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HUMAN RISK ASSESSMENT FOR HEAVY METALS AND As CONTAMINATION IN THE ABANDONED METAL MINE AREAS, KOREA

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Abstract. Cleanup goals for the contaminated sites are established on the basis of risk assessments and rely on the estimated toxicity of the chemicals of concern (COC). Toxicity estimates are based on bioavailability causing risk of adverse health effects on humans. In this study, bioavailability of As, Cu, Pb and Zn in soil was determined by SBET (Simple Bioavailability Extraction Test), and chemical analysis for groundwater and stream water collected from the abandoned mine areas (Dukeum, Dongil, Dongjung, Myungbong and Songchun mine areas) was conducted. High values of cancer risk for As (1.16×10^{-5}) were detected through soil ingestion pathways in the Songchun mine area and assessed through water exposure pathways in the all mines except Dukeum. The hazard index value for As in the Songchun mine area (3.625) exceeded 1.0. The results indicated that the ingestion of As-contaminated soil and water by local inhabitants can pose a potential health threat in these mine areas.

Keywords: arsenic, bioavailability, risk assessment, SBET

1. Introduction

The high level of contamination caused by toxic metals and metalloids is a persistent problem in many mine areas in Korea. Toxic contaminants in the tailings and waste rocks can be released to the stream in the farming area and accumulated in crop plants of the mine areas. It may finally become a significant threat to human health (Davies, 1983). Risk assessment of As and heavy metals for soil environments is often based on total soil concentrations and does not take the actual degree of exposure into account (Crommentuijn *et al.*, 2000). In order to investigate environmental and health risks caused by the contaminants, understanding the bioavailability of contaminants in the soil is required (Ma and Rao, 1997; Mester *et al.*, 1998). Through the substitution of total concentration with bioavailable concentration, risk calculations proved more realistic, thus facilitating an improved cost-benefit analysis of site remediation options (Williams *et al.*, 1998). Unfortunately, the routine estimation of toxicity through total element concentration measurement cannot provide such detailed information. Recently, many scientists have developed extraction tests to estimate the bioavailability of metal from soil to present the quantitative relationship between contamination level and human health (Ruby *et al.*, 1992, 1993, 1996, 1999; Hamel *et al.*, 1998, 1999; Rodriguez and Basta, 1999; Rodriguez *et al.*, 2003; Sarkar and Datta, 2003). The risk assessment using the simple bioavailability extraction test (SBET) based on simulating digestion of the human stomach can account for the risk of the characteristics of a metal in soil. The objectives of this study are (1) to investigate the contamination levels of As and heavy metals, (2) to assess the bioavailability of toxic metals using SBET analysis, and (3) to estimate the risk of adverse health effects on inhabitants around the mine sites.

2. Material and Methods

2.1. Sampling and chemical analysis

The tailings, agricultural soils (paddy and farmland soil) and water (tailing dump leachate, groundwater, and stream water) were collected around the mine areas in Korea (Dukeum, Dongil, Dongjung, Myungbong and Songchun). An approximate 20 kg composite tailings and soil samples were collected using a hand auger at depths of 15 cm. Each tailings and soil samples were composed of approximately 20 subsamples. The all samples were transferred to plastic-lined canvas bags, and transported to the laboratory. Tailings and soil samples were air-dried, disaggregated, and sieved through a –80 mesh sieve ($<180\ \mu$m). The total concentrations of As and heavy metals (Cu, Pb and Zn) were determined by aqua regia (HNO_3 1 mL + HCl 3 mL) to investigate contamination levels. Water samples were stored at 4 °C after acidifying with HNO_3. All the samples were analyzed by ICP-AES and ICP-MS (Thermo Jarrell) for heavy metals (Cu, Pb and Zn) and HG-AAS (5100, Perkin-Elmer) for As.

2.2. Risk assessment procedures

The risk assessment model developed by NRC (National Research Council) and NAS (National Academy of Sciences) was generally used to estimate the risk caused by contaminants in the sites. The risk assessment can be followed by four main steps; (1) hazard identification, (2) exposure assessment, (3) dose-response assessment and (4) risk characterization (NRC/NAS, 1983). The procedure of risk assessment was summarized in Figure 1.

