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Arsenic environmental contamination, chemical speciation and its behaviour in the water system from some abandoned Au-Ag mines, Korea

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Abstract: Mine waters, surface waters and groundwaters were sampled around seven Au-Ag mine areas (Dongil, Okdong, Dongjung, Songcheon, Ssangjeon, Dogok and Gubong Au-Ag mines). The main contamination sources of As in these abandoned Au-Ag mines can be suggested as mine tailings and waste rocks including the sulfide gangue minerals (arsenopyrite). The relatively high concentration of As in mine waters was shown in the Dongil (524 μg/L) and the Dogok (56 μg/L) mine areas. Arsenic concentrations in stream waters from the Dongil (0.9~118 μg/L), the Songchon (0.8~63 μg/L), the Ssangjeon (1.6~109 μg/L) and the Gubong (3.6~63 μg/L) mine areas exceeded the permissible level for stream water in Korea. Groundwaters collected from the Dongil (0.9~64 μg/L), the Okdong (0.2~69 μg/L) and the Gubong (0.5~101 μg/L) mine areas contained high As concentration to cause the arsenicosis in these areas. In As speciation, the concentration ratios of As(III) to As(total) present up to 75 % and 100% in stream waters from the Okdong and the Songcheon mines, and 70% in groundwaters from the Okdong and the Dongjung mines. Arsenic concentration decreases downstream from the tailing dump correlatively with pH and Fe concentration. Highly elevated As concentrations are found in the dry season (such as April and March) than in the wet season (September) due to the dilution effect by heavy rain during summer in stream waters from the Dongil and the Songcheon mine areas.

1. Introduction

The increase in the number of heavy metal emitting sources has aroused and the degree of these metal pollutions has been one of the main topics studied in environmental science (Plant and Raiswell, 1983). Mining is one of the most important anthropogenically-induced sources of heavy metals in the environment owing to various mining activities including processing, transportation of ores, disposal of tailings and waste waters around mines (Adriano, 1986). Especially, inorganic arsenic is classified as human carcinogen and causes serious environmental problems in abandoned gold mine areas. Arsenic-containing minerals are common in gold-bearing rocks and gold mining has been associated with release of high levels of arsenic into the environment. There are over 900 metalliferous mines in South Korea and about 98% of them were abandoned due to a lack of ore minerals. And most of these abandoned mines have been left without any management. Therefore it may pose a potential health risk to residents in the vicinity of mines (Jung and Thornton, 1996; Chon et al., 1998; Lee et al., 2000, 2001).

The objectives of this study are (1) to investigate the extent and degree of As contamination in water system; (2) to identify arsenic species in surface and groundwater to assess environmental toxicity; (3) to examine the variation of As according to pH and Fe concentration; (4) to monitor the seasonal variation of As in water system in some abandoned Au-Ag mine areas.

2. Materials and Methods

Mine, stream and groundwater samples were collected from the Dongil, the Okdong, the Dongjung, the Songcheon, the Ssangjeon, the Dogok and the Gubong Au-Ag mines which are all fissure filling deposit and major metallic minerals of gold-silver. Although the Okdong mine is typical base metal mine, the exploitation of gold-silver was reported. For monitoring seasonal variation of As in water system, water sampling was carried out on April and September, 2002 in the Dongil and the Okdong mine areas and September, 2002 and March, 2003 in the Songcheon mine area. Physical properties such as pH, Eh and Conductivity were measured in the field. The chemical analyses of water samples were carried out using ICP-AES and ICP-MS for cations, and IC for anions. Dissolved total As in some water samples was separated As(III) from As(V) using silica-based anion exchange cartridges and their concentrations were measured by hydride generation-AAS and graphite furnace-AAS with matrix modifier of Pd.

