Environmental Contamination of Arsenic and Heavy Metals around Cho Dien Lead and Zinc Mine, Vietnam

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ABSTRACT

An onsite survey of heavy metal pollution in the environment, including tailings, soil, and water around Cho Dien mine was conducted. There is considerable amount of heavy metal pollutants, such as As, Cu, Cd, Pb, Zn, present at various levels in soil, surface water, and drinking water. Note that, most of the soil samples are being severely polluted with Pb, Zn, Cd and As. The results showed that 100% of soil samples contained Pb and Zn exceeding the Vietnamese standards. Tailings sample was found to contain elevated levels of these pollutants. Overall, surface water and drinking water around the mining area were found to have low As, Cd, and Cu content. Meanwhile, there have been quality problems for drinking water and surface water containing relatively high concentrations of Pb, with 45.4% and 40% of total samples exceeding the allowable standard, respectively. High concentrations of Pb were found in some water samples collected from sites close to the mining area and tailings dam. Mining wastes and tailings are sources of heavy metal pollution and may result in ecotoxicological effects on terrestrial, groundwater and aquatic ecosystems. On the other hand, the source identification of contaminants was made partially by the examination of the distribution map and also based on what other researchers mentioned.

Keywords: arsenic, Cho Dien mine, heavy metals, mining, tailings

INTRODUCTION

Mining activities are known to be potential sources of contamination due to arsenic and heavy metals generated and released into the environment. Release of metals from mining sites occurs primarily through mining-milling, ore concentrating, refining processes and disposal of tailings and waste rock. The extent and degree of heavy metal contamination derived from the mining activities may vary depending upon the type of mineralization, composition of ore minerals, geology, topography, method of mining and smelting (Jung and Thornton, 1996). Toxic metals in tailings and waste rock can be released to streams, farming areas and can accumulate in crop plants of the mining area (Ju *et al.*, 2005; Hye *et al.*, 2008). In many previous studies, the elevated levels of heavy metals that were found in soil, sediment, stream and water surrounding mining areas were derived from weathering processes of minerals, tailings and waste rock (Lee *et al.*, 2001; Israel *et al.*, 2004; Aparajita *et al.*, 2006). The contamination by these metals in water and soils has posed a potential public health risk in many countries such as Bangladesh, Korea, and India (Chowdhury *et al.*, 2001; Alam *et al.*, 2003; Hye *et al.*, 2008).

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In Vietnam, lead and zinc are abundant and are mainly concentrated in the northernmost part of the country with a total ore reserve of up to 97 million tons (Tran *et al.*, 2009). Geological investigation and exploration have shown that lead-zinc ore found within the scope of Bac Kan Province has great potential. Currently, due to the demand for socio-economic development, the searching, exploring and mining work of lead and zinc are expanding constantly. Mining of lead and zinc in the area has become the leading activity of Bac Kan Province. However, due to the geological structure and characteristics as well as the mining conditions, lead and zinc mines are mainly concentrated in this region so the local environment is being polluted.

This study focused on Cho Dien mine in Bac Kan Province, which was estimated to be one of the largest lead and zinc mine in Vietnam. In this province, studies of heavy metal contamination derived from lead and zinc mining activities are limited. Therefore, the aims of this study were: (1) to conduct a preliminary investigation of the contamination levels of As and other heavy metals and (2) to identify the sources of contaminants in soil and water at the study area (i.e. Bac Kan Province, Vietnam).

MATERIALS AND METHODS

Study area

The study area is located in a monsoon tropical climate region with an annual average temperature of 22.3°C and rainfall of 1,822 mm with two distinct seasons: rainy and dry seasons. The rainy season is from April to September, with total rainfall of 100 - 600 mm/month. For the dry season from early October until March next year, rainfall is negligible. Most of the people in this area are involved in agricultural work (70%) and earn an average annual income of about US\$ 320/year. The main source of drinking water for villagers is supplied by Vietnam National Rural Clean Water program (taken from mountain creeks and pre-treated) and used by about 84% of households. A small number of villagers are using bore wells and dug wells (16%). The study area has sparse stream networks with surface flows significantly changing by season, flowing towards the valley. During dry season the branch streams are almost dry. Ban Thi stream is mainly used for irrigation and domestic activities in this region. Residential areas surrounding the lead-zinc mining areas are mostly located below the valley. The study area and sampling locations are shown in Fig. 1.