Hazard identification is to estimate the hazardous effect of contaminant. In the first step, the hazard level is examined by physical and chemical properties of contaminants such as mobility and contaminant levels in the point of exposure where the contaminants is exposed to environment. The second step is exposure assessment estimated by average daily dose (ADD) using the identification of

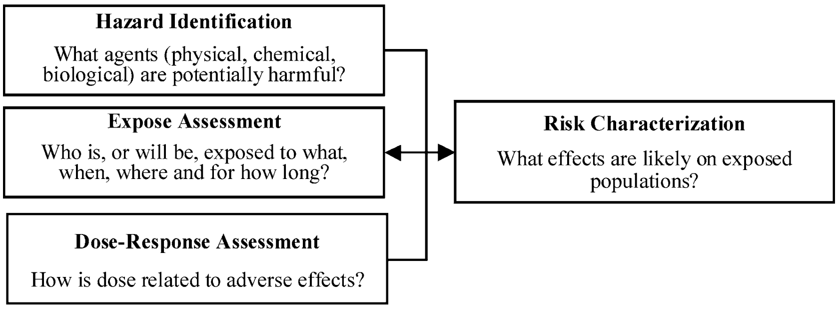

Figure 1. Procedure of risk assessment model (NRC/NAS, 1983).

intensity, frequency, exposure period, and pathway of contaminants. In the third step, dose-response assessment examines the relationship between adverse effects and exposure levels of carcinogenic and non-carcinogenic chemicals. In addition, it prospects an attack of disease caused by exposed contaminants to humans. The two principal toxicity indices are known as SF (cancer slope factor) and RfD (reference dose). The information of SF and RfD was based on toxicity data of contaminants from (1) EPA Integrated Risk Information System (IRIS) on-line database, (2) EPA Health Effects Assessment Summary Tables (HEAST) and (3) EPA Environmental Criteria and Assessment Office (ECAO). Risk characterization is the final step that predicts the level of risk. The results of exposure assessment and dose-response assessment are integrated to derive quantitative estimates of cancer risk and hazard index.

2.2.1. *Hazard Identification*

In order to identify the hazard of mine areas, the concentrations of As and heavy metals in soils and water (stream water and groundwater) were determined. Bioavailable concentrations of As and heavy metals in soils (paddy and farmland soils) were measured by the simple bioavailability extraction test (SBET) modified from PBET (Physiologically Based Extraction Test) suggested by Ruby (1993, 1996, 1999). The SBET method simulates mobilization of contaminants in the artificial gastric fluids (acidic conditions of the stomach). SBET results indicate the amount of contaminants which can be absorbed through the ingestion of soils. The procedure of SBET was shown in Figure 2.

In addition, the concentrations of As and metals for water samples (groundwater and stream water) collected near mine areas were determined using ICP-MS and HG-AAS.

2.2.2. *Exposure Assessment*

2.2.2.1. Exposure Factors and Pathways. In exposure assessment, the average daily dose (ADD) was quantified by the intake of toxic metals through several pathways such as soil and water. Exposure factors for ADD were driven from the

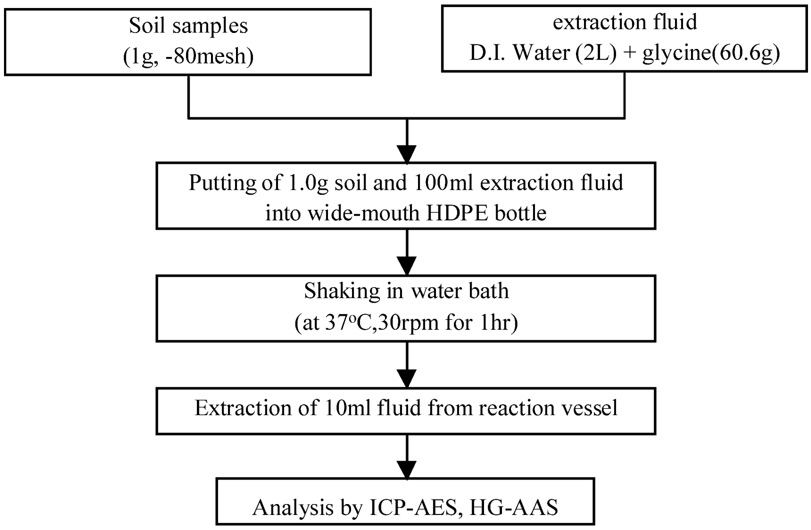

Figure 2. Flowchart showing the procedure of SBET analysis.

report of physical condition and the exposure factors handbook (EPA, 1997). The exposure factors and the input parameters of Koreans farmers to calculate ADD were summarized Table I.