3. Results and Discussion

Hydrogeochemical characteristics of waters

Range and mean concentration of As in mine water, stream water and groundwater in the vicinity of abandoned Au-Ag mine areas are shown in Table 1. Arsenic concentration in mine waters from the Dongil (524 μg/L) and the Dogok mine (56 μg/L) was much higher than that from the Okdong mine (0.4~33 μg/L). Elevated levels of As in stream waters were also found in the Dongil (0.9~118 μg/L), the Songcheon (0.8~63 μg/L), the Ssangjeon (1.6~109 μg/L) and the Gubong (3.6~63 μg/L) mines. Arsenic concentration in groundwaters was highly elevated in the Dongil (0.9~64 μg/L), the Okdong (0.2~69 μg/L) and the Gubong (0.5~101 μg/L) mine areas. Maximum level of As in these areas are higher than the permissible level of As for drinking water suggested by WHO (0.01 mg/L) and Korea (0.05 mg/L). Therefore, it can be possible to the human health risk of arsenicosis because groundwater is used by drinking water in these gold mine areas.

Table 1. Range and mean concentration of As in mine, stream and ground waters from abandoned gold mines (μg/L).

Sample type	Dongil mine	Okdong mine	Dongjung mine	Dogok mine	Songcheon mine	Ssangjeon mine	Gubong mine
Mine water	524	0.4 – 33	-	56	-	-	-
Stream water	0.9 - 118	< 0.1- 50	0.3 - 11	0.1 - 6.0	0.8 - 63	1.6 – 109	3.6-63
Ground-water	0.9 - 64	0.2 – 69	3 - 15	0.7 - 0.8	28	0.5 – 4.8	0.5-101

Chemical speciation of arsenic

In order to assess actual environmental toxicity of As, dissolved total As in some water samples was separated As(III) from As(V) using silica-based anion exchange cartridges (500 mg sorbent of 40 μm particle size and 60-A pore size). Arsenate(V) and arsenite(III) are the primary species of As in soils and natural waters. Of these forms, As(III) is the most soluble and mobile as well as the most toxic species. But, in aquatic system in general, As(V) species is the most common species in waters. Arsenate ($H_2AsO_4^-$) was more dominant than arsenite (H_3AsO_3) in waters collected from all mine areas. However, the concentration ratios of As(III) to As(total) present up to 75 % and 100% in stream waters from the Okdong and the Songcheon mines as compared with the other mine areas. Also, the highest concentration ratios of As(III) to As(total) are found in groundwaters used by drinking water from the Okdong (70%) and the Dongjung (82%) mine areas.

Variation of As according to pH and Fe concentration

To identify the variation As concentration according to pH and Fe concentration, these data were plotted in Fig. 1. In the Dongil mine area, the stream waters collected from the mine site to the downstream. Although As concentration is abruptly decrease downstream from the tailing dump due to the dilution effect, this concentration decreases correlatively with pH and Fe concentration in each sampling period of September in 2001, April and September in 2002 (presented in the separate columns in the Fig. 1). These variations of As concentration were also matched correlatively with pH and Fe concentration in the Dongjung and the Songcheon mine areas.

