HNO $_3$ and 3 mL of HCl (aqua regia) were added to 2 g of a tailing sample that had previously been dried and sieved to a -80 mesh (<180 μ m). These were heated to 70 °C and shaken for 1 h, and 4 mL of deionized water was

Table 1Characteristics of the studied metal mines in Korea and quoted heavy metal concentrations in these bedrock types (Thornton 1983; John 1997)

Mine	Mining element	Ore mineral	Bedrock type	World average concentration of heavy metals in bedrock					
				Cd	Cu	Mn	Pb	Zn	Fe
Ilkwang	Au, Ag, Cu	FeS ₂ , PbS, ZnS	Shale	0.3	57	670	20	80	33,300
			Sandstone	-	_	-	7	16	_
			Granodiorite	0.2	30	1,200	15	60	_
Imchun	Au, Ag, Cu, Pb,	ZnS, FeS ₂ , PbS	Granite	0.1	20	600	20	60	27,000
	Zn		Granodiorite	0.2	30	1,200	15	60	_
			Schist	_	30	900	30	_	4,900
Daduck	Au, Ag, Pb, Zn	CuFeS ₂ , PbS, FeAsS	Gneiss	_	_	_	-	_	_
Dalsung	Au, Ag, Cu, W	FeAsS, CaCO ₃	Andesite	0.09	35	1,200	15	72	58,500
Sujum	Au, Ag, Pb, Zn	_	Shale	0.2	50	850	20	100	_
•	C		Sandstone	_	_	_	7	16	_
Sangdong	Au, Ag, Cu, Bi, W, Mo	-	-	-	-	-	-	-	
Gahak	Au, Ag, Cu, Pb, Zn	-	Gneiss Quartzite	-	-	-	-	-	-
Goobong	Au, Ag, Cu, Pb, Zn	FeS ₂ , PbS, As	Biotite granite	0.1	2	600	20	60	_
Sambo	Pb, Zn, W	-	Biotite schist	_	30	900	30	_	_
Duckum	Au	ZnS, FeS ₂ , PbS	Felsite	_	_	_	_	_	_
		· •/	Granite	0.1	20	600	20	60	27,000

Step 1 Exchangeable Fraction

Extracted Component Exchangeable Ions Reagents NH₄OAc, 1.0 M, pH 7

Step 2 Carbonatic Fraction

Extracted Component Carbonates Reagents NaOAc, 1.0 M, pH 5 with HOAc

Step 3 Easily Reducible Fraction

Extracted Component Mn-Oxides Reagents 0.1 M NH₂OH HCl with 0.01 M HNO₃

Step 4 Moderate Reducible Fraction

Extracted Component Amorphous Fe-Oxides Reagents 0.2 M Oxalate buffer, pH 3

Step 5 Sulfidic/Oragnic Fraction

Extracted Component Sulfides with Organics Reagents 30 % H₂O₂ with 0.02 M HNO₃ pH 2 1M NH₄OAc, In 6% HNO₃

Step 6 Residual Fraction

Extracted Component Lithogenic Matters Reagents HNO₃ and HCl

Fig. 2Sequential extraction procedure modified from Kersten and Förstner (1995)

added to the solution. These solutions were analyzed for Cd, Cu, Pb, and Zn with an atomic absorption spectrometer, and rigorous quality control was carried out (Davidson and others 1994).

A modified version of the sequential extraction analysis set forth by Kersten and Förstner (1995) was used to determine the chemical speciation of the heavy metals. This method consisted of six steps: step 1 was for the exchangeable ion fraction, step 2 was for the carbonate fraction, step 3 was for the Mn oxide fraction, step 4 was for the Fe oxide fraction, step 5 was for the sulfides/organic matter fraction, and step 6 was for the residual fraction known as crystalline mineral form. The sequential extraction procedure is shown in Fig. 2, and a strong acid (HNO₃+HCl) was used for the residual fraction instead of nitric acid.