Sampling

Sampling of tailings, soil, and water was conducted on September 3 - 8, 2010 in and around the lead-zinc mining areas (22° 11' 50" N, 105° 32' 00" E). Since this time was the end of the rainy season, it was sunny during sampling.

Surface soils (0 - 15 cm in depth) were sampled by trowel, from the tailings, forest, and paddy fields. Eighteen locations of natural forest (F1 - F18) on the mountain near the mining sites and at different elevations were selected. Six paddy fields (P1 - P6; P1, P2 at higher positions while P3 - P6 at lower positions) located near the mining area and in the valley below the slope of the mountain were also investigated. All the soil samples were stored in polyethylene bags.

Surface water samples were taken from Ban This stream, small ponds, and several

Fig. 1 - Study site and sampling location map of Cho Dien mine.

stream branches (SW1 - SW20). This water is mainly used for irrigation and cattle watering, and in some places for domestic water supply.

Drinking water was taken from various sources including six bore well stations (BW1 - BW6), fourteen creeks (CW1 - CW14), and two dug well stations (DW1, DW2). Each water slot provides drinking water for one to several households.

All water samples were collected in 500 ml polyethylene containers previously washed with 10% HNO_3 solution and rinsed with deionized water. The values of pH were determined *in situ* by using a portable Twin pH meter.

Analysis

Tailings and soil samples were dried at room temperature (approximately 30°C) and ground to pass a 0.15-mm mesh. Then, 0.25 g of sample was digested using aqua regia ($\text{HCl}/\text{HNO}_3 = 3 : 1$) (USEPA, 2001) by the microwave digestion method. After this process, the digested samples were filtered through Whatman GF/C filter papers into 50 ml borosilicate volumetric flasks and made up to the mark with distilled water. These solutions were analyzed by atomic absorption spectrometry (AAS 800 Perkin Elmer,

USA) for Cd, Cu, Pb and Zn and by inductively coupled plasma mass spectrometry (ICP-MS ELAN 900-Perkin Elmer) for As.

Water samples were filtered through a 0.45 µm membrane filter on site into acid-washed polyethylene bottles and divided into two portions. One part was acidified with HNO₃ (0.2% v/v) for analyzing As and heavy metals, avoiding Fe hydroxide precipitation, while the other was used for anion determination. Water samples were stored in a refrigerator at 4°C before physical-chemical analysis. At least 10% of the samples were spiked and analyzed in duplicate. The relative standard deviation (RSD) was between 2% and 10%.

Ionic constituents (NH₄⁺, NO₃⁻, NO₂⁻, SO₄²⁻, Cl⁻) were analyzed by ion chromatography (IC, DIONEX600, DIONEX, USA) and the other elements (As, Ca, Cd, Cu, K, Mg, Na, Pb and Zn) in all water samples were determined by ICP-MS (Detection limit for Cd and Pb is 0.0001 mg/L while for As, Cu and Zn is 0.001 mg/L). For AAS, the detection limits for Cd, Cu, Pb, and Zn were 0.03, 0.08, 0.45, and 0.02 mg/L, respectively. Bicarbonate (HCO₃⁻) concentration was examined by the titration method.

RESULTS AND DISCUSSION

Heavy metal contamination of water

Drinking water

Drinking water samples were taken from 22 locations, including creeks, dug wells, and bore wells. The average concentrations of the major cations and anions in drinking water are presented in Tables 1 and 2. The values of pH were neutral and slightly basic (pH = 7 - 8.1). The most abundant cation and anion were Ca²⁺ and HCO₃⁻, respectively. Relative ionic abundance in all drinking water samples at the mining area showed the trend: HCO₃⁻ > SO₄²⁻ > Cl⁻ > NO₃⁻ > NO₂⁻ for anions and Ca²⁺ > Mg²⁺ > K⁺ > Na⁺ > NH₄⁺ for cations.