The major exposure pathways for the intake of toxic metals caused by the usage of contaminated water as drinking water and soil ingestion by the agricultural activity and bad hygiene. To calculate ADD of soil ingestion, absorbed amounts of toxic metals were based on the SBET results for paddy and farmland soils. The intake rate of water pathway (stream water and groundwater) was derived from the

TABLE I
Exposure factors and input parameters of Korean farmer

Factor/parameter	Symbol	Units	Residential/ Agricultural	Data source
Exposure duration	ED	years	30	US EPA (1997)
Exposure frequency	EF	Soil (days/year)	210	US EPA (1997)
		water (days/year)	350	US EPA (1997)
Averaging time				
Carcinogens	ATc	years	76.5	KNSO (2001)
Non-carcinogens	ATnc	years	30	US EPA (1997)
Body weight	BW	kg	60	MOCIE (1997)
Ingestion rate				
Soil	IRs	kg/day	100×10^{-6}	US EPA (1997)
Drinking water	IRw	L/day	2.0	KOWACO (2001)

report of Korea Water Resources Corporation.

The ADD of exposed contaminants can be quantitatively calculated by the following numerical formula:

$$\text{ADD} = (\text{C} \times \text{IR} \times \text{ED} \times \text{EF})/(\text{BW} \times \text{AT} \times 365)$$

C = Concentration of contaminant in the environmental sample (e.g. soil, water, air in mg/kg, mg/L, mg/m^3, etc.); IR = Ingestion rate per unit time (kg/day, L/day); ED = Exposure duration (years); EF = Exposure frequency (days/year); BW = Body weight of the receptor (kg); AT = Averaging time (years).

2.2.3. *Dose-Response Assessment*

In order to estimate the carcinogenic and non-carcinogenic risks, dose-response assessment was carried out using slope reference dose (RfD) and factors (SF) driven from IRIS database (EPA, 1997). Table II shows RfD and SF values of As and Zn due to lack of reference dose (RfD) information.

2.2.4. *Risk Characterization*

Risk characterization is quantitatively presented by carcinogenic risk and non-carcinogenic risk. Carcinogenic risk can be estimated by following formula.

$$\text{Cancer risk} = \text{ADD (average daily dose)} \times \text{SF (slope factor)}$$

The estimated value is the probability of an individual developing any type of cancer from lifetime exposure to carcinogenic hazards. The acceptable or tolerable risk for regulatory purposes is in the range of 10^{-6}–10^{-4}.

To determine quantification of non-carcinogenic risks, the hazard quotient (HQ) is estimated by comparing exposure or average intake of hazardous substances with the corresponding reference dose. The hazard index can be estimated by summarizing the hazard quotient. If the hazard index exceeds 1.0, it implies that the contaminants revealed toxicity.

$$\begin{aligned} \text{HQ} &= \text{ADD (from exposure assessment)}/\text{RfD (from IRIS of US EPA)} \\ \text{HI} &= \sum \text{HQs (sum of hazard quotients)} \\ &= \{\text{ADD}_1/\text{RfD}_1 + \text{ADD}_2/\text{RfD}_2 + \cdots + \text{ADD}_i/\text{RfD}_i\} \end{aligned}$$

TABLE II

Reference doses and slope factors of As and Zn obtained from US-EPA IRIS database

Element	RfD (mg/kg-day)	SF $(\text{mg/kg-day})^{-1}$
As	3×10^{-4}	1.5
Zn	3×10^{-1}	n.a.

3. Results and Discussion

3.1. Contamination levels of As and heavy metals

The concentrations of As and heavy metals in the tailings and soils (paddy and farmland soils) in the mine areas (Dukeum, Dongil, Dongjung, Myungbong and Songchun mine) were shown in Table III. As in the tailings, farmland and paddy soils of all mines significantly exceeded average values for the world's normal soil suggested by Bowen (1979). Especially, As concentrations of As in the tailings of Dongil, Myunbong and Songchun mine were 5,505 mg/kg and 3,012 mg/kg, 113,560 mg/kg, respectively. These results indicate that As is seriously contaminated in the mine areas and can act as a potential risk in the study areas.

Contamination levels of Zn in tailings, farmland and paddy soils in all throughout the mine were also high in comparison of average values. However, those of Cu and Pb in all of the samples except tailings of Songchun mine were similar to the average values (70 Cu mg/kg and 30 Pb mg/kg).

