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Evaluation of human exposure to arsenic due to rice ingestion in the vicinity of abandoned Myungbong Au–Ag mine site, Korea

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Abstract

In order to assess the risk of adverse health effects on human exposure to arsenic (As) influenced by past mining activities an environmental geochemical survey was undertaken in the abandoned Myungbong gold–silver (Au–Ag) mine area. Elevated levels of As were found in tailings from the studied mine area. This high concentrations may impact on agricultural soils and waters around the tailing piles. The resulting human health risks to farmers who inhabit the surrounding areas due to consumption of rice grown on contaminated soils are summarized as follows: (1) The non-cancer health hazard index showed that the toxic risk due to As was 7.8 times greater than the reference dose. (2) The increased cancer risk of As for exposed individuals through the rice consumption pathway was 1 in 1 000, exceeding the acceptable risk of 1 in 10,000 set for regulatory purposes. Thus, the daily intake of rice by the local residents from the Myungbong mine area can pose a potential health threat due to long-term As exposure.

Keywords: Arsenic; Risk assessment; Exposure pathway; Rice ingestion; Hazard index; Cancer risk

1. Introduction

Mining can be a significant source of As and heavy metal contamination of the environment owing to activities such as mineral exploitation, ore transportation, smelting and refining, disposal of the tailings and waste waters around mines (Adriano, 2001). In typical metal mine districts, massive stockpiles of sulfide containing refuse and tailings in the inactive mines are weathered and oxidized due to long-term atmospheric exposure. The acidic mine drainage, with elevated levels of heavy metals, are discharged to contaminate the downstream water bodies, and subsequently agricultural soils and food crops. The fugitive metals in the receiving water and soil may pose a potential health risk to the residents in the vicinity of the mines (Davies and Ballinger, 1990; Merrington and Alloway, 1994). There is a need to accurately quantify the toxicological risk to the resident populations in the contaminated environments. Current assessment models derive the total human exposure (Kolluru et al., 1996; Kimmel et al., 1999; Akagi et al., 2000; Alcock et al., 2000; Green et al., 2000; Lee et al., 2000; Paustenbach, 2002; Sekhar et al., 2003; Lee et al., 2004) by evaluating the fate and transport of toxic elements through exposure pathways such as drinking water, food intake, dust inhalation and hand-to-mouth soil ingestion.

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In this study, to assess the risk of adverse health effects on human exposure to As by past mining activities, an environmental geochemical survey was undertaken in the abandoned Myungbong Au–Ag mine area in Korea.

2. Materials and methods

2.1. Sampling and chemical analysis

The Myungbong Au–Ag mine is located in Myungbong-ri, Nodong-myun, Boseong-gun, Chonnam, southwest Province of South Korea. The deposit of this mine was classified as a hydrothermal vein type with Au–Ag minerals in quartz veins and the main geology is Cretaceous granite. Sulfide containing minerals consist of arsenopyrite (FeAsS), pyrite (FeS_2), galena (PbS) and sphalerite (ZnS). The mine was active from 1916 to 1988 and upon closure large amounts of tailings (10,950 m^2) were left behind without proper environmental safeguards. The unprotected mining wastes have acted as a point source of contamination and have been dispersed down the slope by wind and water in the vicinity of this mine.

Sampling of tailings, soils, groundwaters and rice grains in the vicinity of the Myungbong mine was carried out in September, 2005. Surface soil samples (0–15 cm depth) were collected from the agricultural land around the mine area. Each soil comprised a composite of 15–20 subsamples taken across a 1×1 m square. Tailing and soil samples were air-dried, sieved to −10 mesh (< 2 mm), quartered and pulverized into −80 mesh (< 180 μm) for the chemical analysis. Two grams of each soil sample was digested with aqua regia (HNO_3/HCl) and the samples were then heated at 70 °C for 1 h. The total As concentrations in the soil extracts were determined by HG-AAS (5100, Perkin-Elmer).

To prepare the rice samples, rice grains were separated from rice plants. Each sample was thoroughly rinsed with tap water followed by deionized water in an ultrasonic cleaner for 5 min then a final rinsing step with deionized water. After air-drying at room temperature, rice brans were removed and the samples ground with a smaller mill until homogeneous. A 0.5 g of each rice sample was digested in reagents consisting 65 % of HNO_3 (5 ml), H_2O (5 ml), and H_2O_2 (1 ml) and the mixture gently swirled and placed in microwave digestion system (Milestone, Ethos plus). After digestion the solution was filtered and As was measured by inductively coupled plasma-mass spectrometry (ICP-MS).

Waters used as drinking water were filtered through 0.45 μm membrane and then samples were acidified with concentrated HNO_3 for analyses of the cations using the ICP-MS.