The Cd, Cu, and Zn contents of all drinking water samples were lower than their detection limit of 3 µg/L, 2,000 µg/L, and 3,000 µg/L, respectively (Table 3). In some locations, the concentrations observed were below the detection limits.

As can be seen from Fig. 2a, most of the collected samples has As concentration lower than the permissible standard set by the Vietnamese government as well as the USA quality standard for drinking water, except in one creek where the concentration is above 10 µg/L (CW1).

Table 1 - Average and range of concentrations of major cations in drinking water.

Type (Number)	Ca	K	Na	Mg	NH ₄
Bore wells (6)	65.3	1.12	0.28	1.34	0.01
	55.1-91.2	0.89-1.34	ND-0.45	1.07-2.12	ND-0.03
Creeks (14)	58.3	0.25	0.13	0.46	0.01
	42.7-72.1	0.07-0.76	ND-0.24	0.13-0.87	ND-0.03
Dug wells (2)	62.1	0.56	0.22	1.25	0.01
	61.1-63.2	0.35-0.77	0.12-0.32	1.18-1.32	ND-0.02

ND; Not detected

Table 2 - Average and range of concentrations of major anions in drinking water. (mg/L)

Type (Number)	Cl	HCO ₃	NO ₂	NO ₃	SO ₄
Bore wells (6)	8.3 5.4-11.6	236 224-255	0.02 0.01-0.03	0.87 0.45-2.63	27.1 8.5-107.1
Creeks (14)	0.75 0.54-2.12	256 247-263	0.01 ND-0.02	0.67 0.32-4.11	4.36 0.24-6.61
Dug wells (2)	4.28 3.45-5.10	238 230-245	0.015 ND-0.03	0.12 0.11-0.13	8.6 5.5-11.7

ND; Not detected

Table 3 - Average and range of concentrations of As and heavy metals in drinking water sampled around Cho Dien mine. (mg/L)

Type (number)	As	Cu	Cd	Pb	Zn
Bore wells (6)	5.1 (2-8)	29 (2.0-54)	0.5 (ND-2)	30 (ND-121)	71 (ND-155)
Creeks (14)	3.0 (ND-13)	46.5 (5-218)	0.61 (ND-2)	12.0 (ND-34)	58.4 (ND-170)
Dug wells (2)	4.5 (2-7.1)	35 (11-59)	1.5 (1-2)	13.5 (9-18)	472 (2-942)
1329/ 2002/BYT/QĐ	10	2000	3	10	3000
USA drinking water standard	10	1300	5	15	

1329/2002/BYT/QĐ: Hygiene Standard of Drinking Water-Ministry of Public Health, Vietnam

ND; Not detected

Fig. 2 - Distribution of a) As and b) Pb in drinking water of Cho Dien mine.

The concentration of Pb varied widely in the range of ND - 121 µg/L, ND - 34 µg/L and 9 - 18 µg/L for bore wells, creeks, and dug wells, respectively (Table 2). In creek samples, 6 of the total 14 samples had Pb content higher than the permissible standard. These were mainly at sites near the mining areas (CW2, CW3, CW4) and downslope areas (CW8, CW9, CW10).

For bore wells and dug wells, samples were taken in the residential area near the tailings dump and below the valley. The locations where Pb concentration was higher than the allowable standard were at two bore wells near mine tailings (BW5, BW6), and in another well within the valley (BW4). The maximum concentration of Pb was found at BW4 with the concentration of up to 121 µg/L. The dug wells were also found to have Pb contamination. Distribution of Pb in drinking water is illustrated in Fig. 2b.

Relative metal abundance in drinking water at the mining area showed the following trend: $Zn > Pb > Cu > As > Cd$ for bore wells, and $Zn > Cu > Pb > As > Cd$ for creeks and dug wells.

In comparison with the allowable values, the analyzed results indicated that Pb contamination in drinking water is a major concern and caused a significant health risk for residents living in and around Cho Dien mine.

