

# Arsenic cycling within the water column of a small lake receiving contaminated ground-water discharge

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## Abstract

The fate of arsenic discharged from contaminated ground water to a small, shallow lake at a hazardous waste site was examined to understand the role of iron (hydr)oxide precipitation–dissolution processes within the water column. Field and laboratory observations indicate that arsenic solubility was controlled, in part, by the extent of ferrous iron oxidation–precipitation and arsenic sorption occurring near the lake chemocline. Laboratory experiments were conducted using site-derived water to assess the impact of these coupled processes on the removal of dissolved arsenic from the water column. The measured concentration of organic carbon from epilimnetic and hypolimnetic water sampled from the lake was approximately 1.3 mM and 17.0 mM, respectively. Experiments conducted with these samples along with synthetic controls containing no organic carbon demonstrated that observed rates of formation and crystallinity of the precipitated iron (hydr)oxide were dependent on the concentration of organic carbon in the lake water. Increasing dissolved organic matter concentration did not significantly interfere with ferrous iron oxidation, but inhibited iron (hydr)oxide precipitation and subsequent sorption of arsenic. For experiments using water sampled from the lake hypolimnion there was a strong relationship between the fraction of precipitated iron and the fraction of sorbed arsenic. Laboratory- and field-derived iron (hydr)oxide precipitates were characterized to evaluate mineralogy and arsenic distribution. In-situ suspended solids and precipitates formed in laboratory experiments using hypolimnetic water were identified as poorly crystalline 2-line ferrihydrite. These solids were readily dissolved in the presence of dithionite indicating that elevated dissolved iron and arsenic observed in the hypolimnion resulted, in part, from in-situ reductive dissolution of settling 2-line ferrihydrite near the sediment–water interface. These observations support the contention that the levels of dissolved arsenic observed in the shallow lake can be attributed to ground-water discharge and internal recycling of arsenic within the water column. The efficiency of the process resulting in iron (hydr)oxide precipitation and arsenic sorption limits the downgradient export of arsenic derived from ground-water discharge.

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## 1. Introduction

Ground-water discharge can serve as a significant source of water inflow into surface water bodies (Winter et al., 1998; <http://water.usgs.gov/pubs/circ/circ1139/>)

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[pdf/circ1139.pdf](#)). Contaminated ground water may be a significant source of contaminant load into surface water and sediments and it is estimated that within the United States approximately 75% of sites regulated under the Resource Conservation and Recovery Act and the Comprehensive Environmental Response, Compensation, and Liability Act ('Superfund') are located within a half mile of a surface-water body (Tomassoni, 2000; Biksey and Gross, 2001). Successful restoration of contaminated watersheds depends on reliable assessment of the source(s) of contaminant flux and the processes controlling contaminant speciation within the boundaries of the impacted water body. This poses a significant challenge for sites impacted by inorganic contaminants such as arsenic where contaminant mass is conserved and physicochemical speciation may change dramatically within a given site.

Several studies have been conducted to examine the fate of arsenic and other metals within the water column of lakes (e.g., Aggett and O'Brien, 1985; Aggett and Kriegman, 1988; Seyler and Martin, 1989; Balistrieri et al., 1994; Spliethoff et al., 1995; Sohrin et al., 1997; Senn and Hemond, 2002). In many of these studies it has been shown that the fate of arsenic is tied to the chemical cycling of iron. Iron may be continuously recycled within the lake for systems in which anoxia develops within the hypolimnion or underlying sediments. Reduced iron generated in the lower regions of a lake can diffuse upward within the water column and re-precipitate upon contact with oxygen or other oxidants (e.g., Sholkovitz and Copland, 1982; Davison et al., 1982). Dissolved arsenic within the water column can partition to the newly formed iron (hydr)oxides either through coprecipitation at the time of formation or adsorption to settling iron (hydr)oxide particles (Seyler and Martin, 1989; Balistrieri et al., 1994; Spliethoff et al., 1995; Sohrin et al., 1997; Senn and Hemond, 2002). Arsenic partitioned to the settling iron (hydr)oxides can subsequently be remobilized during reductive dissolution within the hypolimnion or following sedimentation depending on the spatial distribution of reducing conditions within the lake system. This 'natural' biogeochemical cycle will overlap with hydrologically driven contaminant fluxes across the ground water–surface water interface and can cause misleading interpretations of the environmental risk that is directly attributable to discharge of contaminated ground water (see Summary and conclusions, Sholkovitz and Copland, 1982).

The purpose of this study was to evaluate the fate of arsenic within a wetland system immediately down-gradient from a Superfund site with arsenic-contami-

nated soils and ground water resulting from industrial activities dating back to the mid-1800s (Aurilio et al., 1994a; Davis et al., 1994; Wick and Gschwend, 1998). The fate of arsenic in this system is coupled to the chemical cycling of iron, sulfur and carbon due to the characteristics of the ground-water plume discharging into the surface water system. This manuscript specifically addresses the fate of arsenic derived from anoxic, contaminated ground water following discharge into a small, shallow lake. Experiments were conducted using water samples collected from the lake to assess the in-situ rate of ferrous iron oxidation within the lake and the degree of arsenic uptake onto iron (hydr)oxide precipitates that formed. Iron (hydr)oxide precipitates collected from experimental systems and from various depths within the lake were characterized to assess the influence of in-situ water chemistry on precipitate mineralogy and stability relative to arsenic desorption and/or re-dissolution. A companion manuscript (Wilkin and Ford, 2006-this issue) describes arsenic partitioning to sediments within this lake due, in part, to deposition of arsenic-bearing iron (hydr)oxides that form within the water column.

## 2. Methods

### 2.1. Study location

The study location is down gradient from a Superfund site in Massachusetts in which historical on-site disposal of arsenic-bearing wastes has resulted in leaching of arsenic into site ground water (Durant et al., 1990; Davis et al., 1994). A portion of the ground water plume discharges into a constructed shallow lake that also receives surface water inputs from a perennial stream (Halls Brook) on its western edge and an intermittent runoff channel at its northwestern end (Fig. 1). The constructed lake was built in the early 1970s for flood control and is referred to as the Halls Brook Holding Area (HBHA) Pond throughout this report. The HBHA Pond has a maximum depth of approximately 5 m and it discharges into a heavily vegetated wetland area. The HBHA Pond discharge reconstitutes Halls Brook, which meanders through the wetland until discharge into the Aberjona River.

Independent estimates of water flux into the HBHA Pond indicate that approximately 7–60% of the water input is derived from ground water throughout the year (Aurilio et al., 1994b; Wick et al., 2000). Based on salt balance calculations Wick et al. (2000) have estimated that a contaminated ground-water plume (denoted 'Q<sub>high</sub>' by the authors) contributes approximately 1–

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