



Field investigations of passive and active processes for acid mine drainage Treatment: Are anions a concern?

Adil Alsaiari, Hao L. Tang*

Environmental Engineering Program, Indiana University of Pennsylvania, Indiana, PA 15705, USA



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ABSTRACT

Acid mine drainage (AMD) treatment processes were investigated for removal of anions including bromide, fluoride, chloride, nitrate, phosphate, and sulfate. The study selected a passive process consisting of an aerated lagoon and aerobic wetlands and an active process composed of a lime dosing silo and a sedimentation pond. The results showed both processes were capable of removing fluoride, nitrate, and phosphate. The passive process removed 21% bromide and 26% chloride as a result of biotic mechanisms which could not be achieved by the active process. Additionally, higher temperature and longer hydraulic retention time promoted the removal of many anions. With abiotic mechanisms such as precipitation, the active process was capable of removing sulfate, which was found unsuccessful by the passive process due to unfavorable conditions for the growth of sulfate reducing bacteria. Although the anions in the AMD waters were not high enough to generate substantial environmental and health concerns, their removals, especially the removals of bromide, phosphate and sulfate, would reduce burdens at downstream drinking water treatment plants.

1. Introduction

Formation of acid mine drainage (AMD) originates from the oxidation of sulphide mineral ores (e.g. pyrite ore (FeS_2) – commonly known as fool's gold) that are exposed to the environment by intensive mining activities, releasing Fe(II) , SO_4^{2-} , and H^+ . Facilitated by sulfur-oxidizing bacteria, the produced Fe(II) reacts with O_2 to form Fe(III) , which in turn acts as a much stronger oxidant than O_2 for expedited pyrite oxidation, exacerbating the generation of acidity and mobilization of iron. In the meanwhile, acidification of mining water provokes the release of a large number of other toxic elements from rocks including Ag, As, Bi, Cd, Co, Cu, Hg, Mo, Ni, Pb, Ru, Sb, Se, Sn, Te, and Zn (Iakovleva et al., 2015). Without pumps to keep the water tables artificially low, the situation can be worse after mines are closed or abandoned (Simate and Ndlovu, 2014). In the United States, there are about 557,000 abandoned mines, many of which are active sources of AMD (RoyChowdhury et al., 2015). The toxic metals and acidity in AMD discharges generate serious environmental concerns as they may endanger aquatic organisms, destroy ecosystems, corrode infrastructures, and pollute water sources (Obrique-Contreras et al., 2015; Nleya et al., 2016).

Along with mobilization of cations including those metals and H^+ , anions such as fluoride, chloride, bromide, etc. could also be dissolution

products as those halide-bearing minerals in rocks weather or interact with AMD (Turner et al., 2005; Nagaraju et al., 2017). The U.S. Geological Survey Coal Quality Database reveals that coals are notable source of halogens with approximately 108 ppm F, 1290 ppm Cl, and 54 ppm Br. These elements may end up in AMD after a series of chemical and biological reactions, imposing negative impacts on water sources. For instance, a discharge of bromide to surface waters, even at low concentrations, can alter the formation and speciation of disinfection by-products (DBPs) at downstream drinking water treatment plants (DWTPs) significantly (Wang et al., 2017). During the water disinfection – an inevitable step for inactivation of microorganisms at DWTPs, an increased bromide promotes the formation of hypobromous acid, which then reacts with organic matter to form more toxic brominated DBPs than their chlorinated analogues (Hua et al., 2006). Therefore, additional concerns may arise from deterioration of product water quality at DWTPs drawing water from AMD-impacted water sources as a result of discharges of those anions.

The eastern United States has a long history of dealing with AMD, both passively and actively. The passive treatment was originally developed in the Appalachian coalfield (Skousen et al., 2017), and it utilizes the chemical, biological, and physical removal processes that occur naturally in the environment (Clyde et al., 2016). Wetlands as a low-cost biogeochemical system is highlighted as a typical passive

* Corresponding author.

E-mail address: htang@iup.edu (H.L. Tang).

ecological restoration process for AMD, as it has the ability of accumulating large quantities of metals at economic advantages of no chemical inputs and low maintenance needs (Humphries et al., 2017). In contrast to passive treatment, another category of treatment method for AMD is the active process, which is usually more effective regarding the process time by direct application of alkaline chemicals (e.g., limestone (CaCO_3), lime (CaO), hydrated lime (Ca(OH)_2), soda ash (Na_2CO_3), caustic soda (NaOH), etc) to AMD streams for precipitation of metals and raise of pH (Kefeni et al., 2017). Although selecting an appropriate treatment process depends on a variety of factors, the decisions are usually prioritized based on removal of cations. Processes are rarely designed or optimized for removal of anions, especially for the halides. In the meanwhile, the monovalent anionic species are a big challenge to remove at downstream DTWPs. Since they do not settle out or form complexes as metals do, conventional coagulation and sedimentation that are employed at DTWPs are ineffective in removing these anions. Therefore, it would be desirable to have those anionic species taken care of by processes ahead of the downstream DTWPs, preferably at the AMD sites, in order to attenuate their impacts on water quality and human health. To date, there is inadequate information on the capability of AMD treatment processes on removal of anions. Studies on evaluating the potential of AMD treatment processes on removing the anionic species of concern are needed.

