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Hydrochemical assessment of environmental status of surface and ground water in mine areas in South Korea: Emphasis on geochemical behaviors of metals and sulfate in ground water

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ABSTRACT

In this study, hydrochemistry of ground and surface water collected around six metalliferous mines and one coal mine in South Korea was investigated to evaluate the status of mining-related impact and pollution. Groundwater system under the mine impact shows varying degrees of immobilization and retardation of heavy metals during the flow. SO_4^{2-} is shown to be the most reliable indicator of the mining impact on groundwater, as it reflects the degree of initial sulfide oxidation even after subsequent removal of metals; SO_4^{2-} is also less prone to sorption, precipitation, and geochemical reduction. A good correlation between the concentrations of SO_4^{2-} and the sum of Ca and Mg is observed, indicating that SO_4^{2-} represents the degree of dissolution of Ca- and Mg-bearing carbonates and silicates (i.e., neutralization of acidic water to circumneutral pH) by generated H^+ that is proportional to SO_4^{2-}. The higher Zn/Cd ratios of ores and water than those of tailings and precipitates indicate a more preferential immobilization of Cd, and this ratio also reflects the inherent composition of each ore. The modified HPI (Heavy Metal Pollution Index) consisting of the measured concentrations and respective environmental standards of Fe, Mn, Al, Zn, Pb, Cd, Cu, As, and SO_4^{2-} is suggested as MPI (Mine Pollution Index) to overall reliably evaluate the status of mining-related water pollution. The plot of SO_4^{2-} versus MPI is very effective to identify the contaminated mine water and its evolution, including the source and the pathways consisting of immobilization (precipitation, sorption) and dilution of contaminants in groundwater system. This study shows that a careful examination of the relationship between MPI and SO_4^{2-} can be very useful to identify diverse geochemical processes occurring in groundwater affected by mine drainage.

1. Introduction

Mining activity causes the exposure of sulfide minerals to oxidative environment; therefore, the mine wastes from metalliferous and coal mines act as the source of metalliferous leachate and subsequent contamination of surface and ground water systems (Yun et al. 2001; Herbert 2006; Cidu et al. 2009; García-Lorenzo et al. 2012; Lghoul et al. 2014; Resongles et al. 2015; Shim et al. 2015). The oxidation of sulfides in mine areas is also one of major anthropogenic contributions to biogeochemical sulfur cycle (Berner and Berner 1987; Stüeken et al. 2012). There have been numerous studies on water geochemistry in mine areas over the world. For instance, Toran (1987) investigated the oxidation of sulfide minerals and subsequent neutralization of acidic water by carbonates at the Pb-Zn mine area in carbonate aquifer near Shullsburg, Wisconsin, USA. Fuge et al. (1993) presented the geochemical behavior of Zn and Cd in water and solid samples in abandoned metalliferous mines in Wales, UK. Plumlee and Nash (1995) investigated the geoenvironmental model according to the types of mineral deposits. In addition, there have been investigations about the behaviors of metal species and arsenic in mine drainage (e.g., Smith et al. 1994). There have also been several comprehensive studies of mine tailings as a major contamination source (Nordstrom and Southam 1997; Holmström et al. 2001; Blowes et al. 2003, 2014; Jambor 2003; Johnson and Hallberg 2003; Huang et al. 2010; Lindsay et al. 2015). However, most studies on the geochemistry of mine water have been restricted to surface water in a specific area or ore deposit; therefore, understanding the immobilization of pollutants in groundwater is still poor.

Several indices have been suggested for the overall evaluation of water quality influenced by municipal or agricultural pollutants (SDD

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Fig. 1. Locations of the studied seven mines in Korean peninsula.

1976; Mano 1989; Cude 2001; Bordalo et al. 2006; Nikolaidis et al. 2007) or metal species (Mohan et al. 1996; Lee et al. 2009; Milijašević et al. 2011; Amadi et al. 2012; Brankov et al. 2012; Prasad et al. 2014). Some of those indices have used the combination of the weighting of individual parameter (Mohan et al. 1996; Prasad et al. 2014). Most of such studies using pollution indices have also focused on a specific watershed, river, or mine area. Therefore, for a better evaluation of the overall water quality in mine area, the suggestion and use of a more versatile index are still needed.

