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Health Risk Assessment through Residents Exposure to Toxic Metals in Soil and Groundwater in the Vicinity of Sanyang Metal Mine

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Abstract

BACKGROUND: Metal mines were actively developed in the early twentieth century in Korea; however, most of these mines were closed and abandoned without proper management. Therefore, toxic metal contamination in the vicinity of Korean abandoned metal mines has been reported. A risk assessment for these metals was performed for residents near by abandoned Sanyang metal mine.

METHODS AND RESULTS: Soil and groundwater samples were collected from May to October 2007 around the mine. After pretreatment of these samples, metal concentrations were measured and then a risk assessment was performed using the Korean soil-contamination risk assessment guidelines. Cancer risk was the highest from inhalation of Pb-contaminated soil, followed in descending order by As-contaminated soil inhalation and water ingestion. The sum of carcinogenic risks was 3.35×10^{-3}. The noncarcinogenic risk was observed for inhalation of Hg-contaminated soil (5.71).

CONCLUSION: Inhalation of soil in dust was the principal pathway to cause the health risk and most of the risk was attributed to As, Pb,Cd, and Hg contamination.

Key Words: Heavy metals, Mining, Phytoaccumulation, Risk assessment, Soil

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Introduction

Metal mines were actively developed in the early twentieth century in Korea; however, most of these mines were closed and abandoned without proper management in the 1970s because of economic conditions and the exhaustion of ore reserves (Kim et al., 2005). Currently, 936 abandoned metal mines exist in Korea (Susaya et al., 2010), and preliminary investigations for soil contamination in mines have been performed. Recently, precise investigations for approximately 300 mines have been performed over the three years from 2007 to 2009. It has been reported that heavy metal contamination is one of primary environmental problems around the abandoned mines (Susaya et al., 2010). However, risk assessments for those mines have been rarely performed.

The reported risks for humans in the vicinity of several metal mines have been summarized in Table 1. Rice and water ingestion were the primary pathways for As or Cd exposure and posed both carcinogenic and noncarcinogenic health risks to residents in the Dongil (Lee and Chon, 2005; Lee et al., 2006), Okdong (Lee and Chon, 2005), Dongjung (Lee and Chon, 2005), Dokok (Lee and Chon, 2005), Hwacheon (Lee and Chon, 2005), Myungbong (Lee and Chon, 2003), Songchun (Lee et al., 2006), and Songcheon (Lee et al., 2005b; Lim et al., 2008) mine areas. Carcinogenic and noncarcinogenic health risks from As ingestion with soil were observed only in the Songchun (Lee et al., 2006) mine area. Carcinogenic health risk by dermal contact with As-contaminated water was observed only in the Songcheon (Lim et al., 2008) mine area. Farmer may be exposed to dust through agricultural activities, nonetheless, none of these reports considered the pathway of dust inhalation.

Table 1. Abandoned metal mines with carcinogenic and non-carcinogenic risks through exposure pathways of target metals

Pathways		Abandoned metal mines
Soil ingestion	Car (As)	Songchun
	Noncar (As)	Songchun
Water ingestion	Car (As)	Dongil, Okdong, Dongjung, Dogok, Hwacheon, Myungbong, Songchun, Songcheon
	Noncar (As)	Dongil, Okdong, Myungbong, Songchun, Songcheon
	Noncar (Cd)	Songcheon
Rice ingestion	Car (As)	Dongil, Okdong, Hwacheon, Myungbong, Songcheon
	Noncar (As)	Dongil, Okdong, Hwacheon, Myungbong, Songchun, Songcheon
	Noncar (Cd)	Myoungbong
Soil dermal contact		Observed mine is not available
Water dermal contact	Car (As)	Songcheon

Car: carcinogenic risk, Noncar: noncarcinogenic risk, Parenthesis is the risk metal.

The abandoned Sanyang metal mine was selected for an assessment of health risk for residents in the vicinity of the mine. The purpose of this study was to investigate the metal contamination levels of soil and groundwater, to estimate the daily intake of metals, to estimate carcinogenic and noncarcinogenic risks by various metal pathways including inhalation of dust, and to assess the risks for residents in the vicinity of the abandoned Sanyang metal mine following Korean soil-contamination risk assessment guidelines.

