



Comparison of arsenic co-precipitation and adsorption by iron minerals and the mechanism of arsenic natural attenuation in a mine stream



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ABSTRACT

Mine stream precipitate collected from Ilkwang mine, Korea, contained high concentrations of arsenic (As), while water collected from the same site had negligible As concentrations, indicating natural attenuation of As occurred in the mine stream. The mechanism of attenuation was explained by comparison of X-ray absorption near edge structure (XANES) of As(V) co-precipitated with or adsorbed to iron (Fe) minerals in mine precipitates. Arsenic in the mine precipitate was present as As(V) and schwertmannite was the main Fe mineral. Arsenic co-precipitation with schwertmannite was the major mechanism of As removal in the mine stream, followed by As adsorption by goethite and As co-precipitation with ferrihydrite. Schwertmannite and ferrihydrite were formed in acid mine drainage and As was incorporated in their structure during formation. Additionally, schwertmannite and ferrihydrite may transform to goethite with As adsorbed onto the goethite surface. Based on the results of batch experiments of As co-precipitation and adsorption, co-precipitation of As with ferrihydrite and schwertmannite was the most effective As sequestration mechanism in the removal of As(V) from acid mine drainage.

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1. Introduction

Acid mine drainage causes severe environmental problems, however, iron (Fe) and manganese (Mn) oxide precipitation from acid mine drainage may contribute to the attenuation of metal transport. Acid mine drainage contains inorganic acids, Fe^{2+} and sulfates (Flores et al., 2012). These precipitate as hydrous Fe oxides by chemical neutralization and act as metal adsorbents (Smith et al., 1998). Iron minerals such as ferrihydrite, goethite and schwertmannite are found in mine environments producing acid mine drainage. The Fe minerals precipitate as byproducts of pyrite oxidation (Mazzetti and Thistlethwaite, 2002).

The geochemistry of metals and metalloids in acid mine drainage can be controlled by several parameters, including mineralogy of the primary rocks, weathering conditions, hydrogeological factors and newly formed minerals (Acero et al., 2006). Formation of Fe minerals contributes to the removal of hazardous elements. Iron minerals such as schwertmannite and goethite

retain As, leading to natural attenuation of As in streams (Asta et al., 2010). Arsenic retention can be achieved by either co-precipitation or adsorption to the surface of Fe minerals (Courtin-Nomade et al., 2005). Arsenic co-precipitation occurs when the Fe minerals are formed in the presence of As while As adsorption does when As is added after the formation of Fe minerals (Crawford et al., 1993). Since the environmental conditions are not known when Fe minerals are formed, it is not clear whether As is co-precipitated in or adsorbed to Fe minerals.

Ferrihydrite is ubiquitous in precipitates and acts as a scavenger of contaminants because of its high surface area and reactivity. Individual ferrihydrite crystallites are generally less than 10 nm in size. Their structure contains 20% tetrahedrally and 80% octahedrally coordinated Fe (Michel et al., 2007). Schwertmannite is an amorphous Fe-oxyhydroxy sulfate mineral ($\text{Fe}_8\text{O}_8(\text{OH})_x(\text{SO}_4)_y$; $x = 8 - 2y$, $1 < y < 1.75$) commonly found in acid mine drainage as a brownish yellow precipitate (Bigham et al., 1994; Paikaray et al., 2011). Schwertmannite collected from acid mine drainage of the Kristineberg Zn–Cu mine, northern Sweden, was in the form of $\text{Fe}_8\text{O}_8(\text{OH})_{5.02}(\text{SO}_4)_{1.49} \cdot 0.5\text{H}_2\text{O}$ (Jönsson et al., 2005). When dissolved sulfate and Fe^{2+} in acid mine drainage are transported away

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and exposed to air in the presence of Fe oxidizing bacteria, Fe^{2+} is oxidized to Fe^{3+} and precipitates as secondary Fe minerals. Jarosite is found in solutions rich in sulfate at low pH ($< \text{pH } 3$). Schwertmannite is more common than jarosite in ochre deposits between pH 3 and 4 (Bigham and Nordstrom, 2000; Jönsson et al., 2005). Schwertmannite has also been reported to scavenge As from acid mine drainage (Acero et al., 2006; Carlson et al., 2002). Goethite ($\alpha\text{-FeOOH}$) is also a common component of ochre precipitates of acid mine drainage; it is poorly to moderately crystalline. It occurs along mine stream and forms light orange mixture with schwertmannite (Valente and Gomes, 2009). Goethite can be subsequently transformed from schwertmannite after the formation of schwertmannite at low pH and high SO_4 conditions (Fukushi et al., 2003). Goethite is known to have As(V) adsorption capacity through surface complex formation (Lakshmipathiraj et al., 2006).

Although it is known that Fe minerals, such as ferrihydrite and schwertmannite, play an important role in the removal of As, few studies have compared As co-precipitation with adsorption by various Fe minerals and applied the mechanism design for As natural attenuation. Therefore, the objective of this study is to elucidate the mechanism of As natural attenuation by comparing the co-precipitation of As in Fe minerals and the adsorption of As on the surface of Fe minerals. Naturally occurring Fe minerals in the Ilkwang mine stream were collected and the mechanism of As removal in the acid mine stream was examined. Ferrihydrite, schwertmannite and goethite were tested for As co-precipitation and adsorption as reference materials. Arsenic in mine precipitates was compared with co-precipitated and adsorbed As on ferrihydrite, schwertmannite and goethite, and their contributions to As natural attenuation were quantitatively evaluated.