Results and discussion

Geochemical characteristics of the tailings

The pH values of the tailings from the Daduck, Dalsung, Duckum, Ilkwang, Imchun, and Sujum mines were less than 4.0, but those from the Gahak, Goobong, and Sangdong mines were greater than 8.0 (Table 2). The mobility of the cation generally increased under acidic conditions, such that a great number of heavy metals in the tailings from Daduck, Dalsung, Duckum, Ilkwang, Imchun, and

Sujum may be more mobile and more easily extracted than those from other mines.

The heavy metals that are in cation form (e.g., Cd²⁺, Cu²⁺, Pb²⁺, and Zn²⁺) tend to be adsorbed on the surface of clays, which are negatively charged. The mobility of these elements may decrease with increasing clay content. The tailings from the Daduck, Dalsung, Duckum, and Sangdong mines, and especially the Sujum mine, all had larger contents of clay (>20%, Table 2); therefore, the sorption capacities of these tailings are expected to be high. Copper, lead, and some other metals have a high affinity for soil organic matter, which has both a cation exchange property and chelating ability (Adriano 1986). The loss on ignition values of the tailing samples were relatively small, and the Imchun mine had the largest value (3.81%; Table 2). The cation exchange capacity (CEC) is a measure of the number of ions that can be adsorbed, in an exchangeable fashion, on the negative charge sites of the soil (Bache 1976). The CEC of the tailing samples ranged from 3.53 to 9.03 meq/100 g (Table 2). The largest value of CEC was found in the tailings from Sujum mine, which may be related to the relatively high clay content. The concentrations of heavy metals in the tailings by aqua regia digestion are listed in Table 2. Generally, the aqua regia digestion extracts from 70 to 90% of the total concentration of Cd, Co, Cr, Cu, Fe, Mn, Ni, and Pb in soils (Alloway 1990). The concentration of Cd in all the tailing samples was greater than the tolerable level which is considered to be phytotoxically excessive: 300 µg/g Zn, 100 μg/g Pb, 100 μg/g Cu, and 3 μg/g Cd (Kabata-Pendias and Pendias 1984). In particular, the concentration of Cd in the tailings from the Daduck and Goobong mines was 50 times greater than its tolerable level.

The concentration of Cu in the tailings from Duckum, Sambo, and Sujum was less than the tolerable level, while the tailings from the Dalsung mine contained 1% of Cu. The concentration of Pb in all the tailings, except those from the Sambo and Sangdong mines, was ten times greater than its tolerable level. The concentration of Zn in the tailings from the Daduck, Imchun, and Sujum mines were significantly high, and the tailings from the Daduck mine contained up to 4% of Zn.

Chemical speciations of heavy metals in the tailings

The chemical speciation of heavy metals is affected by various environmental conditions. The exchangeable ion

Physical properties and concentrations of heavy metals in tailings from closed metal mines

Mine	pН	Sand (%)	Silt (%)	Clay (%)	LOI (%)	CEC (meq/100 g)	Cd (mg/kg)	Cu (mg/kg)	Pb (mg/kg)	Zn (mg/kg)
Daduck	2.67	27	47	26	0.72	7.28	148	1,280	3,886	40,000
Dalsung	3.56	57	31	22	1.65	6.33	8	10,000	2,450	1,160
Duckum	2.40	52	27	21	1.34	6.32	8	108	1,560	840
Gahak	8.23	74	23	3	0.20	5.94	28	560	1,050	4,360
Goobong	8.02	24	63	13	0.10	6.42	136	400	5,822	4,840
Ilkwang	2.65	68	21	11	2.33	4.79	4	400	1,826	284
Imchun	1.91	71	19	10	3.81	3.53	46	220	9,780	13,600
Sambo	6.61	77	19	4	0.40	4.51	4	40	538	4,400
Sangdong	8.26	25	51	24	0.60	7.08	16	560	166	880
Sujum	3.46	10	33	57	1.61	9.03	44	160	4,774	14,800