The concentrations of As and heavy metals in water samples (leachate, groundwater and stream water) from each of the mine areas were shown in Table IV. As concentrations of water samples in the Dukeum, Dongil, Dongjung, Myungbong mine areas did not exceed 50 μg/L and concentration of heavy metals was also lower than the permissible levels of drinking water regulated by Ministry of Environment, Korea (1.0 Cu mg/L, 0.05 Pb mg/L, 1.0 Zn mg/L). However, the average concentration of As in leachate and groundwater of the Songchun mine was 371 and

TABLE III

Concentration of As and heavy metals in tailing and soils of the mine areas (unit in mg/kg_{soil})

Mine	Type (Num.)	As	Cu	Pb	Zn
Dukeum mine	Tailings (1)	74.0	76.7	38.7	309
	Farmland soil (2)	68.4–131 (99.6)	49.6–50.3 (49.9)	2.0–11.3 (6.7)	153–322 (237)
	Paddy soil (2)	15.3–49.8 (32.5)	84.4–199 (141)	183–418 (300)	399–400 (400)
Dongil mine	Tailings (1)	5505	84.1	1.2	100
	Farmland soil (2)	23.8–32.3 (28.1)	16.8–17.7 (17.2)	7.0–10.6 (8.8)	96.6–384 (241)
	Paddy soil (2)	25.6–26.0 (25.8)	18.8–25.2 (22.0)	19.6–27.2 (23.4)	102–117 (109.5)
Dongjung mine	Tailings (3)	44.6–91.2 (71.3)	55.2–183 (135)	167–590 (317)	182–399 (309)
	Farmland soil(2)	4.1–24.6 (14.3)	13.6–152 (82.7)	0.4—184 (92.3)	37.5—231 (134)
Myungbong mine	Tailings (2)	2673—3351 (3012)	35.9–50.5 (43.2)	107–113 (110)	135–144 (140)
	Farmland soil (1)	30.4	16.6	11.8	66.9
	Paddy soil (2)	84.7–130 (107)	20.0–23.2 (21.6)	12.2–21.1 (16.6)	73.5–85.5 (79.5)
Songchun mine	Tailings (1)	113,560	163.5	19,075	513.3
	Farmland soil (2)	25.2–368 (196)	15.5–174 (94.7)	21.1–129 (75.0)	115–363 (239)
World's normal soil (average)[a]		6.0	70	30	35

[a]Bowen (1979).

TABLE IV
Concentration of As and heavy metals in groundwater and stream water in the selected mine areas (unit in μg/L)

Mine	Type (Num.)	As	Cu	Pb	Zn
Dukeum mine	Leachate (3)	1.3–2 (1.7)	29.3–114 (66.3)	96.5–610 (271)	7347–2346 (3676)
	Stream water (3)	0.2–0.5 (0.4)	0.7–15.9 (6.2)	0.4–3.7 (1.6)	11.1–23.3 (18.8)
	Groundwater (3)	0.3 (0.3)	3.0–12.4 (6.9)	0.4–0.7 (0.6)	137–166 (148)
Dongil mine	Leachate (1)	34.1	116	0.6	342
	Stream water (1)	9.3	18.1	0.4	12.6
	Groundwater (3)	8.0—15.2 (12.8)	1.6–22.9 (12.3)	0.3–0.7 (0.4)	10.2–37.9 (26.1)
Dongjung mine	Leachate (2)	2.6–2.7 (2.7)	3.2–4 (3.6)	0.6–1.6 (1.1)	236–9234 (4735)
	Stream water (2)	4.0–6.2 (5.1)	2.2–2.5 (2.4)	1.1 (1.1)	225–2819 (1522)
	Groundwater (5)	1.5–3.9 (2.7)	0.6–1.9 (1.3)	0.1–1.0 (0.5)	3.1–17.9 (7.9)
Myungbong mine	Leachate (1)	17.2	14.2	0.3	9.2
	Stream water (3)	1.9–13.3 (6.3)	3.1–5.6 (4.4)	0.0–0.3 (0.1)	0.0–20.1 (11.5)
	Stream water (6)	0.4–5.7 (2.5)	1.3–5.6 (3.2)	0.0–0.4 (0.2)	0.0–30.7 (13.2)
Songchun mine	Leachate (2)	223–518 (371)	0.0–47.2 (23.6)	0.0–0.5 (0.3)	9.1–58.5 (33.8)
	Stream water (4)	0.6–36.2 (11.3)	7.0–14.3 (7.4)	0.0–0.3 (0.2)	2.8–15.8 (7.4)
	Groundwater (6)	1.1–220 (86.1)	0.0–16.2 (9.9)	0.0–0.3 (0.2)	1.5–41 (16.3)

86.1 μg/L, respectively. These results suggested that the contamination of leachate and groundwater was affected by tailings containing high level of As.