2. Methods

2.1. Sampling of Ilkwang mine precipitates

Mine water and precipitate samples were collected from a mine adit and two different streams near the abandoned Ilkwang mine,

Pusan, Republic of Korea (Fig. 1). Mine water upstream was directly affected by the mine. Along the mine water course two samples (mine water upstream and downstream) were collected. The Ilkwang mine stream flows into the Jawgwangcheon stream, which was also sampled upstream and downstream of the confluence. Acid mine drainage only affected the Jawgwangcheon downstream of the junction. Sampling locations are marked in Fig. 1. At each site, stream water and precipitates were sampled separately. Yellowish red colored precipitates were collected in streams near the Ilkwang mine. The precipitates were collected using a syringe and then wet sieved through $150 \mu\text{m}$ sieves and freeze-dried. Samples were digested with HCl, HNO_3 , HClO_4 and HF. Total elemental concentrations were analyzed using inductively coupled plasma atomic emission spectroscopy (ICP-AES, Perkin Elmer). Wet precipitate samples were reacted at $300 \text{ }^\circ\text{C}$ and under 88 kg/cm^2 pressure for 2 h in a high pressure reactor system (HR-8200, Hanwoul); changes in crystallinity and mineralogy were evaluated for transformation of schwertmannite and ferrihydrite to goethite. The reacted precipitate was analyzed using a transmission electron microscope (TEM, Tecnai G2 F30) and X-ray diffractometry (XRD, PHILIPS X'pert MPD) equipped with a Cu-K α source at a working voltage 40 kV and current 30 mA. Water samples were analyzed for pH and EC using a calibrated pH and EC meter (Orion Star A211, Thermo Scientific); anion concentrations were measured by ion chromatography (IC, Metrohm) and cations by inductively coupled plasma optical emission spectrometry (ICP-OES, Ultima 2, Horiba), after passing through a $0.45 \mu\text{m}$ syringe filter.

2.2. Synthesis of Fe minerals

Ferrihydrite, schwertmannite and goethite were synthesized as reference materials (model minerals), as mine precipitates are generally comprised of these minerals and a similar mineral composition was expected at the Ilkwang mine study site. Two-line ferrihydrite, which shows two broad X-ray diffraction peaks because of poor crystallinity, was synthesized by dissolving 20 g of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ in 250 mL deionized water, adding 165 mL of 1 M

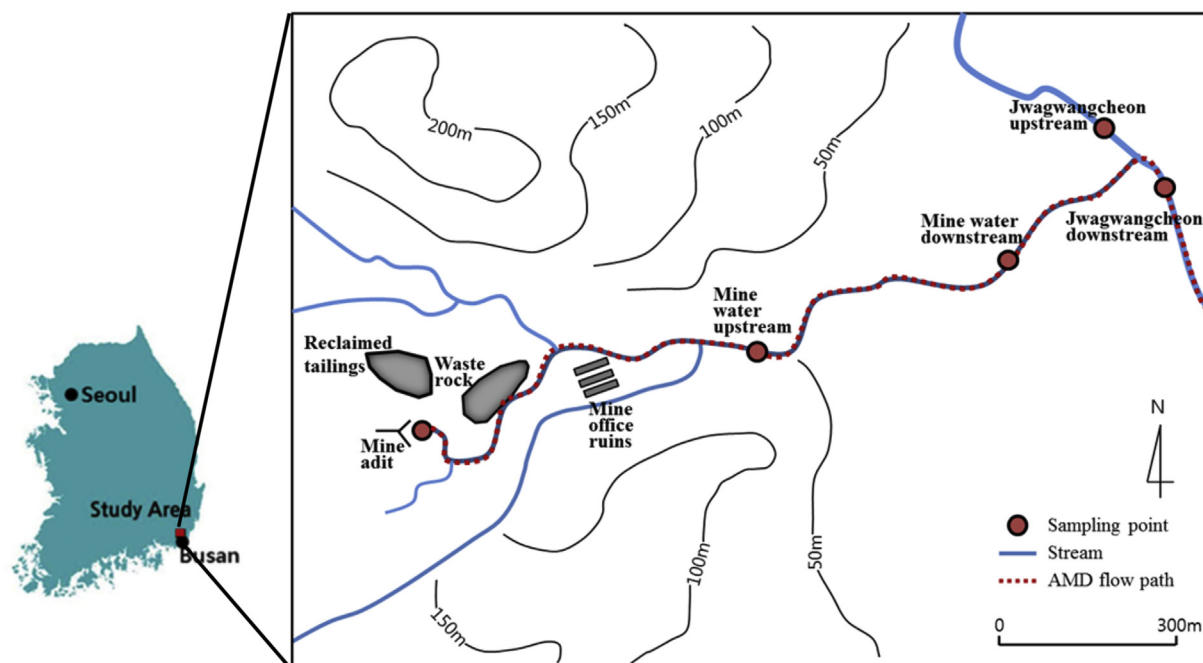


Fig. 1. Sampling locations at Ilkwang mine site.

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