



Enhancement of particle aggregation in the presence of organic matter during neutralization of acid drainage in a stream confluence and its effect on arsenic immobilization



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HIGHLIGHTS

- Organic matter associated to Fe and Al phases during neutralization of acid drainage.
- Dissolved organic matter promoted aggregation of Fe phases at pH 4.5.
- Particle size distributions' multimodality depended on organic matter type.
- Aggregation of humic acids removed metals in acidic conditions (pH ~3).

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ABSTRACT

Acid drainage (AD) is an important environmental concern that impacts water quality. The formation of reactive Fe and Al oxyhydroxides during the neutralization of AD at river confluences is a natural attenuation process. Although it is known that organic matter (OM) can affect the aggregation of Fe and Al oxyhydroxides and the sorption of As onto their surfaces, the role of OM during the neutralization of AD at river confluences has not been studied. Field and experimental approaches were used to understand this role, using the Azufre River (pH 2) - Caracarani River (pH 8.6) confluence (northern Chile) as model system. Field measurements of organic carbon revealed a 10–15% loss of OM downstream the confluence, which was attributed to associations with Fe and Al oxyhydroxides that settle in the river bed. Laboratory mixtures of AD water with synthetic Caracarani waters under varying conditions of pH, concentration and type of OM revealed that OM promoted the aggregation of Fe oxyhydroxides without reducing As sorption, enhancing the removal of As at slightly acidic conditions (pH ~4.5). At acidic conditions (pH ~3), aggregation of OM - metal complexes at high OM concentrations could become the main removal mechanism. One type of OM promoted bimodal particle size distributions with larger mean sizes, possibly increasing the settling velocity of aggregates. This work contributes to a better understanding of the role of OM in AD affected basins, showing that the presence of OM during processes of neutralization of AD can enhance the removal of toxic elements.

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1. Introduction

The exposure of sulfide minerals from mining activities – mainly Fe sulfides such as pyrite and pyrrhotite – to oxygen and water produces a drainage of low pH and high concentrations of

sulfate and metals like Fe and Al, commonly referred as Acid Drainage (AD) (Amos et al., 2015; Lindsay et al., 2015). The dissolution of these minerals can lead to the release of toxic metals like As, Pb and Zn as they are often associated to these Fe sulfides, for example, through substitution of metals in their structure (Cheng et al., 2009; Nordstrom et al., 2015) or incorporated as nanoparticles (Deditius et al., 2011). Whether natural or anthropogenic, the presence of metals such as As, Pb and Zn in riverine systems poses a challenge for sustainability, as they cause several negative impacts in ecosystems (Byrne et al., 2012) and human health (Abdul et al., 2015; Järup, 2003). This is particularly relevant for water stressed watersheds, like those in northern Chile (Aitken et al., 2016), in which water quality issues may limit availability of water resources.

The most widespread scheme for AD remediation is neutralization, in which the addition of alkaline materials results in a raise in pH that promotes the precipitation of Fe and Al oxyhydroxides (Johnson and Hallberg, 2005). These particles are able to aggregate into larger flocs (Duan and Gregory, 2003) with potential to settle (Akcil and Koldas, 2006). These solids are also capable of sorbing toxic metals like As (Giles et al., 2011; Lizama A. et al., 2011; Mohan and Pittman, 2007), allowing for the immobilization of trace metals (Lee et al., 2002). Neutralization of AD through mixing with more alkaline streams, and subsequent Fe and Al oxyhydroxides formation, has played a key role in natural attenuation of metals contamination in watersheds affected by AD, as in the Tinto River – Odiel River (Asta et al., 2015), the Cement Creek – Animas River (Schemel et al., 2007), the Amous River – Reigous Creek (Adra et al., 2013) and the Azufre River – Caracarani River confluences (Guerra et al., 2016a).

The fate of As and other contaminants during these processes is controlled by a complex interaction of hydrologic, hydrodynamic and chemical factors. Hydrologic and hydrodynamic conditions lead to specific time frames and zones in which precipitation of Fe and Al oxyhydroxides occurs (Guerra et al., 2016a, 2016b). Chemical conditions leading to the formation of primary particles and their flocs affect their physical and chemical properties, such as particle size and surface charge (Wang et al., 2014). For example, ferrihydrite – an amorphous Fe oxyhydroxide present in acid waters – precipitates as nanoparticles of ~5 nm (Giles et al., 2011) that can aggregate into flocs ranging from <1 μm to hundreds of μm (Lo and Waite, 2000), with increasing Fe concentrations and pH promoting the formation of larger flocs (Tsubaki et al., 2012). Aggregate size is an important property of Fe particles, as it controls their settling velocity (Droppo et al., 2000) and their removal from the water column. Therefore, it is relevant to study the size distribution of Fe particles aggregates and the factors controlling the aggregation process. Despite this, few studies have considered the analysis of the particle size distribution (PSD) of the byproducts of acid drainage neutralization (Abarca et al., 2017; Guerra, 2016).