fraction is likely to be affected by sorption-desorption processes that occur on the surface of clays, oxides, and organic matter. Trace metal associated with carbonate fraction is susceptible to changes in pH. The oxide fractions for Fe and Mn, which are excellent scavengers for trace metals, are thermodynamically unstable under anoxic conditions. The fraction of sulfides/organic matter can be degraded, leading to a release of soluble trace metals under oxidizing conditions in natural waters. The residual fraction, in which trace metals may be held in a crystal structure, is not expected to release trace metals in solution over a reasonable time span under the conditions normally encountered in nature (Tessier and others 1979). The results of the sequential extraction analysis are listed in Table 3, and the relative percentages of each fraction are plotted in Fig. 3. In the case of Cd, the fraction of sulfides/ organic matter or residual was larger than any other fraction. The total concentration of Cd was similar in the tailings from Daduck and Goobong mines (139 and 144 μg/g, respectively), but the proportions of the exchangeable ion fraction of Cd were significantly different. The exchangeable Cd concentration was 40 μg/g for the Daduck mine, and 2 µg/g for the Goobong mine. This indicated that the Cd in the tailings from the Daduck mine may be easily released and transported into the nearby soil

The largest portion of Cu was associated with sulfides/ organic matter, because Cu has a high geochemical affinity for sulfides or organic matter. The total concentration of Cu was remarkably high in the tailings from the Dalsung mine, and the proportion of the exchangeable ion fraction was also high. In the case of Pb, the residual fraction was absolutely predominant, except for the tailings from the Imchun mine, which contained 2,020 $\mu g/g$ of exchangeable ion fraction. However, it was obvious that the carbonate

fraction was relatively predominant at the Gahak and Sambo mines.

Because the sulfides/organic matter and residual fractions were the major fractions in Zn, it can be inferred that Zn may not be released under the normal conditions (Fig. 3). However, the proportion of the exchangeable Zn ion fraction in the tailings from the Daduck and Duckum mines were 30 and 63%, respectively. In particular, the concentration of the exchangeable ion fraction of Zn in the tailings from the Daduck mine was 9,400 $\mu g/g$, and high level of Zn contamination in the soil and water system around the Daduck mine is expected from this contamination source.

Priority list of remediation for the contaminated mining areas

Based on the results of chemical analyses, it was possible that the tailings of the investigated mines will have an impact on nearby soil and water systems, making a strong argument for carrying out a remediation program in these mining areas. In this study, a prioritized list for remediation was suggested based on the geochemical characteristics and chemical speciation of heavy metals in the tailings. Blume and Brümmer (1991) suggested a method that predicts heavy metal behavior in soils based on the results of simple field tests. According to this method, the relative metal-binding capacity of soil in relation to pH, redox potential, clay, organic matter, and iron oxide content can be calculated. For the ten closed metal mines investigated in this study, the relative binding capacity of the tailings was calculated for Cd, Cu, Pb, and Zn using Blume and Brümmer's (1991) method (Table 4). The redox potential and iron oxide content of the tailings were not used for this calculation because these data were not available. The relative metal-binding capacities of the tailings from the

Table 3The concentrations of each chemical form from the sequential extraction analysis in tailings

Element	Fraction	Daduck	Dalsung	Duckum	Gahak	Goobong	Ilkwang	Imchun	Sambo	Sangdong	Sujum
Cd (mg/kg)	Exchange	40	2	5	5	2	0.2	3	1	1	14
, , ,	Carbon.	15	2	0.2	4	2	1	1	1	2	3
	Mn-oxide	5	0.2	0.2	1	1	0	0.2	0.2	1	1
	Fe-oxide	4	0.2	0.2	0.2	1	1	0.2	0.2	0.2	1
	Sulf./Organ.	60	4	1	11	110	1	7	1	5	20
	Residual	15	0.2	8	0.2	30	1	30	1	0.2	1
Cu (mg/kg)	Exchange	33	1,900	9	19	6	5	17	3	7	1
	Carbon.	42	1,040	4	28	4	16	6	8	15	4
	Mn-oxide	1	9	1	2	1	1	1	1	1	1
	Fe-oxide	5	204	6	19	1	26	12	4	10	3
	Sulf./Organ.	500	4,600	34	211	215	165	144	21	211	43
	Residual	225	1,965	32	120	60	165	75	15	105	45
Pb (mg/kg)	Exchange	70	1	36	42	237	1	2,020	89	1	75
	Carbon.	310	14	48	450	913	3	413	155	31	123
	Mn-oxide	16	1	1	7	150	1	92	39	9	18
	Fe-oxide	125	24	70	61	85	49	90	74	1	137
	Sulf./Organ.	710	87	581	300	923	1,210	303	57	76	760
	Residual	2,565	2,205	952	240	3,690	555	4,200	150	165	3,270
Zn (mg/kg)	Exchange	9,400	66	639	150	44	1	520	102	7	1,000
	Carbon.	5,300	30	64	420	122	4	43	250	37	1,200
	Mn-oxide	1,900	5	10	92	44	1	10	107	16	110
	Fe-oxide	2,500	8	8	420	111	5	9	124	83	76
	Sulf./Organ.	9,000	230	93	1,500	7,100	86	1,800	800	95	8,000
	Residual	3,150	210	192	375	765	105	4,650	1,155	90	1,095