In this research, we performed field investigations over a 12-month period at one passive and one active AMD treatment sites in Indiana County – a historic center of mining activities in Pennsylvania, USA. The objectives of the research were to (1) assess the feasibility of using the existing passive and the active AMD treatment process to remove the anionic species of concern, including bromide, fluoride, chloride, nitrate, phosphate, and sulfate; and (2) discuss whether these anions are of any environmental concern after such treatment. The study extends current knowledge of ecological engineering approaches for control of undesirable anionic species in AMD.

2. Materials and methods

2.1. Site description

The passive process under investigation was the Tanoma AMD discharge site (N 40° 41' 36", W 79° 2' 57") in Indiana County, Pennsylvania. The site map is shown in Fig. 1a. As an abandoned mine since 1960s, the site is the first major pollution input to the Crooked Creek Watershed, impacting Crooked Creek with approximately 23 tons of iron each year if untreated. An ecological restoration effort was accomplished by employing an aerated lagoon followed by aerobic wetlands for iron removal (Fig. 2a). The wetlands consist of a series of shallow ponds that contain emergent-rooted wetland vegetation. In addition to wetlands that are considered as a typical passive process, the aerated lagoon is also highlighted as “passive”, because the air is provided by a water-powered air compressor (i.e. trompe) with no moving parts. The aeration in the lagoon drives off carbon dioxide dissolved in the water to raise pH, and adds oxygen to the water, which facilitates the oxidation of Fe(II) and formation of Fe(OH)_3 that settles in the lagoon. The following aerobic wetlands function as an additional layer of filter to catch the unsettled solids, benefiting from the established vegetation growing in the shallow wetland ponds. There is also a carbonate rock drain at the Tanoma site, serving as an emergency spillway.

The active process under investigation was an AMD discharge site in the Waterworks Conservation Area (N 40° 35' 55", W 79° 7' 26") in Indiana County, Pennsylvania. The site map is shown in Fig. 1b. The mine was abandoned in 1967 after 20 years of service. The AMD discharge enters nearby Two Lick Creek, impacting the Creek with 17 tons of iron and aluminum if untreated. The remediation effort includes a 75-ton lime dosing silo over the AMD stream followed by a sedimentation pond (Fig. 2b). The process is considered as “active”, because

chemicals and man powers are needed for its operation and maintenance.

2.2. Sample collection

Grab samples were collected approximately bimonthly from a total of 6 sampling locations of both passive and active processes over a one-year period from November 2016 to October 2017. As denoted in Fig. 2, the sampling locations were distributed along the treatment train of each process. Sampling Point (SP) A and SP D were the raw AMD influents to the processes without any treatment. Since each process consisted of two stages, SP B and SP E were the effluents right after Stage 1 treatment. For the passive process, Stage 1 treatment was the aerated lagoon; and for the active process, it was the lime dosing. Stage 2 treatment included aerobic wetlands and sedimentation pond for the passive and active processes, respectively. SP C and SP F were the final effluents after the Stage 2 treatment and prior to the discharge to the surface waters. The samples were collected in 500-mL polypropylene bottles without headspace, stored in an ice box, transported to the laboratory, and kept refrigerated at 4 °C before analyses of anions.

2.3. Sample analysis

pH was analyzed by a Vernier pH probe (Vernier Software & Technology, Oregon, USA) at the sites. For analysis of anions including bromide, fluoride, chloride, nitrate, phosphate, and sulfate, samples were pre-filtered with 0.45- μm syringe filters and then determined by a Metrohm 850 ion chromatograph (Metrohm AG, Herisau, Switzerland) with a Metrosep A Supp 5 column. The eluent was 64 mM sodium carbonate/20 mM sodium bicarbonate solution. The filtered samples were also analyzed for total organic carbon (TOC) by a TOC analyzer (Shimadzu TOC-LCSH, Shimadzu Corporation, Japan).

2.4. Statistical analysis

The obtained anions and TOC concentrations were validated against a comprehensive set of procedural blanks, matrix spikes, and replicate samples. The data used in the study were averages of duplicate analyses. Descriptive statistics on removal percentages were then calculated for each parameter by each process over the period of investigation. Statistical analyses were implemented using Minitab 15 (Minitab Inc, Pennsylvania, USA). A nonparametric Mann–Whitney procedure was used and the level of significance was set to $p < 0.05$ (Tang et al., 2012).

3. Results and discussion

3.1. Removal of halides

3.1.1. Bromide

The starting water quality parameters with halide concentrations prior to the passive (Tanoma site) and active (Conservation site) AMD treatment processes are presented in Table 1. The data presented were averages of samplings between November 2016 and October 2017. The bromide, fluoride, and chloride concentrations at the Tanoma site were 0.088, 0.69, and 16.6 mg/L, respectively. The Conservation site was found with lower bromide (0.045 mg/L) and chloride (12.7 mg/L) compared to the Tanoma site, while the fluoride concentration (0.63 mg/L) was similar. The chloride/bromide ratios of 188 and 282 for the two sites were comparable to those of shallow ground water (Davis et al., 1998; Katz et al., 2011). Considering the bromide concentrations in inland surface waters in the United States can be as low as 0.014 mg/L (VanBriesen, 2014), both AMD waters appeared to contain non-negligible amounts of bromide. Moreover, the TOC of AMD waters (11.6 and 1.2 mg/L for Tanoma and Conservation sites, respectively) suggests the presence of organic DBP precursors (Liu et al.,

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