3.2. Human health risk assessment

3.2.1. *Bioavailablity of As and Heavy Metals*

Recently, researchers have developed in-vitro test methods to measure the fraction of a chemical solubilized from soils under simulated gastrointestinal conditions (Imber, 1993; Ruby *et al.*, 1993, 1996, 1999; Medlin, 1997; Williams *et al.*, 1998). In this experiment, the SBET (simplified PBET) was applied to determine the bioavailable fractions of contaminant through the ingestion of soils.

From the results of the SBET, the highest concentration of As extracted was 20.6 and 12.4 mg/kg in farmland soil and paddy soil of the Songchun mine, respectively (Figure 3). The average Cu value was the highest in farmland soil (7.9 mg/kg) from the Dukeum mine and paddy soil from the Dongil mine (24.1 mg/kg). In the case of Pb and Zn, the highest values were found in farmland soil (75.0 and 94.7 mg/kg) from the Songchun mine and paddy soil (300.6 and 141.4 mg/kg) from the Dukeum mine.

In comparison between total concentration and bioavailability of As and heavy metal in each mine, the bioavailability of As, Cu and Zn value which can be digested by human beings were much lower than the total concentration. However, the bioavailabilty of Pb in farmland and paddy soil was similar to total concentration in all the mines.

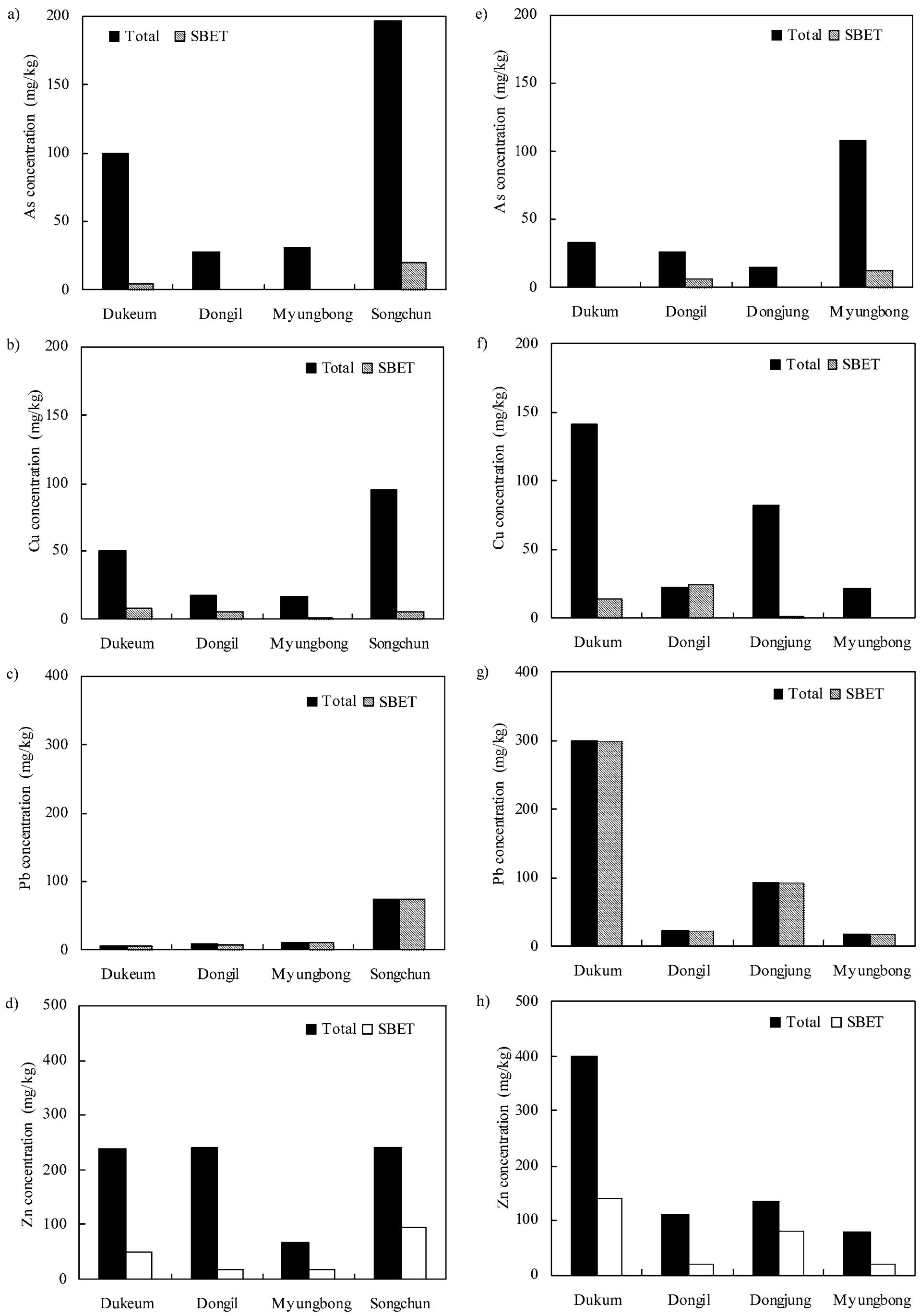

Figure 3. Concentration of As and heavy metals by the SBET in soils from the mine areas (farmland soil: a, b, c, d and paddy soil: e, f, g, h).

TABLE V
Average daily dose (ADD) of As and Zn through soil pathway and water pathway

Mine	Pathway	As	Cu	Pb	Zn
Dukeum mine	Soil pathway	1.76×10^{-6}	7.53×10^{-6}	6.39×10^{-6}	4.79×10^{-5}
	Ground pathway	3.76×10^{-6}	2.21×10^{-4}	1.92×10^{-5}	4.73×10^{-3}
Dongil mine	Soil pathway	1.06×10^{-6}	2.88×10^{-5}	1.46×10^{-5}	1.88×10^{-5}