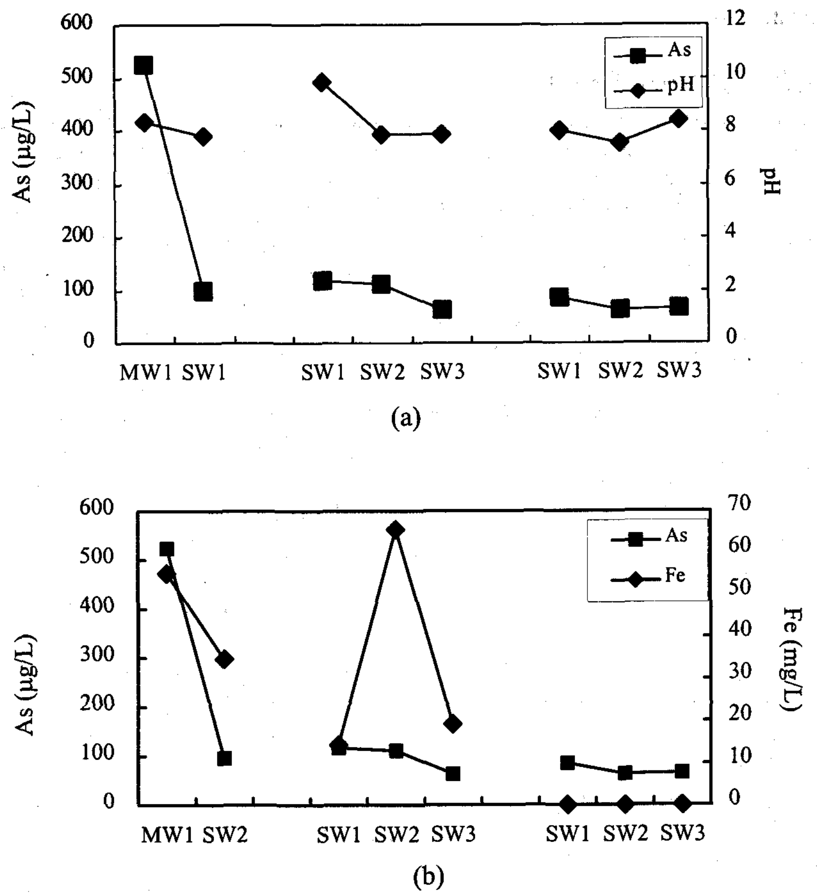

Fig. 1. The variation of As concentration according to pH (a) and Fe concentration (b) in the Dongil mine area.

Seasonal variation of As concentration

In monitoring of As contamination level in the dry and the wet seasons, highly elevated As concentrations were found in the dry season(on April, 2001) than in the wet season which is the period after heavy rain in summer(on September and October, 2001 and 2002). Arsenic concentration decreases in the order of Apr., 2002(117.6 μg/L) > Sep., 2001(97.0 μg/L) > Sep., 2002(85.3 μg/L) in stream water collected from the same sampling point in the Dongil mine area(Fig. 2). Also, in the Songcheon mine area, the stream waters collected near the mine site showed the decrease of As concentration in the order of May, 2003(764 μg/L) > Sep., 2002(165 μg/L). This indicates that the As contamination of water system on September was influenced dilution effect by heavy rain in summer which is characteristic climate in Korea.

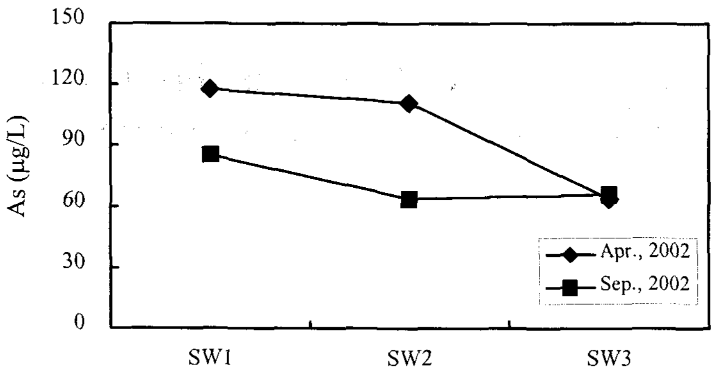

Fig. 2. Seasonal variation of As concentration in the stream water samples from the Dongil mine area.

4. Conclusions

Elevated levels of As were found in mine waters from the Dongil (524 μg/L) and the Dogok (56 μg/L) mines, and in stream waters from the Dongil (0.9~118 μg/L), the Songcheon (0.8~63 μg/L), the Ssangjeon (1.6~109 μg/L) and the Gubong (3.6~63 μg/L) mine areas. Arsenic concentration in groundwaters was highly elevated in the Dongil (0.9~64 μg/L), the Okdong (0.2~69 μg/L) and the Gubong (0.5~101 μg/L) mine areas. In As speciation, arsenate ($H_2AsO_4^-$) was more dominant than arsenite (H_3AsO_3) in waters from all mine areas. However, the concentration ratio of As(III) to As(total) present up to 75 % and 100% in stream waters from the Okdong and the Songcheon mines. Also, the concentration ratio of As(III) to As(total) is the highest in groundwaters from the Okdong (70%) and the Dongjung (82%) mines. Therefore it can be possible to the human health risk due to the more toxic As(III) species in the groundwater used by drinking water in the Okdong and the Dongjung mine areas. In the Dongil, the Dongjung and the Songcheon mine areas, variations of As concentration decrease downstream from the tailing dump correlatively with pH and Fe concentration. In monitoring of As contamination level in the dry and the wet seasons, highly elevated As concentrations are found in the dry season (April and March) than in the wet season (September) due to the dilution effect by heavy rain in summer in stream waters from the Dongil and the Songcheon mine areas.

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