Materials and Methods

Investigation of metal contamination

The Sanyang Au-Ag mine is located in Banseok-ri, Boseong-gun, Chonnam in a southwestern province of South Korea. The geology of this mine is classified as Cretaceous granite. Granitic gneiss is composed of biotite, quartz, muscovite, plagioclase, and feldspar. This mine had been developed to exploit gold and silver from 1935 to 1957. After closure of the mine in 1992, tailings and waste rock were left behind without proper environmental treatment. The mining wastes have been dispersed down into paddy fields by water and wind near the mine (Jo, 2001).

Soil samples were collected from agricultural fields and groundwater samples were collected from wells for residents in the vicinity of the mine. All these samples were collected from May to October 2007.

Contaminant analysis of soil was performed according to the Korean Standard Test (KST) Methods for Soils (MOE, 2007). All soil samples were air-dried and sieved to-10 mesh (< 2 mm). Ten grams of sieved soil was extracted with 50 mL of 0.1 Nand 1 N HCl for analysis of Cd, Pb, Cu, and Cr and As, respectively. The extracted solution was filtrated through 5B filter paper (Advantec, Toyo Roshi Kaisha, Japan). For Zn analysis, dried soils were pulverized and sieved to-100 mesh(< 0.15 mm). 3 g of sieved soil was digested with 28 mL of 3:1 HCl/HNO_3 solution (aqua regia) for one hour at 70℃. The extracted or digested solution was filtrated through 5B filter paper, and the concentrations of metals were then measured by inductively coupled plasma-atomic emission spectrometry (ICP-AEX) (2100DV, Perkin-Elmer). Sampled soil was used in a Direct Mercury Analyzer (DMA-80, Milestone) for Hg analysis.

Groundwater used as drinking water was sampled around the mine area. The samples were filtered through a 0.45 µm membrane (PTFE Smart Por, Woongki science, South Korea), then acidified with concentrated HNO_3, and finally the metal concentrations in the water samples were measured using inductively coupled plasma-mass spectrometry (ICPM-8500, Shimadzu, Japan).

Health risk assessment

The purposes of risk assessment are to provide information to decision-makers about the consequences of possible actions, the source treatment and disposal options, the remediation of contaminated sites, and the cleanup standards. The risk assessment consisted of four main analyses: hazard identification, exposure assessment, toxicity (dose-response) assessment, and risk characterization (NRC, 1983).

Hazard identification

To identify hazards, soils and groundwater used for drinking were collected from the vicinity of the mine. The concentrations of metals in those samples were measured.

Exposure assessment

Fig. 1 shows the exposure pathways of contaminated soils to human as expressed in the Korean guidelines (MOE, 2009). Four pathways (groundwater ingestion as drinking water, soil ingestion, and soil contact and dust inhalation through outdoor and indoor air)are suggested for metal exposure in the Korean guidelines. In exposure assessment, the average daily intake (ADI) was calculated by summing the intakes of toxic metals through the suggested pathways. The intake of metals with exposure can be quantitatively calculated using the formula in Table 2. The input parameters in Table 3 were used to determine the ADI values via human exposure pathways such as inadvertent soil ingestion, soil dermal contact, groundwater ingestion, and dust inhalation (MOE, 2009).

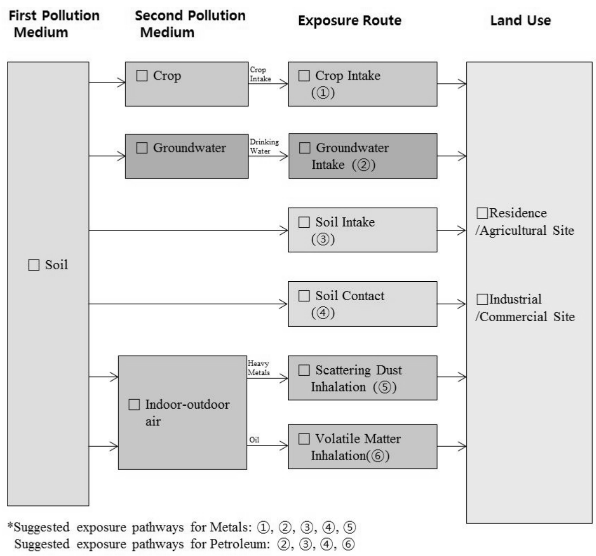

Fig. 1. Suggested exposure pathways of contaminants in soils to humans (MOE, 2009).