Surface water

The measured concentrations of chemical species in surface water are shown in Tables 4 and 5. Similar to drinking water samples, the most abundant cation and anion were Ca^{2+} and HCO_3^- , respectively. For anions: most of the analyzed samples showed levels of Cl^- concentration that change very little from 0.38 - 3.17 mg/L. This was followed by SO_4^{2-} . For cations: Ca^{2+} was the most dominant in the range of 41.7 - 69.8 mg/L, while the amounts of Mg^{2+} ranked second (1.59 - 3.99 mg/L). The stream water was characterized by high concentration of Ca^{2+} which was derived from carbonate minerals. Sodium ion (Na^+) was almost undetectable in all samples.

Table 4 - Average and range of concentrations of major cations in surface water sampled around Cho Dien mine. (mg/L)

Elements	Range	Average (n=20)
pH	6.7-8.1	7.5
Ca	41.7-69.8	60.0
K	0.28-1.18	0.39
Na	ND-0.126	0.021
Mg	1.59-3.99	2.24
NH_4	ND-0.05	0.02

ND; Not detected

Table 5 - Average and range of concentrations of major anions in surface water sampled around Cho Dien mine. (mg/L)

Anions	Range	Average (n=20)
Cl	0.38-3.17	1.55
HCO_3	223-269	235
NO_2	ND-0.009	0.004
NO_3	0.17-1.93	1.00
SO_4	3.57-107	24.3

ND; Not detected

Compounds containing nitrogen were found in small amounts. The concentration of NO_2^- was below 0.01 mg/L in surface samples, while NO_3^- concentration ranged from 0.17 - 1.93 mg/L (Table 5). The same trends of relative ion abundance in surface water as drinking water were also observed at the mining area in the order: $\text{HCO}_3^- > \text{SO}_4^{2-} > \text{Cl}^- > \text{NO}_3^- > \text{NO}_2^-$ for anions and $\text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+ > \text{Na}^+ > \text{NH}_4^+$ for cations.

According to the results presented in Table 6, the surface water around the mining areas contain elements such as As, Cu, Cd, Pb and Zn with average concentrations of 18.2 $\mu\text{g/L}$, 146 $\mu\text{g/L}$, 3.3 $\mu\text{g/L}$, 107 $\mu\text{g/L}$ and 1,040 $\mu\text{g/L}$, respectively. The levels of As, Cu and Cd in surface water were generally less than 50 $\mu\text{g/L}$, 500 $\mu\text{g/L}$ and 10 $\mu\text{g/L}$, respectively. However, SW3 (95 $\mu\text{g/L}$), SW1 (850 $\mu\text{g/L}$) and SW20 (12 $\mu\text{g/L}$) exceeded the standard for As, Cu, and Cd, respectively.

The distribution of Pb in surface water is shown in Fig. 3b. The highest level of Pb was up to 564 $\mu\text{g/L}$ and it was measured at SW4, which received the effluent discharging from the mining excavation at upper stream. The concentrations of Ca^{2+} and SO_4^{2-} were also found the highest at this point, suggesting that there was an input of acid mine drainage. The concentration of Pb was also above the allowable level at the downstream SW8 (179 $\mu\text{g/L}$), at some locations near mining area SW2 (423 $\mu\text{g/L}$), close to tailings dam SW16 (52 $\mu\text{g/L}$) and SW17 (83 $\mu\text{g/L}$), and the confluence SW20 (108 $\mu\text{g/L}$). In comparison, Pb concentration in surface water was higher than its measured

Table 6 - Average and range of concentrations of As and heavy metals in surface water sampled around Cho Dien mine. (mg/L)

Elements	Range	Average (n=20)	QCVN-B1 08:2008/BTNMT
As	ND-95	18.2	50
Cd	ND-18	3.3	10
Cu	3-865	146	500
Pb	2-564	107	50
Zn	10-4,170	1,040	1,500

ND; Not detected

Fig. 3 - Distribution of a) As; b) Pb and c) Zn in surface water of Cho Dien mine.

concentration in drinking water. Quang *et al.* (2006) reported that the percentage of samples with Pb concentration higher than the allowable standard was 45.2% (n = 31) in surface water of the study area.

Contamination level of Zn was much higher, varying from 10 - 4,170 µg/L. The Zn content was observed to decrease from upstream to downstream. At the points SW1, SW4 and SW5, Zn concentrations exceeded the allowable standard (as shown in Fig. 3c). The highest concentration of Zn reached 4,170 µg/L at SW1. However, for the surface water samples adjacent to the tailings dam, the concentration of Zn still remained within the permissible limit.

