



Research papers

Arsenic and metallic trace elements cycling in the surface water-groundwater-soil continuum down-gradient from a reclaimed mine area: Isotopic imprints



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ABSTRACT

One decade after closure of the Salsigne mine (SW France), As contamination persisted in surface water, groundwater and soil near and down-gradient from the reclaimed ore processing site (OPS). We assess the fate of As and other associated chalcophilic MTEs, and their transport in the surface-water/groundwater/soil continuum down-gradient from the reclaimed OPS, using Sr-isotopic fingerprinting. The Sr-isotope ratio was used as a tracer of transfer processes in this hydro-geosystem and was combined to sequential extraction of soil samples to evaluate the impact of contaminated soil on the underlying phreatic groundwater. The contrast in Sr isotope compositions of the different soil fractions reflects several Sr sources in the soil. In the complex hydro-geosystem around the OPS, the transport of As and MTEs is affected by a succession of factors, such as (1) Existence of a reducing zone in the aquifer below the reclaimed OPS, where groundwater shows relatively high As and MTEs contents, (2) Groundwater discharge into the stream near the reclaimed OPS causing an increase in As and MTE concentrations in surface water; (3) Partial co-precipitation of As with Fe-oxyhydroxides, contributing to some attenuation of As contents in surface water; (4) Infiltration of contaminated stream water into the unconfined aquifer down-gradient from the reclaimed OPS; (5) Accumulation of As and MTEs in soil irrigated with contaminated stream- and groundwater; (6) Release of As and MTEs from labile soil fractions to underlying the groundwater.

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1. Introduction

Tracking the geochemical cycles of metallic trace elements (MTEs) and arsenic in a post mining context is essential for a better orientation of environmental management, remediation strategy and neighborhood protection. Arsenic contamination is a major risk to human health and is a prominent environmental cause of cancer (Smith, 2002). Mining activities involving As ores have an impact on the environment and on human health that may persist for many decades after mine closure (Smedley and Kinniburgh, 2002).

High As concentrations are observed at numerous mining sites (Azcue et al., 1994; Azcue and Nriagu, 1995). In an As-Au mine, the

natural occurrence of As is associated with sulfide minerals, especially arsenopyrite (FeAsS), and realgar (AsS) (Cullen and Reimer, 1989; Matschullat, 2000; Moore et al., 1988; Smedley and Kinniburgh, 2002). In addition to mining effluents derived from water treatment, the oxidation of sulfides, such as FeAsS, is a common process, resulting in the release of As into the total environment (surface water, groundwater, soil and sediment) (Basu and Schreiber, 2013). Even after mine decommissioning, As sources remain in the form of tailings dams, dumps, or As-bearing ore stored on site.

Arsenic is among the most problematic and most studied metalloid elements in the environment because of its mobility over a wide range of redox conditions (Smedley and Kinniburgh, 2002). As (III) is the dominant form under reducing and acidic conditions, while As (V) is the prevalent form under oxidizing conditions. In soil, sediment and water, adsorption and desorption are the main processes controlling As transport and fate (Burton et al., 2009;

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Deschamps et al., 2005; Dousova et al., 2012; Feng et al., 2013; Garcia-Sanchez and Alvarez-Ayuso, 2003; Mukherjee et al., 2008a; Román-Ross et al., 2006; Sørensen et al., 2008). Arsenic adsorbs on Fe- and Mn-oxides, carbonates, clay and organic materials (Bardelli et al., 2011; Bauer and Blodau, 2006; Guan et al., 2009; Postma et al., 2010; Román-Ross et al., 2006; Sørensen et al., 2008; Song et al., 2006; Yokoyama et al., 2012; Zheng et al., 2004). These processes are controlled by the prevailing physical and chemical context, mainly redox conditions, pH, and temperature (Kelderman and Osman, 2007; Mukherjee et al., 2008a, 2008b; Polizzotto et al., 2006; Robertson, 1989; Xie et al., 2009). Under reducing conditions, the increase of dissolved As is often explained by the dissolution of As-bearing Fe-oxyhydroxide and ferric arsenate (Deuel and Swoboda, 1972; Muehe et al., 2013; Welch and Lico, 1998).

The Salsigne mesothermal Au-As mine (southern France) lies in the southern external zones of the French Variscan orogen (Fig. 1). It was considered as the greatest Au-As mine in France and has been exploited for 96 years. Since the start of mining activities, a considerable amount of As has been transmitted to the environment surrounding the mining and ore-processing areas, either in particulate form (dust), or as dissolved fractions (acid mine drainage), exposing the entire local ecosystem to As contamination. After mine closure, extensive remediation actions were carried out at the reclaimed ore processing site (OPS) (Fig. 1). Nevertheless, As concentrations in surface water and groundwater down-gradient from the reclaimed OPS remain significantly higher than that of the regional background (Khaska et al., 2015).