Table 2. Equations of average daily intake with different exposure pathways (units in mg/kg-day)

Exposure pathways	Average daily intake
Ingestion of soil	Is = (Cs × IR ×EF × ED)/(BW × AT)
Ingestion of water	Iw = (Cw × IR × EF × ED)/(BW × AT)
Dermal contact with soil	Ids = (Cs × CF × SAe × AF × ABs × EF × ED)/(BW × AT)
Inhalation of dust	Iid = (TSP × frs × CRi × t × tf) × Cs × fr × fa × EF × ED/(BW × AT)

C = concentration of metal in the medium (Cs for soil and Cw for water), and other parameters and factors are as listed in Table 3.

Toxicity (dose-response) assessment

Dose-response relationships for carcinogens are reported as lifetime incidence of cancer (i.e., probability) versus dose. The slope factor (SF) represents the carcinogenic potency of the chemical. Unlike carcinogens, there is a threshold effect for noncarcinogens. In other words, below a specific dose, no adverse health effect occurs in exposed populations. This threshold is defined as reference dose (RfD). Carcinogenic and noncarcinogenic risks for contaminants were estimated using SF and RfD respectively as given in the Korean guidelines (MOE, 2009). As, Cd and Pb are classified as carcinogenic metals, and the other metals are classified as noncarcinogens.

Risk Characterization

Risk characterization is quantitatively expressed by carcinogenic risk and noncarcinogenic risk. Carcinogenic risk may be defined as the average daily intake (calculated in the exposure assessment) multiplied by the carcinogenic slope factor (selected by the toxicity assessment) and can be calculatedusing the following formula:

Carcinogenic risk = I (average daily intake) × SF

where the values of SF for As, Cd and Pb were as given in the Korean guidelines. The estimated value is the probability of an individual's developing any type of cancer from lifetime exposure to carcinogenic chemicals. The total carcinogenic risk equals the sum by exposure route to carcinogenic risks from all individual substances. The acceptable or tolerable total risk for regulatory purposes is in the range of 10^{-6}-10^{-4} (MOE, 2009).

Noncarcinogenic risk is normally characterized in terms of a hazard index. This index is defined as the ratio of the average daily intake from exposure (calculated in the exposure assessment) to the reference concentration (selected by the toxicity assessment) and can be calculated as follows:

Hazard index (HI, non-carcinogenic risk) = I / RfD

where the values of RfD for metals were as given in the Korean guidelines. For exposure to multiple noncarcinogens, the hazard index scores for all noncarcinogens are summed to provide the final measure of the risk for noncarcinogenic adverse effects. If the sum of hazard indices exceeds 1.0, this implies that the contaminants had noncarcinogenic adverse effects.

Results and Discussion

44 soils (32 from paddies and 12 from farmland soils) and two groundwater sources were sampled

from the vicinity of the mine. The mean concentrations of As, Cu, Cu, Pb, Zn, and Hg in these media are shown in Table 4. All metals measured were detected in soils, and the Zn concentration was the highest as 69 mg/kg. As and Zn metals were detected in the groundwater.

Estimated daily metal intake rates from the various media are shown in Table 5. The parameters shown in Table 3, which are listed in the Korean guidelines (MOE, 2009), were used to estimate the daily intake rate. The daily intake rate of metals was the highest for Zn, followed in descending order by As, Pb, Cd, Hg and Cu. The metal intake rate through the various pathways was the highest for inhalation of soil in dust, followed in descending order by groundwater ingestion, soil ingestion, and soil contact.

Table 3. Parameters used for an adult Korean farmer (EPA, 1997; MOE, 2009)

Factor/Parameter	Symbol	Media	Units	Residents
Exposure duration	ED	all	years	Carcinogen-70
				Non-carcinogen-30
Exposure Frequency	EF	all	days/year, events/year	365
Averaging time	AT	all	days	ED×EF
Body weight	BW	all	kg	60
Ingestion rate	IR			
		soil	kg/day	0.0001
		water	L/day	2
Skin area exposed	SAe	soil	cm^2	5700
Adherence factor	AF	soil	mg/cm^2	0.07
Absorption factor	ABS	soil-As	/event	0.03
		soil-Cd	/event	0.001
Total suspended particle	TSP	soil	mg/m^3	
		Indoor		0.07
		outdoor		0.053
Soil fraction in dust	frs	soil	-	
		indoor		0.8
		outdoor		0.5
Inhalation rate	Cri	soil	m^3/day	20
Exposure time/day	t	soil	h/d	8
Exposure ratio	tf	soil	-	
		indoor		2.86
		outdoor		0.143
Retention factor particles in lung	fr	soil		0.75
Relative Absorption factor	fa	soil		1