The presence of organic matter (OM) also influences the fate of As. Several authors have reported that OM interacts with systems containing Fe, Al and As. For example, dissolved organic carbon (DOC) has been shown to sorb onto Fe and Al oxyhydroxide (Gu et al., 1995), displacing metals to the bulk solution and increasing their mobility (Bauer and Blodau, 2006; Ko et al., 2004). Organic matter can also change the surface charge of these particles and their flocculation behavior (Angelico et al., 2014). The removal of metals by the sedimentation of Fe and Al oxyhydroxides in natural and engineered systems requires (1) metals to be sorbed onto these solid phases and that (2) the formed flocs are large enough to settle in the river bed or in a sedimentation tank. This shows that OM could greatly affect the fate of metals in systems affected by AD.

Most research regarding the effects of OM in the fate of As focused either on chemical interactions (sorption and desorption of

As, and fractionation between dissolved, colloidal and particulate phases) or physical properties of particles (surface charge, flocculation properties). Wang et al. (2014) briefly studied the effects of OM in the removal of As through settling of particles, combining both approaches, although not in a context of acid drainage. Furthermore, studies involving particle size characterization have focused mainly in the nm size range, which is not relevant for removal by sedimentation because of their negligible settling velocities. For example, ferrihydrite particles in the nm range can take hours to days to fall a 10 cm height (Tsukimura et al., 2010). Therefore, emphasis must be placed in μm sized aggregates when considering removal by sedimentation.

This work aims to study the effects of Dissolved Organic Matter (DOM) in the removal of As by settling of Fe and Al oxyhydroxide aggregates in the μm size range after neutralization of AD in river confluences. In particular, the effects of DOM in (1) the PSD of Fe and Al oxyhydroxide particle aggregates and (2) the partitioning of As in the dissolved (<0.45 μm) and particulate fractions were studied. This knowledge could be useful to propose engineered solutions to enhance removal of As in natural attenuation processes like river confluences or during treatment schemes.

2. Materials and methods

2.1. Study site

Our study model was the Azufre River – Caracarani River confluence, which is located in the upper basin of the Lluta River, northern Chile (Fig. 1a). The Azufre River has low pH (<2), high electric conductivity ($\text{EC} > 9 \text{ mS cm}^{-1}$) and high metal concentrations ([Al] ~ 200 mg L^{-1} , [Fe] ~ 90 mg L^{-1} , and [As] ~ 2 mg L^{-1}) due to contributions from geothermal springs and the oxidation of sulfur tailings from an abandoned sulfur mine (Leiva et al., 2014). The Caracarani River (pH ~ 8.5, [Al] ~ 0.5 mg L^{-1} , [Fe] ~ 0.5 mg L^{-1} , and [As] ~ 0.1 mg L^{-1} , total alkalinity between 20 and 230 $\text{mg CaCO}_3 \text{ L}^{-1}$) neutralizes the Azufre River, promoting the formation of Fe and Al oxyhydroxides that scavenge toxic elements like As (Guerra et al., 2016a, 2016b).

2.2. Field measurement of physicochemical parameters and water sampling

A field campaign in May 2015 focused on OM concentrations, measurement of hydro-chemical parameters (pH, electric conductivity and turbidity) and sampling for metal and Particle Size Distribution (PSD) in order to understand the role of OM on the formation and aggregation of Fe and Al oxyhydroxides. In situ measurements were performed in both rivers upstream the confluence and along 3 cross-sections downstream the confluence (50, 100 and 200 m, Fig. 1b), with 7–12 measurements per cross-section. Sample collection was performed at one point per cross-section.

Samples for total metals, dissolved metals (<0.45 μm using Nylon Syringe Filters, Agela Technologies, DE, USA) and total organic carbon (TOC) were collected on field, while non-filtered samples for dissolved organic carbon were collected and filtered within 24 h through 0.45 μm MCE filters (Merck Millipore, Merck, Germany) on a glass filtration apparatus (Kontes Ultraware, Kimble Chase, TN, USA). Samples for dissolved and total metal analyses were stored in 15 and 50 mL polypropylene Falcon tubes, respectively, and acidified to pH 2 with HNO_3 (1:1 v/v, EMSURE, Merck, Germany). Samples for organic carbon analyses were stored in 150 mL glass bottles and acidified to pH 2 with H_3PO_4 (1:1 v/v, EMSURE, Merck, Germany). Additional non-filtered non-acidified samples were stored in 350 mL polyethylene bottles and measured for PSD and Total Suspended Solids (TSS).

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