2.2. Risk assessment process

The US National Academy of Sciences (NRC, 1983) defines risk assessment as the process of estimating the probability of the occurrence of an event and the probable magnitude of adverse health effects on human exposures to environmental hazards (Kolluru et al., 1996; Paustenbach, 2002). A fully fledged risk assessment consists of four interactive and iterative steps, namely the hazard identification, exposure assessment, dose–response (toxicity) assessment and risk characterization. The basic frame-work for risk assessment was adopted for assessing the health risk to an adult farmer who resides in the affected area and is exposed to As present in the contaminated soils and waters. The hazard identification process was accomplished through field sampling of the tailings, soils, waters and crop plants, and the subsequent determination of the contaminant level of As in these samples.

2.2.1. Exposure assessment

The exposure assessment identifies the pathways by which humans are potentially exposed the toxicants and estimates the magnitude, frequency and duration of these actual and/or potential exposures. Conducting an exposure assessment involves analyzing contaminant releases; identifying exposed populations; identifying all potential pathways of exposure; estimating exposure point concentrations for specific pathways; and estimating contaminant intakes for specific pathways.

The average daily dose (ADD) of the contaminant via the identified pathways (i.e. soil ingestion, rice grain and drinking water pathways) indicates the quantity of chemical substance ingested per kilogram of body weight per day (Kolluru et al., 1996; Paustenbach, 2002) that:

$$\mathrm{ADD} = \frac{C \times \mathrm{IR} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT} \times 365} \tag{1}$$

Where C is the concentration of the contaminant in the environmental media (mg/kg or mg/L), IR is the ingestion rate per unit time (mg/day or L/day), ED is the exposure duration (years), EF is the exposure frequency (days/year), BW is the body weight of the receptor (kg), and AT is the averaging time (years), equal to the life expectancy for carcinogen, and 365 is the conversion factor from year to days. The principal exposure factors that have been taken into account to carry out the risk assessment calculations are shown in Table 1.

2.2.2. Dose–response assessment

In order to estimate the carcinogenic and non-carcinogenic risks, a dose–response assessment was

Table 1
Exposure factors for an adult Korean farmer

Factor/parameter	Symbol	Units	Residential/agricultural	Data source
Exposure duration	ED	years	30	US EPA, 1997
Exposure frequency	EF	days/year	350	US EPA, 1997
Averaging time	AT			
Carcinogens	AT_C	years	76.5	KNSO, 2001
Non-carcinogens	AT_{NC}	years	30	US EPA, 1997
Body Weight	BW	kg	60	MOCIE, 1997
Ingestion rate				
Soil	IR_S	kg/day	100×10^{-6}	US EPA, 1997
Drinking water	IR_W	L/day	2.0	US EPA, 1997
Rice grain	IR_r	kg/day	0.358	KNSO, 2005

carried out using reference dose (RfD) and slope factor (SF) derived from the IRIS (Integrated Risk Information System) database (US EPA, 1997). Table 2 shows RfD and SF values for As.

2.2.3. Risk characterization

Toxic risks refer to the non-carcinogenic harms incurred due to the exposures and the extent of the harm is indicated in terms of a hazard quotient (HQ) that:

$$HQ = ADD/RfD \tag{2}$$

The reference dose is the daily dosage that enables the exposed individual to sustain this level of exposure over a prolonged time period without experiencing any harmful effect. Toxic risk estimates are based on a comparison of actual exposure to the reference dose for the particular chemical involved. The RfD for chemicals is derived from toxicological data. When more than one potential toxicant or exposure pathway is present, the interactions must be considered. The HQs may then be summed to arrive at the overall toxic risk, the hazard index (HI) (Kolluru et al., 1996; Paustenbach, 2002) where:

$$HI = \Sigma HQ_i, \qquad i = 1 \ldots n. \tag{3}$$

If the calculated HI is less than 1.0, the non-carcinogenic adverse effect due to this exposure pathway or chemical is assumed to be negligible.

Table 2
Reference dose and slope factor for As

Element	RfD (mg/kg-d)	SF $(mg/kg\text{-}d)^{-1}$
As[a]	3×10^{-4}	1.5

[a] US EPA IRIS database (http://www.epa.gov/iriswebp/iris/index.html).

The cancer risks are expressed in terms of the probability one may develop cancer at a given lifetime exposure level. The cancer risk probability is determined from the slope factor of the dose–response curve in the low-dose region where the relationship between the exposure dose (measured in mg/kg-day) and response (measured in terms of probability of developing cancer) is assumed to be linear. Mathematically, the SF denotes the probability of developing cancer per unit exposure level of mg/kg-day. The lifetime exposure level (ADD_{life}) is arrived by prorating the exposure incurred over the exposure duration over the expected life span. Cancer risk is then calculated as follows (Kolluru et al., 1996; Paustenbach, 2002):

$$\text{Cancer risk} = ADD_{life} \times SF \tag{4}$$

This equation applies to a linear low-dose cancer risk model that is valid for risks below 0.01.