Overall, metal content was relatively low in surface water at the studied area even for the samples at the tailings dam and at the downstream station. The pH value of water samples, which was neutral to slightly basic (pH = 6.8 - 8.1), could be the main factor causing this. According to Travainen *et al.* (1997), the high pH in water (> 7) may enhance toxic metal precipitation.

Heavy metal contamination of tailings and soil

The range and average concentrations of As and heavy metals (Pb, Zn, Cu, Cd) in tailings, paddy soil and forest soil are given in Table 7. As can be seen from the table, elevated levels of As (10,200 mg/kg) and heavy metals such as Pb (10,000 mg/kg), and Zn (2,800 mg/kg) are measured in tailings sample. These values significantly exceeded the Vietnamese standards and are also higher than the world's normal soil composition as reported by Bowen (1979).

According to the land use, contents of As and heavy metals in soils were quite different. The average concentration of these metals was highest in the forest soil and lowest in paddy soil. This is also similar with the study of Ju *et al.* (2005) during the assessment of As and heavy metal contamination in the vicinity of the abandoned mine in Korea.

Arsenic, cadmium, copper and lead were recorded to have heavily polluted the surface layer of the natural forest soil with 100% of soil samples containing Pb and Zn exceeding the Vietnamese standards. High levels of metals were found near the mining areas and tailings dam with the highest concentrations of As, Cd, Pb, Zn (F2). The concentrations of these metals reached 489 mg/kg, 23.6 mg/kg, 22,400 mg/kg, and 6,820 mg/kg, respectively. Pho *et al.* (2006) also identified that the soil in the study area was seriously polluted by heavy metals with 100% Pb and Zn and 43% of Cd content higher than the allowable standards. In comparison with various studies, Li and Thornton (1993) found elevated levels of As, Cd, Pb and Zn to be 42.3 mg/kg; 38.3 mg/kg; 28,900 mg/kg; and 3,180 mg/kg, respectively. The surface soils (0 - 5 cm) of a mining area in Mexico containing 19 - 17,400 mg/kg of As was reported by Israel *et al.* (2004). However, the content of As, Cu, Cd, Pb, and Zn in forest soil decreased with distance from the mining area (Fig. 4). In some cases, increased concentrations were observed, especially As concentration was found to be relatively high (62.2 mg/kg) at a location quite far from the mining area (F18).

Average concentrations of As, Cu, Cd, Pb, and Zn in paddy soil were higher than the permissible level as well as the world's normal values, especially, the maximum levels

of As and Cd in paddy soil were up to 71.9 mg/kg and 3.44 mg/kg, respectively. The ratios of As and heavy metals (> 1) shown in Fig. 5 indicate a certain elevation of contaminants. Various studies reported As and heavy metal contamination of paddy soil associated with mining and smelting activities. Lee (2006) reported the high As contents (84.7 - 130 mg/kg) in the surface layer of paddy soil in the vicinity of an abandoned metal mine in Korea. Elevated levels of Cd, Cu, Pb and Zn were also found in paddy soil taken from Daduk Au-Ag-Pb-Zn mine in Korea (Lee *et al.*, 2001).

Table 7 - Average and range of concentrations of As and heavy metals in tailings and soils of the mining areas. (mg/kg)

Type (Number)	As	Cu	Cd	Pb	Zn
Tailings (1)	10,200	900	11.4	10,000	2,800
Forest soil (18)	228 (3.7-489)	147 (11.9-697)	12.6 (0.66-56.2)	4,010 (273-23,300)	3,540 (231-8,430)
Paddy soil (6)	20.8 (1.89-71.9)	128 (32.7-467)	1.36 (0.13-3.44)	370 (254-592)	557 (215-872)
QCVN 03:08/BTNMT	12	70	2	100	200
World's normal value	6.0	30	0.35	35	90

QCVN 03:08/BTNMT (Vietnam): National technical regulation on the allowable limits of heavy metals in soils

Fig. 4 - Distribution of a) As and Cd; and b) Pb and Zn in soil of Cho Dien mine.

