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Sorption and redox processes controlling arsenic fate and transport in a stream impacted by acid mine drainage

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Abstract

Reigous acid creek originating from the Carnoulès tailings impoundment supplies high concentrations of arsenic under soluble (up to ~4 mg/l) and particulate (up to 150 mgAs/g) phases to the Amous river, situated at the drainage basin of the Rhône river (Southern France). The metalloid is present as As(III) (>95%) in Reigous creek water while As(V) predominates (50–80%) in the solid phase, i.e. schwertmannite. At the confluence between acid (pH<5) creek and alkaline Amous river, As(III) concentrations decrease ten-fold through dilution and formation of As-rich ferrihydrite (As/Fe=0.02–0.1) containing 10–30% As(III). However, these attenuation processes are not efficient in the summer heatwave of 2003 since As concentrations in Amous river water (≥20 μg/l) and As/Fe ratios in particulate matter (≥0.07) are closed to those of Reigous creek (≤22 μg/l and ≤0.02, respectively) or even higher. Downstream the confluence, processes involved in the transport of aqueous As along Amous river flowpath vary seasonally. Arsenic is transported conservatively in the aqueous phase away from the confluence in the cooler months; thus, dilution by unpolluted tributaries is the only process that decreases As concentrations. However, As(III) is rapidly oxidized and As(V) remains in solution. In contrast, during the warm season, desorption from As-rich sediment occurs which results in an increase of As(V) and As(III) concentrations along Amous river flow until they reach up to ~20 μg/l each. Therefore, Amous river seems not to be totally recovered from mine-related arsenic contamination after 3.5 km and may affect freshwater resources further downstream.

Keywords: Arsenic; Speciation; AMD; Transport processes; Mine-impacted river

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1. Introduction

The concentrations of arsenic in acid mine drainage (AMD) can reach hundreds of mg/l, as a result of the oxidation of As-rich sulfides (Williams, 2001). Such

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As concentrations are potentially harmful for the aquatic ecosystem located downstream from mining sites and compromise a significant part of freshwater resources.

In AMD, aqueous arsenic occurs either as As(III) or As(V) (Williams, 2001). Although in rare cases the precipitation of tooeleite, an As(III)–Fe(III) mineral, has been observed (Morin et al., 2003), the As(III) form, which is believed to be the most toxic, is rather stable in solution, especially at low pH. On the contrary, As(V) presents a strong affinity for Fe(III). In AMD from the former Carnoulès mine (France), the formation of an amorphous As(V)–Fe(III)-oxyhydroxysulfate precipitate has been reported (Leblanc et al., 1996; Morin et al., 2003). More frequently, As(V) forms scorodite (Roussel et al., 1999) or adsorbs on schwertmannite and ferrihydrite as a function of pH (Carlson et al., 2002).

When transported downstream during floods, these minerals may dissolve upon water neutralization and release their As. The dissolution of As may be counteracted by the precipitation of Fe- or Al-oxyhydroxides which efficiently scavenge As(V) (Kimbal et al., 1995). Depending on local site conditions, the As concentration and partitioning between the solid phase and the solution may be further modified along the flowpath by pH and redox condition changes (Fuller and Davis, 1989).

A principal issue for the assessment of As mobility is the elucidation of the forms in which arsenic is transported or immobilized.

The aim of this study was to deepen the understanding of the processes which control the transport and fate of arsenic in mining environments. Therefore, we studied arsenic speciation and partitioning in the Amous river, which receives acid mine drainage from an abandoned Pb–Zn mine in Southern France.

2. Site description

The mine of Carnoulès (Fig. 1a) is located in Southern France in the drainage basin of the Rhône river. The mining activity, stopped in 1962, has left about 1.5 Mt of sulfidic wastes containing 0.7% Pb, 10% Fe and 0.2% As deposited over a segment of the Reigous Creek (Leblanc et al., 1996). The seepage water, which percolates through the wastes, emerges at the bases of the impoundment and forms the spring of the Reigous creek. The water is acidic (pH 3–4.5) and contains up to 250 mg$\cdot$l^{-1} of As in the As(III) form (Casiot et al., 2003). The As concentration decreases within the first 30 m of the creek in relation with the precipitation of amorphous As(V)–Fe(III) and As(III)–Fe(III) gels and tooeleite (Casiot et al., 2003; Morin et al., 2003). At 1.5 km from its source,

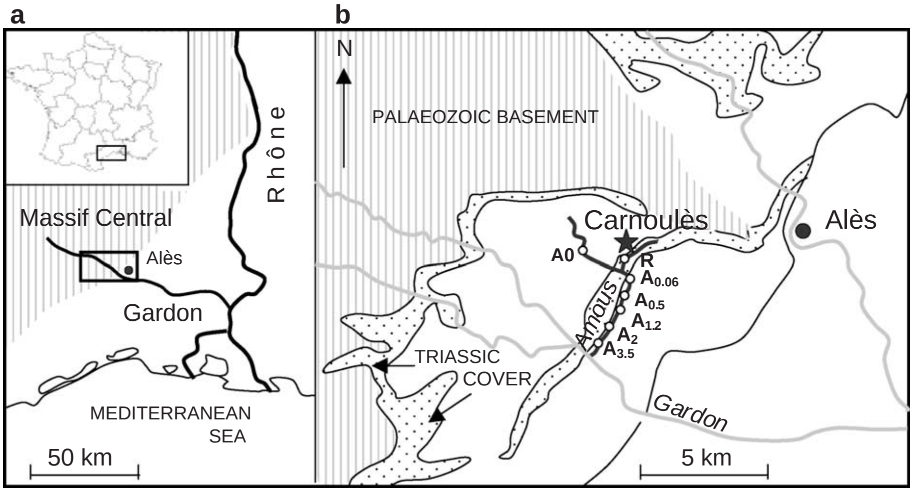

Fig. 1. Site localisation (a) and sampling stations (b). The station in Reigous creek (R) is situated 3 m upstream from the confluence; in Amous river, stations are situated 1.5 km upstream from the confluence (A_0) and 0.06 km ($A_{0.06}$), 0.5 km ($A_{0.5}$), 1.2 km ($A_{1.2}$), 2 km (A_2) and 3.5 km ($A_{3.5}$) downstream from the confluence.