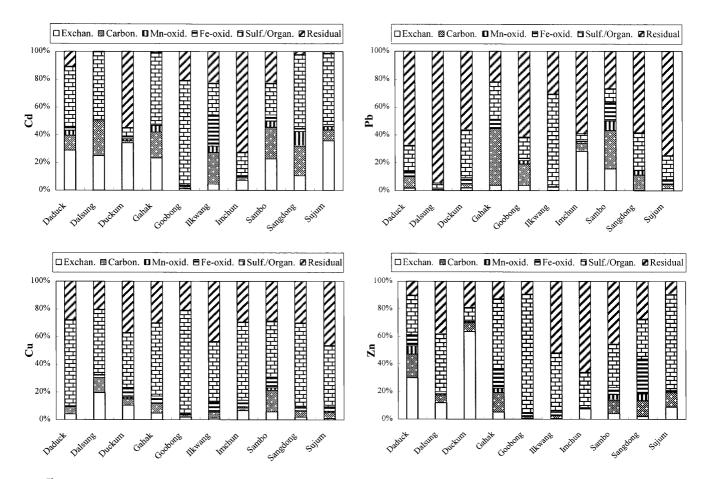


Fig. 3
Partitioning of sequential extracted metal concentrations in tailings

Table 4 Relative metal-binding capacity, pollution index (*PI*), and danger index (*DI*) of tailings

Mine	Relative	metal-binding o	PI	DI			
	Cd	Cu	Pb	Zn			
Daduck	0.5	1.0	1.0	0.5	58.6	634.6	
Dalsung	1.0	2.0	3.5	1.5	32.8	67.8	
Duckum	0.5	1.0	1.0	0.5	5.5	46.4	
Gahak	5.0	5.0	5.0	5.0	10.0	14.8	
Goobong	6.0	6.0	6.0	6.0	34.9	26.4	
Ilkwang	0.5	1.5	1.5	0.5	6.1	0.3	
Imchun	0.5	1.0	1.0	0.5	40.2	233.3	
Sambo	4.5	5.0	5.0	5.0	5.4	15.6	
Sangdong	6.0	6.0	6.0	6.0	3.9	0.8	
Sujum	1.5	2.5	4.0	2.0	28.3	74.1	

^aRating: <1, very weak; 2, weak; 3, medium; 4, strong; >5, very strong

Daduck, Duckum, and Imchun mines were less than 1, which means that heavy metals in these tailings can easily move into the nearby environment. On the other hand, the tailings from Gahak, Goobong, Sambo, and Sangdong mines showed a very strong metal-binding capacity. Heavy metal contamination in surface environment is usually associated with a mixture of contaminants rather than a single element. Many researchers have used a pollution index (PI) of geological materials to identify the multi-element contamination that may increase the overall metal toxicity (Nishida and others 1982; Chon and others

1995; Kim and others 1998). The pollution index is computed by averaging the ratios of the total concentration of heavy metals to the hazard criteria (the tolerable level). The tolerable level is derived from the threshold that indicates the phytotoxically excessive level (Kabata-Pendias and Pendias 1984).