Table 4. Concentrations of metals in media around the abandoned Sanyang metal mine

Type	Number of Samples	Conc.	As	Cd	Cu	Pb	Zn	Hg
soil	44	mg/kg	5.53±10.0	0.44±2.3	2.19±1.6	3.09±3.9	69.2±37	3.6E-2±0.19
water	2	mg/L	6.5E-3	0	0	0	0.07	0

Table 5. Daily intake water of metals from each medium

Type	Pathway	Unit	As	Cd	Cu	Pb	Zn	Hg	Total
soil	ingestion	mg/kg-d	9.23E-6	7.43E-7	3.65E-6	5.15E-6	1.15E-4	6.0E-8	1.34E-4
	contact		1.10E-6	2.96E-9					1.10E-6
	inhalation		7.55E-2	6.08E-3	0.0	0.042	0.944	4.91E-4	1.07
	subtotal		7.55E-2	6.08E-3	3.65E-6	0.042	0.944	4.91E-4	1.07
water	ingestion		2.17E-4	0	0	0	2.33E-4	0	4.5E-4
	subtotal		2.17E-4	0	0	0	2.33E-4	0	4.5E-4
	Total		7.58E-2	6.08E-3	3.65E-6	0.042	0.944	4.91E-4	1.07

Estimated carcinogenic risks of metals from the various media are shown in Table 6. Slope factors of As and Pb for ingestion of soil and water were given as 1.5 and 0.0085 respectively in the Korean guidelines. The slope factor of As for soil contact was given as 61. The slope factors of As, Cd, and Pb for soil inhalation were given as 0.015, 0.0063, and 0.042 respectively. The cancer risk of metals was the highest for lead (Pb) (1.77×10^{-3}), followed in descending order by As and Cd. Cancer risk by pathway was the highest for soil inhalation, followed in descending order by groundwater ingestion, soil contact, and soil ingestion. Considering both metals and pathways, cancer risk was highest from inhalation of Pb-contaminated soil, followed by inhalation of As-contaminated soil and ingestion of As-contaminated water.

Table 6. Carcinogenic risk estimates via metal exposure pathways around abandoned mines

Type	Pathway		As	Cd	Pb	Zn	Hg	Total
soil	ingestion	SF	1.5		8.5E-3	-	-	
		risk	1.38E-5		4.38E-8			1.39E-5
	contact	SF	61	-	-	-	-	
		risk	6.74E-5					6.74E-5
	inhalation	SF	0.015	6.3E-3	0.042	-	-	
		risk	1.13E-3	3.83E-5	1.77E-3			2.94E-3
	subtotal		1.21E-3	3.83E-5	1.77E-3			3.02E-3
water	ingestion	SF	1.5	-	8.5E-3	-	-	
		risk	3.25E-4					3.25E-4
	subtotal		3.25E-4	0	0	0	0	3.25E-4
	Total		1.54E-3	3.83E-5	1.77E-3	0	0	3.35E-3

※SF (kg-day/mg): Safety Factors are from the Korean guidelines (MOE, 2009)

Carcinogenic risk is the probability of an individual developing any type of cancer from lifetime exposure to carcinogenic chemicals. The acceptable or tolerable total risk for regulatory purposes is in the range of 10^{-6}-10^{-4} (MOE, 2009). Considering 10^{-4} as an acceptable risk, which means that one additional case in 10,000 people is acceptable, there was considerable carcinogenic risk because of As and Pb in this area. The cancer risks frominhalation of Pb-contaminated soil, inhalation of As-contaminated soil, and ingestion of As-contaminated groundwater exceeded the acceptable risk. However, the risks from ingestion of soil and contact with soil did not exceed the acceptable risk. The sum of all carcinogenic risks was 3.34×10^{-3}. This value is equivalent to the probability that approximately three cancers per 1,000 people will occur. Most of the risk was due to Pb and As contaminations, and therefore, Pb and As were considered to be the principal carcinogens for humans in this area.