	Ground pathway	1.60×10^{-4}	3.93×10^{-4}	1.28×10^{-5}	8.34×10^{-4}
Dongjung mine	Soil pathway	1.25×10^{-7}	1.20×10^{-6}	8.85×10^{-5}	7.93×10^{-5}
	Ground pathway	3.38×10^{-5}	4.16×10^{-5}	1.60×10^{-5}	2.53×10^{-4}
Myungbong mine	Soil pathway	2.34×10^{-6}	4.47×10^{-7}	1.36×10^{-5}	1.83×10^{-5}
	Ground pathway	3.13×10^{-5}	1.02×10^{-4}	6.39×10^{-6}	4.22×10^{-4}
Songchun mine	Soil pathway	7.74×10^{-6}	5.35×10^{-6}	7.19×10^{-5}	9.08×10^{-5}
	Ground pathway	1.08×10^{-3}	3.16×10^{-4}	6.39×10^{-6}	5.21×10^{-4}

3.2.2. *Exposure Assessment*

The ADD of exposure pathways in the Dukeum, Dongil, Dongjung, Myungbong and Songchun mine areas was shown in Table V.

In the case of soil pathways, the ADD of As was determined in the order of Songchun > Myungbong $\cong$ Dukeum $\cong$ Dongjung > Dongil mines. The ADD of Cu showed in the order of Dongil > Dukeum $\cong$ Songchun > Dongjung > Myungbongmine and the ADD for Pb and Zn was similar in all the mines.

In the case of water pathways, the highest exposed dose of As was shown in the Songchun mines in comparison with other mines because groundwater was seriously contaminated with As.

3.2.3. *Carcinogenic Risk*

Carcinogenic risk for As was only yielded by average daily dose due to lack of slope factor (SF) information (Figure 4). The acceptable carcinogenic risk is 1×10^{-5}, indicating that cancer can be caused by the ingestion of contaminated soil and water.

Carcinogenic risk of As through soil ingestion pathway in the Songchun mine (1.16×10^{-5}) is higher than the acceptable risk. In addition, the ingestion of water from all the mine areas except Dukeum mine area can cause serious health problems. These results indicate that the ingestion of soil and water over a long period of time may be harmful to human health, increasing the probability of cancer.