Pollution Index (PI) =

 $\frac{\sum (Total conc. of element/Tolerable level of element)}{Number of elements}$

The tolerable levels of Zn, Pb, Cu, and Cd are 300, 100, 100, and 3 μ g/g, respectively. If a pollution index is greater than 1, it indicates that, on the average, the metal concentrations are greater than the permissible levels. The calculated PI for the tailing samples ranged from 5.4 to 58.6 (Table 4), which indicated that all the tailings contained heavy metals at a level that causes toxicity to the ecosystem. The order of pollution severity from highest to lowest for the mines in this study was Daduck, Imchun, Dalsung, Goobong, and Sujum.

The short-term impact of tailings on the soil-plant-human system is dependent on the availability of the heavy metal in the tailings. If metals exist in a crystal lattice or as sulfides, it may take a long time to cause adverse effect on plants and animals. Availability is concerned with metals existing in an easily releasable form. To select the contaminated mine that has a possibility of a short-term impact on the nearby environment, the danger index (DI) modified from the PI was suggested in this study. The DI was calculated by averaging the concentration of the exchangeable fraction, determined by sequential extraction analysis instead of the total concentration. The thresholds of the available metal concentrations extracted by a weak acid (10 µg/g Cu, 3.4 µg/g Pb and 5 µg/g Zn) (Adriano 1986) were used instead of the tolerable level suggested by Kabata-Pendias and Pendias (1984).

Danger Index (DI) =

 $\underline{\sum (\text{conc. of exchangeable fraction/threshold of available metal conc.})}$ Number of elements

A significantly high value of the DI was found in the Daduck and Imchun mines (634.6 and 233.3, respectively), and the other mines ranked in descending order of the DI were Sujum, Dalsung, Duckum, Goobong, Sambo, Gahak, Sangdong, and Ilkwang (Table 4).

The danger indices were compared with the pollution indices (Fig. 4), revealing several trends. The pollution indices of the tailings from the Ilkwang and Sangdong mine were higher than 1.0, but the danger indices were lower than 1.0. This indicated that the tailings from Ilkwang and Sangdong were contaminated in terms of total heavy metal concentration, but their short-term impact on the nearby environment would not be significant.

The PI of the Goobong mine (PI=34.9) was higher than those of the Dalsung (PI=32.8) and Sujum (PI=28.3) mines, but the DI of the Goobong mine (DI=26.4) was lower than those of the Dalsung (DI=67.8) and Sujum (DI=74.1) mines (Fig. 4). In other words, the total concentrations in the tailings of the Goobong mine were higher than those of the Dalsung and Sujum mines, but the fractions of exchangeable ions were smaller in the tailings from the Goobong mine than other mines, indicating that it is more urgent to remediate the Dalsung and Sujum mines before the Goobong mine. Considering the results of the relative metal-binding capacity, the PI, and DI in combination, the priority of remediation was determined to be Daduck, Imchun, Dalsung, and Sujum.

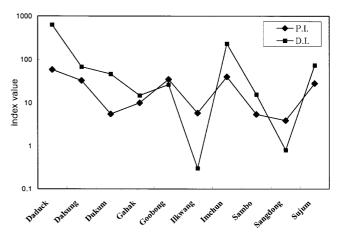


Fig. 4 Comparison between danger index (*DI*) and pollution index (*PI*) of tailings from metal mining areas

Conclusions

- 1. The relative metal-binding capacities based on pH, the clay, and organic contents were very weak in the tailings from the Daduck, Duckum, and Imchun mines, whereas the tailings from the Gahak, Goobong, Sambo, and Sangdong mines had a very strong metal-binding capacity.
- 2. The concentrations of the heavy metals in most tailings were up to one hundred times larger than the tolerable levels, and all the tailings contained heavy metals to the extent of causing phytotoxicity. In particular, the tailings from the Daduck mine contained up to 4% of Zn.
- 3. The greatest proportion of each element was included in the sulfides/organic matter and the residual fractions of the tailings. However, the concentration of the exchangeable ion fraction of Zn in the tailings from the Daduck mine was up to 9,400 μ g/g, and the high level of Zn contamination in the soil and water system around the Daduck mine was expected from this contamination source.
- 4. From the calculation of the relative metal-binding capacity, the PI, and DI, it can be concluded that the Daduck and Imchun mines were in the most urgent need of remediation.

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