Fig. 5 - Ratios of metal concentrations in paddy soil to the corresponding Vietnamese standard (VS).

Causes of As and heavy metal contamination in soil and water

The study area has been contaminated by toxic elements. The concentrations found in high levels were near the lead-zinc mining area and tailings dam. However, high content of As and heavy metals were also found in some locations far from the mine. Several reasons can be identified as follows:

- Due to exploitation activities: The exploitation processes of lead-zinc ore at the study site are both open pit and underground. Firstly, open pit process creates an appropriate condition for distributing and associating ores with elements in soil of nearby areas. In addition, the exploitation of sulfide ores has left a great volume of rock containing arsenopyrite (FeAsS). Secondly, underground process also dumps waste rock and discharge mine water into the surrounding environment, leading to surface water pollution.
- Due to the nature of ore deposit: Quang *et al.* (2006) found that the mineral composition of the Cho Dien mine is complex. The principal minerals of this region are sulfides, including sphalerite (ZnS)-Galena (PbS) (Tran *et al.*, 2009). Besides, this deposit also contains a minor but complex suite of trace minerals and elements such as Ag, Cu, As, Cd, Ge, and Ni. Minor and trace minerals may include arsenate minerals, arsenopyrite, barite, bravoite, bornite, and chalcopyrite among others. These minerals are exposed to the surface and they can be oxidised. As a result, the processes will release the toxic elements into soil and water.
- Composition of ore surrounding rocks: This factor plays a major role in the weathering process of sulfide ore. At Cho Dien mine, limestone and other carbonate rocks are common. When acid solution runs into limestone or carbonate rocks, the solution is gradually neutralized. After that, pH increases slightly from 6 to 8. These will create appropriate conditions for the deposition of mineral carbonate material such as cerussite (PbCO_3) and smithsonite (ZnCO_3). At the study site, the wastes contain elevated levels of Ca, ranging from 5.2 - 13.7%, which are derived from the original mineralized limestone and from fluxes such as calcite CaCO_3 and lime CaO ,

and Ca(OH)_2 added during the smelting process. The relatively neutral soil pH in the study area shows that As and heavy metals will be immobile in the local soil profile. Navarro *et al.* (2008) reported that the presence of high amounts of carbonate in the soil around the mining area revealed the stabilization of all metals studied.

- Due to topography: Cho Dien mine is located on high terrain in which surface water and groundwater flow towards low zones. The water will carry minerals and toxic elements contained in the ore.

In addition, the sources that caused soil contamination could be identified in this area by examining the distribution maps (as shown in Figs. 2 to 4). Elevated levels of As and heavy metals were measured in tailing and soil samples near the tailings dam. According to these maps, the highest contamination was found closest to the tailings dam and mining areas. The results indicated that tailings might be important contamination source of As and heavy metals in the environment and can pose a potential risk in the study area.

CONCLUSIONS

It was found that there were no remarkable differences in As and heavy metal contents in surface water as compared with drinking water. The contents of As, Cu, and Cd were relatively low in all samples while Pb was the most existent. The concentration of Pb in drinking water and surface water exceeded the Vietnamese standards of 10 $\mu\text{g/L}$ and 50 $\mu\text{g/L}$ at 45% and 40% of sampling locations, respectively. The results suggested that Pb contamination in drinking water and surface water is a major concern.

There was a serious soil pollution in Cho Dien mine. Soil located near mine spoils had extremely high concentration of As and heavy metals such as Cu, Cd, Pb, and Zn. The highest average concentration of these metals was found in tailings, then in forest soil and the lowest concentration was found in paddy soil.

Tailings dam and mining activities were the main sources of As and heavy metal pollution in the studied area. On the other hand, the important factors such as the type of mining, composition of materials and rocks, geological characteristics, forms of exploitation, and creation of waste dump also release these substances into this area. The relatively neutral pH of soil surrounding Cho Dien mine plays an important role in metal mobility.

There should be further studies about the impacts of As and heavy metals on human health in the study area. Moreover, researches about the mobility characteristics and distribution mechanism of these metals in different types of environment should be carried out. The local government should have appropriate solutions to manage the mining activities and waste dumps, especially the remediation of soil in this area.

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