3. Results and discussion

3.1. Contamination level of arsenic

The range and mean concentration of As in the tailings, soils, groundwaters and rice is shown in Table 3. Maximum concentration of As in the tailings from the Myungbong Au–Ag mine were 3090 mg/kg. This represents the contaminant level at the pollution source that could seriously impact the soils and waters

Table 3
Range and mean concentration of As in tailings, soil, groundwaters and rice grains from the Myungbond Au–Ag mine

Sample type		Mean	Range	S.D.**
Tailings (mg/kg) (N*=3)		2383	1946–3090	618
Soils (mg/kg)	Paddy soil (N=5)	71.8	25–131	45
	Natural value of Korea[a]	9.6	6.1–13.0	–
	World's natural soil[b]	6.0	–	–
Groundwaters (mg/L) (N=2)		0.008	0.007–0.009	0.001
Rice grains (mg/kg)	Rice grains (N=5)	0.41	0.24–0.72	0.184
	Natural value of Korea[c]	0.09	0.02–0.15	–
	World's natural soil[d]	–	0.11–0.20	–

*N = no of samples , **S.D. = standard deviation.
[a]Ahn (2000), [b]Bowen (1979), [c]Rhu et al. (1988), [d]Kabata-Pendias and Pendias (1984).

Table 4
The ADD value of As with exposure pathways (mg/kg-day)

Exposure pathway	ADD
Soil ingestion pathway	7.8E-05
Water ingestion pathway	2.56E-04
Rice grain ingestion pathway	2.3E-03

around the tailing piles. As the soils and vegetations in the entire region have been affected by the emissions and fallouts of the mining operation, it is not be possible to establish the background concentration levels for the elements. The mean concentration of As in agricultural soils (paddy fields) was considerably higher than the typical contents of comparable soils around the world (Bowen, 1979) and average concentrations of uncontaminated soils of Korea (Ahn, 2000). The soil As concentration is alarming as the element in significant quantities would be expected to transfer from soils to rice grains and other crops grown in these agricultural fields.

Arsenic concentration in groundwaters used for drinking water from the Myungbong mine area was 0.008 mg/L, below the permissible level (0.01 As mg/L) for drinking water of WHO and USA. In comparison with natural concentration in rice grains grown in a non-contaminated area in Korea (Rhu et al., 1988), an elevated concentration of 0.41 As mg/kg in rice grains from the Myungbong mine area was observed. Regular consumption of rice by the local population could pose a potential health problem from a long-term As exposure in the vicinity of this mine.

3.2. Evaluation of human exposure

In this study, the conceptual model was based on the exposures of a typical Korean farmer residing close to the mine. The exposure scenario in this situation was narrowed down to the intake of arsenic through drinking water, crop plant and direct ingestion of soil caused by improper personal hygiene that transferred soils attached on hand to mouth. A major component of the Korean diet is rice, and the food pathway through direct ingestion of rice grains was considered in our case. The dosage of the exposures may be estimated by the expected quantity of As in the ingested waters, rice grains and soils.

Table 4 summarizes the outcomes of the ADD estimations for As with the three exposure pathways. The average daily intakes of As via the rice grain pathway were 10^2 times higher than those of the soil ingestion pathway.

3.3. Evaluation of toxic risk

The HQ value for As via soil and water exposure pathways was all < 0.1 (Table 5). Therefore, there is no soil and water ingestion toxic risk in the Myungbong mine area. However, the HQ value for As by the rice grain exposure route was 7.8 (Table 5). The resulting HI value for As is much greater than 1.0 and this toxic risk due to rice ingestion in the Myungbong mine area is significant.

3.4. Evaluation of cancer risk

Arsenic is a human carcinogen by classification of chemicals according to the weight of evidence from animal experiments, clinical experience and epidemiologic studies by US EPA. The increased cancer risks of being exposed to As by the soil ingestion, rice consumption and drinking water routes, according to Eq. (4), are 5E-5 (4.6×10^{-5}), 1E-3 (1.4×10^{-3}) and 2E-4 (1.5×10^{-4}), respectively. The As cancer risk via the exposure pathway of rice consumption exceeds the acceptable risk of 1 in 10,000 set for regulatory purposes (Kolluru et al., 1996; Paustenbach, 2002). Thus, the daily intake of rice grain by the local residents can pose a potential health threat due to long-term As exposure.

4. Conclusions

Tailings of the Myungbong mine, closed since the 1980s, contained 2383 As mg/kg. This concentration is considerably elevated above the background levels of soils and can seriously impact the soils and waters of the surrounding areas through the surface erosions caused by weathering and transport. Mean concentration of As in paddy soils was 72 mg/kg.

The outcomes of the risk assessment show that the toxic risk HI (hazard index) for As for the exposed individuals (adult farmers) in the affected area is significantly greater than 1 due primarily to exposure from rice consumption that is grown on arsenic-contaminated soils. The cancer risk due to As for the exposed individuals via the rice consumption pathway

Table 5
Hazard index (HI)and hazard quotient (HQ) for As in the Myungbong mine site

Exposure route	HQ
Soil ingestion	0.13
Water ingestion	0.85
Rice grain ingestion	7.82
HI	8.80

exceeded the probability of one cancer case in ten thousand threshold set by regulation. Thus, rice ingestion is a significant pathway to As exposure in humans. Rice consumption poses a significant human health risk to the inhabitants of the abandoned Myungbong mine area.

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