3.2.4. *Non-Carcinogenic Risk*

In order to quantify non-carcinogenic risk, the hazard quotient (HQ) was estimated by using average dairy dose (ADD) and reference dose (RfD). The hazard index can be estimated by summarizing hazard quotient (Figure 5).

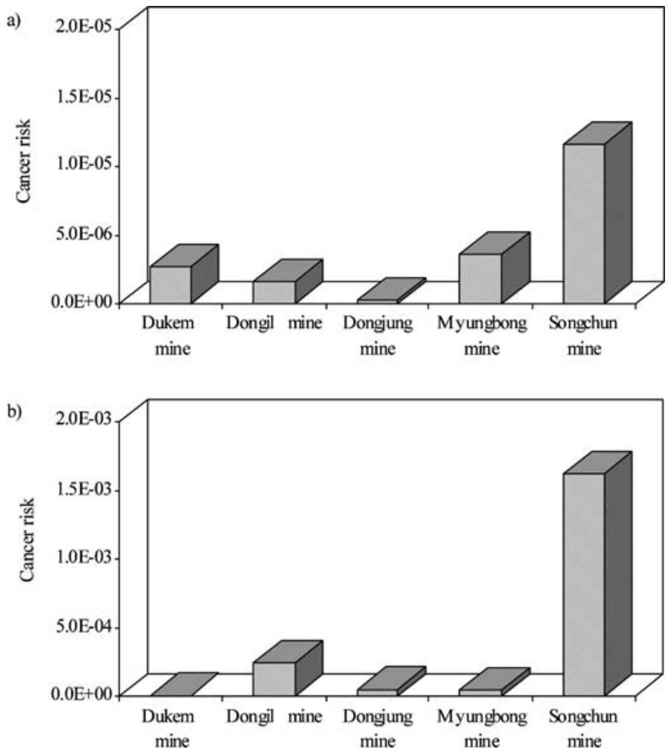

Figure 4. Carcinogenic risk of As through (a) soil pathway and (b) groundwater pathway in the Dukeum, Dongil, Dongjung, Myungbong and Songchun mine.

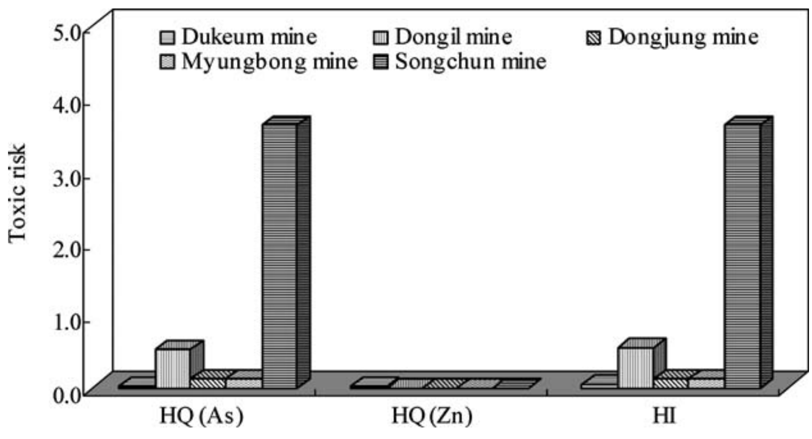

Figure 5. Hazard index (hi) for As and Zn in the Dukeum, Dongil, Dongjung, Myungbong and Songchun mine areas.

The hazard indexes of all mine areas except the Songchun mine were lower than 1.0. If the hazard index exceed 1.0, it may imply that toxicity can be appeared by contaminants. From the results of hazard index, it is verified that As is the main contaminant, and toxic human risk can be caused by ingestion of soil and water in the Songchun mine area.

4. Conclusions

The concentrations of As and heavy metals in the tailings from the study area were significantly high, and the contamination level of As in the paddy and farmland soils was higher than the average value of world's normal soil. Mine wastes such as tailings may act as sources of As and heavy metals near the mine area. The results of cancer risk using data from SBET showed that high values of cancer risk were detected from soil pathway of the Songchun mine area (1.16×10^{-5}) and from water pathway of all mines except Dukeum mine. The hazard index for As in the Songchun mine was 3.625, indicating largely exceeded 1.0. In conclusion, contaminants from the ingestion of soils and water can cause toxic risk for the inhabitants of the Songchum mine areas. Therefore, remediation should be applied to prevent harmful effects of the health of the inhabitants.

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