The objectives of this study are as follows: 1) to understand the behavior of heavy metals and major ions (especially, Ca, Mg, and SO_4^{2-}) in ground and surface water around several mines, and 2) to verify and use geochemical indicators to better assess the overall impact and environmental contamination in mine areas. Especially, the relationships among several hydrochemical parameters and suggested indicators are investigated to evaluate diverse processes such as precipitation, sorption, and dilution occurring in groundwater affected by mine drainage. In addition, removal of heavy metals and arsenic in groundwater system is also evaluated.

2. Studied mines

Five abandoned metallic ore mines and one coal mine under operation in South Korea (Fig. 1) were selected for the overall evaluation of the status of contamination of ground and surface water by mining activities. The climate of South Korea is monsoonal temperate with daily average temperatures ranging between −6 and 27 °C (annual average: 10–16 °C; KMA, 2015). Annual average amount of precipitation was about 1000–1900 mm during 1981–2010, of which ~70% occurred between June and September (KMA 2015). Ore types, contamination sources, and major species of contamination in the studied mines are summarized in Table 1.

In the abandoned Sambo Pb-Zn-Ba mine, Pb and Zn were major products, with minor amounts of Ba and Ag. Ore minerals occur in fissure-filling hydrothermal quartz veins in metamorphic rocks and consist mainly of sphalerite, galena, and barite with minor amounts of chalcopyrite and pyrite; gangue minerals include quartz, rhodochrosite, calcite, fluorite, dolomite, and sericite (So et al. 1984; Kim and Chon 1993). In the Sambo mine area, there are two huge dumps of tailings in different catchment areas (Fig. 2a): tailing dumps #1 and #2 have the horizontal extent of 51,345 m^2 and 18,249 m^2, and the depth of 6.9–17.5 m and 5.5–9.9 m, respectively, with the total volume of 722,000 m^3. Leachates are generated from each dump. In particular, the leachate from tailing dump #1 is flowing out to the land surface with the average flow rate of 131 m^3 day^{-1} (period of measurements: Nov. 2008–Sep. 2009, n = 46), contaminating downstream paddy fields where water table is higher than the ground surface. The paddy fields are prohibited to be cultivated because of soil contamination by Cd, Pb and Zn (KME 2003). A drainage with the average flow rate of 230 m^3 day^{-1} (period of measurements: Nov. 2008–Sep. 2009, n = 46) is flowing out through an abandoned mine adit near the tailing dump #2 and then joins the leachate from dump #2 (MIRECO 2009a).

The abandoned Buddeun Au-Ag-Cu mine consists of hydrothermal quartz veins in granitic rocks. Major ore minerals are chalcopyrite, sphalerite, pyrite, arsenopyrite, galena, electrum, argentite and native silver; gangue minerals include quartz, rhodochrosite, ankerite, and calcite (Park et al. 1988). Major sources of water contamination are two large tailing dumps (#1 and #2) with the area of 14,779 m^2 and

Table 1
Mining history, major ore types, and contamination sources of studied mines.

Name	Province	Operation period	Ore type	Water contamination	
				Sources	Contaminants
Buddeun	Gyeongbuk	1915–1989	Hydrothermal; Au-Ag-Cu	Tailings and adit water	Al, Cd, Cu, Mn, Pb, Zn
Dangdu	Chungbuk	1937–1992	Skarn; Au-Ag-Pb-Zn	Adit water	Cd
Gomyeong and Dongbo	Gangwon	1978–1993	Hydrothermal; Au-Ag	Tailings and adit water	As, Mn
Sambo	Gyeonggi	1956–1991	Hydrothermal; Pb-Zn-Ba	Tailings and adit water	Al, Cd, Mn, Pb, Zn
Samgwang	Chungnam	1928–1996	Hydrothermal; Au-Ag-Pb-Zn	Tailings and adit water	As, Mn
Sangdeok	Gangwon	Under operation	Anthracite coal	Waste rock	Al, Cd, Fe, Mn, Pb, Zn