It has been reported that the residents around the following mines have been exposed to carcinogenic health threats: Dongil, Okdong, and Hwacheon mines due to ingestion of As-contaminated water and soil (Lee and Chon, 2005); Dongil, Dongjung, Myungbong, and

Songchun mines due to ingestion of As-contaminated water (Lee *et al*., 2006); Myungbong mine due to ingestion of As-contaminated rice(Lee *et al*., 2008); Songchun mine due to ingestion of As-contaminated soil, water, and crops (Lim *et al*., 2008); and Okdong and Hwacheonmines due to ingestion of As-contaminated rice and water (Lee *et al*., 2005a). Carcinogenic risk in the Koran abandoned mine area by ingestion of contaminated soil, water, and crops has been reported as mentioned above; however, the risk of As and Pb contamination by inhalation of contaminated soil has not been reported up to now. In this study, exceeded cancer risk was observed from inhalation of Pb-and As-contaminated soil in abandoned Sanyang mine. The risk of inhalation of metal-contaminated soil should be considered in a carcinogenic risk assessment along with ingestion of water and crops in abandoned mine areas.

Estimated noncarcinogenic risks of metals from the various media studied are shown in Table 7. The reference dose (RfD) values of As, Cd, Pb, Zn, and Hg for intake from soil and water were given in the Korean guidelines. The noncarcinogenic risk (hazard index, or HI) of metals was the highest for mercury (Hg), with a value of 5.71, followed in descending order by As, Pb, Cd, and Zn. By pathway, the highest noncarcinogenic risk was in the inhalation of soil (5.71), followed in descending order by ingestion of water, ingestion of soil, and contact with soil. Considering both metals and pathways, the risk was the highest for inhalation of Hg-contaminated soil (5.71), followed in descending order by ingestion of As-contaminated water (0.72) and ingestion of As-contaminated soil (3.08×10^{-2}).

Table 7. Noncarcinogenic risk estimates via metal exposure pathways around abandoned mines

Type	Pathway		As	Cd	Cu	Pb	Zn	Hg	Total
soil	ingestion	RfD	3.0E-4	5.0E-4	-	5.0E-4	0.3	3.0E-4	
		HI risk	3.08E-2	1.49E-3		1.03E-2	3.84E-4	2.0E-4	4.31E-2
	contact	RfD	3.0E-4	5.0E-4	-	5.0E-4	0.3	2.1E-5	
		HI risk	3.81E-3	2.28E-4		0	0	0	4.04E-3
	inhalation	RfD	-	-	-	-	-	8.6E-5	
		HI risk	0	0		0	0	5.71	5.71
	subtotal		3.46E-2	1.71E-3		1.03E-2	3.84E-4	5.71	5.75
water	ingestion	RfD	3.0E-4	5.0E-4	-	5.0E-4	0.3	3.0E-4	
		HI risk	0.722	0		0	7.78E-4	0	0.723
	subtotal		0.722	0		0	7.78E-4	0	0.723
	Total		0.757	1.71E-3	0	1.03E-2	1.16E-3	5.71	6.48

※RfD (mg/kg-day): Reference Dose values are from the Korean guidelines (MOE, 2009)

The HI values for As, Cd, Pb, and Zn for each pathway were less than one, and their sum was also less than one (Table 7). Therefore, the noncarcinogenic risk from these metals in this area is not substantial. However, the HI values for Hg for all pathways were 5.71. This value is significantly greater than one, and its noncarcinogenic risk is therefore high and substantial for the residents in the vicinity of the abandoned Sanyang mine. The highest noncarcinogenic risk is in the inhalation of Hg-contaminated soil in this area. However, the HI values for ingestion of water, ingestion of soils and contact with soils were less than one.

It has been reported that the residents in the following mines have been exposed to noncarcinogenic health threats: Dongil, Okdong, Dongjung, and Hwacheon due to ingestion of As-contaminated water and soil (Lee and Chon, 2005); Duckum, Dongil, Dongjung, and Songchun mines due to the ingestion of As-contaminated groundwater (Kim *et al*., 2005); Songchun mine due to the ingestion of As-contaminated water (Lee *et al*., 2006); Myungbong mine due to the ingestion of As-contaminated rice (Lee *et al*., 2008); Songchun mine due to the ingestion of As-contaminated soil, water, and crops (Lim *et al*., 2008); Okdong and Hwacheon mines due to the ingestion of As-contaminated rice and water; and Dokok mine due to the ingestion of Cd-contaminated groundwater (Lee *et al*., 2005a). A noncarcinogenic risk of As or Cd contamination in the area of the Korea abandoned mine by ingestion of water and crops has been reported as mentioned above; however, the risk of Hg by inhalation of contaminated soil has not been reported until now. The risk of inhalation of metal-contaminated soil should be considered for noncarcinogenic risk assessment along with ingestion of water and crops in abandoned mine areas.

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