Dissolved arsenic and MTEs such as Cu, Zn, Co, Ni, Sb, Sn, Cd in soil solution may be transported outside the system by downward mass transfer during floodwater infiltration to deeper soil layers, or even into the underlying aquifer, or by upward mass transfer during plant uptake, or during microbial As-methylation processes producing volatile methylarsines (Casentini et al., 2011; Ravenscroft et al., 2009; Smedley and Kinniburgh, 2002). At the same time, MTEs are involved in chemical reactions occurring in the soil solid phase. MTE concentrations in soil solution are governed by interrelated processes, such as inorganic/organic complexation, oxidation/reduction-, precipitation/dissolution- and adsorption/desorption reactions (Kabata-Pendias, 2004). Predicting the concentration of a given dissolved metallic element in soil solution depends on the accuracy of predictions of changes in soil-environmental conditions over time, such as the degradation of organic matter, changes in pH, redox potential, or the chemical composition of soil solution. Speciation may also evolve accordingly.

Sequential-extraction techniques have been used for characterizing the distribution of metallic elements in the different soil and sediment fractions (Gleyzes et al., 2002; Li and Thornton, 2001), for predicting their mobility (e.g. Aubert et al. 2004); and for determining their origin (Campanella et al., 1995; Tessier et al., 1979). A seven-step sequential extraction procedure was used in this paper on a representative soil sample in order to: i) investigate the distribution of MTE and As contents associated with the different soil fractions, and ii) determine the potential impact of the contaminated soil on unconfined groundwater.

Furthermore, such techniques can be combined with isotopic composition determination, e.g. Pb isotopes (Cary et al., 2015; Teutsch et al., 2001), to distinguish between various possible metal sources. Here, we use the Sr isotopic ratio for tracking the fate of As and other associated chalcophilic MTEs, such as Ni, Cd, Co, Cr, Zn, Cu, Sn and Sb, and their transport mode in the surface-water/groundwater/soil continuum, to assess environmental pollution and human exposure in a post mine closure context. $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is widely used in pollution studies to track natural and/or anthropogenic processes (Aquilina et al., 2015; Cary et al., 2014;

McArthur et al., 2001; Négrel and Petelet-Giraud, 2005; Pennisi et al., 2009). In particular, at this study site Khaska et al. (2015) showed that, $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is a powerful tool for distinguishing between sources of Sr and As. In this context, Sr isotopes were used as tracers to differentiate the natural and anthropogenic sources of As and other trace elements released from a mining context where CaO is used in the water treatment process. They evidenced the mobilization of As stored in unconfined aquifer to the stream water during dry season, and the mobilization of As with surface runoff during intense rainy events at the vicinity of the reclaimed OPS.

The present study aims to track the fate of As and MTEs down-gradient of the reclaimed OPS in the surface water/groundwater system, and to determine the influence of As and MTE soil contents on the underlying unconfined aquifer. Furthermore, Sr isotopes were coupled to the geochemical composition of the different fractions of the soil, derived from sequential extraction, in order to determine the As and MTE labile fractions.

The objectives of the present study are to:

1. Determine the fate of dissolved As and other MTEs, at the interface between the reclaimed OPS and surface water, between surface water and the Fe-oxyhydroxide film precipitated on the streambed, and in the surface-water-groundwater-soil continuum;
2. Assess the impact of contaminated soil on water quality of the subjacent unconfined aquifer by means of isotopic study and sequential extractions,
3. Study the distribution of As and MTE in the different phases of the contaminated soil, in order to determine the labile soil fraction able to influence the composition of groundwater.

We first discuss the processes of MTE release in association with As in surface water near the reclaimed OPS. Then we assess the mechanism for As and MTE accumulation in the solid phase precipitated from contaminated surface water. Thereafter, the interaction between surface water and groundwater is discussed, the accumulation of As and MTE in soil, as well as the impact of contaminated soil on the underlying aquifer. Finally, we provide a conceptual model showing the mechanism of As transfer into the continuum of surface, groundwater and soil.

2. Geological setting

The study area is located in the southern Massif Central, SW France. On a local scale, the area lies in the southern part of the Orbiel Valley (Fig. 1), 4 km down-gradient from the reclaimed OPS. The lithology of the unconfined aquifer near the reclaimed OPS is described in Khaska et al. (2015). The aquifer matrix mainly consists of Tertiary formations and Quaternary molasse deposits, with a lithologic facies dominated by fluvial sand, loam, clay and carbonate rocks. The heterogeneous aquifer comprises fine to coarse quartz-rich sand with abundant schist material in the shallow part of the aquifer, interbedded with clayey sand and gravel. Gravel is commonly encountered at 3 m below ground surface in the aquifer, and indurated sand forms discontinuous lenses at shallow depth where past water-table fluctuations occurred (Berger et al., 1993). The thickness of the unconfined aquifer is about 10 m on average.

3. Material and methods

Concentrations of minor- and trace-elements were determined for collected water samples (surface water and groundwater) and on solid samples (soil, sequential extraction of As and MTE in the solid soil phase, and Fe-oxyhydroxide film precipitated in the

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