



## Hydrogeochemical niches associated with hyporheic exchange beneath an acid mine drainage-contaminated stream



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### SUMMARY

Biological low-pH Fe(II)-oxidation creates terraced iron formations (TIFs) that remove Fe(III) from solution. TIFs can be used for remediation of acid mine drainage (AMD), however, as sediment depth increases, Fe(III)-reduction in anoxic subsurface areas may compromise treatment effectiveness. In this study we used near-surface electrical resistivity imaging (ERI) and *in situ* pore-water samplers to spatially resolve bulk conductivity changes within a TIF formed in a stream emanating from a large abandoned deep clay mine in Cambria County, Pennsylvania, USA. Because of the high fluid electrical conductivity of the emergent AMD (1860  $\mu\text{S}$ ), fresh water (42  $\mu\text{S}$ ) was added as a dilution tracer to visualize the spatial and temporal extent of hyporheic exchange and to characterize subsurface flow paths. Distinct hydrogeochemical niches were identified in the shallow subsurface beneath the stream by overlaying relative groundwater velocities (derived from ERI) with pore-water chemistry profiles. Niches were classified based on relatively “fast” versus “slow” rates of hyporheic exchange and oxic versus anoxic conditions. Pore-water concentrations and speciation of iron, pH, and redox potential differed between subsurface flow regimes. The greatest extent of hyporheic exchange was beneath the center of the stream, where a shallower (<10 cm) Fe(II)-oxidizing zone was observed. Meanwhile, less hyporheic exchange was observed near the channel banks, concurrent with a more pronounced, deeper (>70 cm) Fe(II)-oxidizing zone. At these locations, relatively slower groundwater exchange may promote biotic Fe(II)-oxidation and improve the long-term stability of Fe sequestered in TIFs.

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### 1. Introduction

Environmental impacts associated with active and historic coal mining activities impair or threaten ecosystems throughout much of the Appalachian region of the United States. The Appalachian coal fields extend over 1600 km in length from Alabama to Pennsylvania; however, a disproportionate number of mine-impacted streams are found in Pennsylvania because of the regional coal geology (PA DEP, 1998). Ecological impairment to downstream water bodies arise as sulfide-bearing coal seams and surrounding

strata are subject to chemical and physical weathering processes that generate acidity, frequently referred to as acid mine drainage (AMD) or, more specifically, coal mine drainage (CMD). CMD is characterized by elevated concentrations of dissolved metals (primarily iron, aluminum and manganese) and sulfate, and high fluid electrical conductivity (Kirby and Cravotta, 2005; Cravotta, 2008). Depending on site geology and hydrogeology, the pH and acidity of CMD can vary from low-pH and net-acid to circumneutral-pH and net-alkaline.

Our research focuses on systems where anoxic, low-pH, Fe(II)-rich AMD emerges from artesian springs, enters a stream, and immediately begins to react with the atmosphere and stream water. In these hydrogeochemical settings, biological low-pH Fe(II) oxidation leads to the precipitation of Fe(III) in the form of ferric sulfate-(hydr)oxide minerals such as schwertmannite [ $\text{Fe}_8\text{O}_8(\text{OH})_6(\text{SO}_4)$ ] (Bigham et al., 1996) or ferric oxyhydroxides such as ferrihydrite. Biological low-pH Fe(II) oxidation, therefore,

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is an attractive option for AMD remediation because it can remove substantial amounts of Fe(III) before conventional neutralization processes. For example, limestone beds are common in passive treatment systems; however armoring of limestone and/or clogging of the limestone beds due to precipitation of Fe(III) minerals limits performance, but can be minimized by first removing Fe(III) via low-pH Fe(II) oxidation. Furthermore, the process decreases pH, which can promote alkalinity generation from limestone.

In natural or engineered settings, biologically mediated low-pH Fe(II) oxidation will create terraced iron formations (TIFs). The morphology of these TIFs is dependent on site specific hydrodynamic conditions (Veysey Li and Goldenfeld, 2008), water chemistry, and kinetics of Fe(II) oxidation and Fe(III) precipitation. Under certain conditions TIF sediment depth can become substantial. For example, Fe-rich sediments >1 m in depth have been reported for TIFs formed from CMD in Appalachia (DeSa et al., 2010) and AMD in the Iberian Pyrite Belt (Sánchez España et al., 2007). For TIFs to remain as part of an AMD treatment system, the immobilization of Fe (via oxidative precipitation at the surface) must exceed any possible remobilization in the subsurface (via reductive dissolution). Previous studies have shown that hydrodynamic conditions (velocity and shear) in surface streams control biofilm architecture (Bottacin-Busolin et al., 2009) and microbial community composition (Macalady et al., 2008). Geochemical conditions also exert an effect on microbial community composition, especially in extreme low-pH environments (Baker and Banfield, 2003). However, hydrodynamic or geochemical conditions alone often cannot fully explain microbial diversity (Flores et al., 2012). Hydrogeochemical niches can be identified by overlaying hydrodynamic and geochemical conditions, and may improve our ability to understand and predict microbial diversity. These data are relatively straightforward to obtain in surface streams but are far more difficult to obtain in the subsurface.

In-stream measurements, or measurements from limited available wells, make characterization of groundwater-surface water exchange processes difficult. Geophysical tools are starting to become commonly used in these systems; in particular, electrical resistivity is now frequently used to monitor exchange and understand flow paths (Ward et al., 2010; Cardenas and Markowski, 2010; Coscia et al., 2011; Toran et al., 2012; Doro et al., 2013). Electrical methods have long been used to image groundwater contamination associated with AMD (Merkel, 1972; Ebraheem et al., 1990; Rucker et al., 2009); however, application of these techniques to quantify hyporheic exchange in an AMD-impacted river, to the best of our knowledge, has not yet been considered. Here, we capitalize on this previous work in a new setting.

The objectives of the current study were to (1) resolve the spatial extent of oxic and anoxic conditions within a TIF, (2) to characterize the subsurface geochemistry in these zones, and (3) identify hydrogeochemical niches in the shallow subsurface. Geophysical techniques such as near-surface electrical resistivity imaging (ERI) used with a conductive tracer can visualize hyporheic exchange beneath a stream (e.g., Nyquist et al., 2008; Ward et al., 2010). In the current study, we used low conductivity water as the tracer because of the intrinsically high conductivity of the AMD-impacted stream. ERI methods coupled with a constant-rate, long-term tracer addition provided a 2-dimensional view of subsurface hydrodynamic conditions beneath the stream and the spatial and temporal extent of hyporheic exchange. Subsurface pore-water samplers were then deployed along electrode transects to measure geochemistry in areas of contrasting extent of hyporheic exchange. Combined, these approaches identified a variety of hydrogeochemical niches beneath the AMD-impacted stream.

## 2. Methods and materials

### 2.1. Site description

The AMD field site was located near Brubaker Run (BR) (40°37'1.42"N; 78°28'35.76"W), a small tributary of Clearfield Creek (Fig. 1); located approximately 3.2 km northwest of Altoona, PA and 2 km east of Dean, PA. The Dean Clay Mine was a 1,011,800 m<sup>2</sup> underground kaolinite mine that flooded after being abandoned (ca. 1950). The Dean Clay Mine now discharges AMD to Brubaker Run as a large artesian spring that was located above a former air shaft in the collapsed entry to the mine. Water flows downhill from the artesian spring across a large TIF (6000 m<sup>2</sup>) in a highly uncontrolled, multi-directional manner, except for one large channel that was the former adit into the mine. This large channel was used for all tracer studies because of its well-constrained, uni-directional flow characteristics (Figs. 1 and 2). A second artesian spring (referred to as BR80) is located along the left-bank of this channel. All left and right-bank designations refer to a downstream orientation. The water chemistry of BR80 and the uphill artesian spring are essentially identical and BR80 is thought to emerge from the “toe” of the mine pool (versus up through the former air shaft). A large fraction of the water from the uphill spring is captured by the adit channel. The top of the uni-directional portion of this channel is approximately 20 m from the center of the uphill spring (and approximately 5 m lower in elevation). The uni-directional portion of the channel then runs for 60 m before dispersing in a multi-directional manner across a lower series of iron terraces.

Because of the high flow and high conductivity of the Dean Clay Mine discharge, a “dilution tracer” was selected containing low conductivity water collected from an adjacent headwater creek (see Table 1). Laurel Run is located near Dysart, PA, approximately 4.8 km from Brubaker Run and was never impacted by coal-mining activities. A water-hauling company was contracted to collect water from Laurel Run and deliver it via a 3000-gallon water truck parked at the test site. A coarse screen was placed on the hose during collection from Laurel Run, and the water was then pumped through 0.5 mm filter bags when filling the parked water truck. Water was pumped from the parked tanker through a Neptune flowmeter that was continuously monitored and maintained at 6.3–6.4 L/s for the complete 10 h tracer addition period. With no tracer addition, we estimated that approximately 3.5 L/s was entering the channel just upstream of the tracer addition zone (0 m datum), and we measured a flow of 15.8 L/s at the end of the uni-directional portion of the channel (60 m downstream). Water was pumped approximately 90 m (~10 m elevation gain) from the truck to the injection zone (BR100). The tracer addition zone was located essentially at the start of the uni-directional portion of the channel and is referred to as the 0 m datum for stream locations in this study. The injection zone consisted of a 55-gallon plastic drum equipped with a 3-in. diameter stand pipe positioned to deliver water to the bottom of the drum. Water then overflowed the drum, ran onto plywood placed beneath the drum and then into the channel (Fig. 2A). Wading surveys to measure stream flow, using a Marsh-McBirney velocimeter and wading rod, were conducted 60 m below the injection zone before and during tracer injection to evaluate how much of the tracer exited the uni-directional portion of the channel.

### 2.2. Electrical resistivity imaging

ERI data were collected on a network of electrodes installed in three transects oriented laterally across the stream channel using a 10-channel Syscal Pro Resistivity Meter (IRIS Instruments,

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