



The combined effect of abandoned mines and agriculture on groundwater chemistry

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ABSTRACT

Although it is well known that both mining and agriculture disturb groundwater quality, their mutual interactions are much less well documented, though agricultural activities may prevail once mining operations have ended. To study these potential interactions and their impacts on water chemistry, we monitored the chemical composition of groundwater at the outlet of a gold exploration gallery in an area of intensive agricultural activity along with an isotopic study of the groundwater, a reactive artificial tracer test that involved injecting H_2O_2 into the gallery, and geochemical modelling.

The isotopic study revealed denitrification of the NO_3^- -bearing groundwater that takes place through oxidation of the sulphide minerals associated with the gold deposit and leads to anomalous concentrations of some metals such as Zn, Co and Ni. It also contributes to liberating As into the groundwater, where the tracer test confirmed that As is sensitive to the redox conditions. The currently observed high arsenic concentrations in the groundwater are interpreted as resulting mainly from the former mining activities through a remobilization of As sorbed on or co-precipitated with the iron oxides that formed when the gallery was excavated. The geochemical modelling enabled us to calculate the respective role of each process involved in the As accumulation in the groundwater. It is also inferred that NO_3^- contamination from agricultural activities disturbs arsenic remobilization – by consuming available electron donors (e.g. organic matter), NO_3^- limits the reduction of iron oxides and consequently the release of arsenic.

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1. Introduction

Underground mining invariably disturbs the hydrogeological environment primarily due to the fact that the mine has to be dewatered. Then, once the mine has been worked-out and/or abandoned, subsequent flooding contributes to piezometric recovery from the previously developed cone of depression and leads to significant groundwater contamination.

The oxidation of sulphide minerals, such as pyrite, during the dewatering phase will give rise to acid waters that pose a risk to the environment in that they commonly contain

metals and metalloids. The leached metals and metalloids, however, may also be sorbed on or co-precipitated with a solid phase, as is the case with arsenic being adsorbed onto ferric hydroxide (Waychunas et al., 1993), or other adsorbents such as Al- or Mn-oxides (Ladeira and Ciminelli, 2004), or precipitating as scorodite ($FeAsO_4 \cdot 2H_2O$) (Paktunc et al., 2004; Loredó et al., 2005) under acidic and oxidizing conditions. The effect of these reactions on the hydrogeological environment is then delayed until the flooding phase, when dissolution and/or desorption takes place in response to the higher pH and more reducing conditions, so increasing As concentration in the groundwater.

Once mining operations have ended, one can consider various site-remediation options (Jonhson and Hallberg, 2005); these may involve changes in land use, which may

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in turn also affect the environment. Agricultural activities, in particular, introduce nitrate into groundwater through the use of fertilizers and manure; this contributes to the redox control of the groundwater, modifying water–rock reaction paths and consequently the fate of the metals and metalloids already disturbed by the mining activities.

Our aim is to assess the fate of metals and metalloids in groundwater from a case study where intensive agricultural activities were developed concurrently with mining activities that were abandoned in the early 1990s. Its main goal is to highlight the interaction and feedback between the potentially polluting mine-related and agricultural-related anthropogenic activities.

Particular emphasis is placed on arsenic, whose release into groundwater depends on the hydrogeochemical characteristics of the aquifer (e.g. Smedley and Kinniburgh, 2002; Varsányi and Kovács, 2006; Nath et al., 2008) and whose elevated concentration in many groundwater bodies around the world represents a major public health hazard. Arsenic is a well-known toxicant that can cause skin lesions and hypertension. It is also a proven carcinogen that gives rise to cancer of the liver, bladder, kidney, prostate and lungs (Agency for Toxic Substances and Disease Registry, 2000; Jain and Ali, 2000; Clewell, 2005); hence the many studies into the pathway of arsenic metabolism in humans and the impact of cofactors (e.g. Vahter, 2000; Christian et al., 2006).

The present paper focuses on arsenic and metal concentrations in groundwater around an abandoned exploration gallery. The key issues have been to (i) identify the key parameters favouring As and metals leaching from the solid phase to the groundwater, and (ii) distinguish the respective impact of gallery flooding and biogeochemical reactivity between the nitrate and the solid phase (denitrification) on concentrations. We adopted a combined approach using (i) the time series of chemical parameters at the gallery outlet during the pumping/flooding sequence in the gallery, (ii) a geochemical and isotopic characterization of the groundwater within and around the gallery, (iii) a controlled experimental injection of hydrogen peroxide within the gallery, and (iv) geochemical modelling of the mineral dissolution and As desorption.

2. Experimental section

2.1. Study site

The study site is an abandoned exploration gallery investigating gold-bearing arsenopyrite ore near Loperec in north-western France (Fig. 1). The ore, located at the Devonian–Carboniferous transition characterized by acid and basic volcanism associated with epithermal polysulphide mineralization, consists of arsenopyrite-rich quartz veins and Fe–Ca-rich chert hosted by acid volcanic rocks (rhyolite and tuff) intercalated in calc-schist and black schist. The ore is rich in sulphide minerals such as arsenopyrite, pyrite, pyrrhotite, chalcopyrite, sphalerite and galena. The arsenic content of the ore is around 2000 ppm, as against a concentration of around 1000 ppm in the volcanic rocks. In addition to arsenopyrite, As is present in Co-gersdorffite and copper sulphides (tennantite and tetrahedrite). The black schist is also sulphide rich (pyrite and arsenopyrite).

The Loperec deposit was identified from a geochemical survey in 1985 and drilled in 1987. The mineralized bodies were then investigated in 1991 by a gallery at around 50 m depth. A lack of sufficient reserves led to the site being abandoned in 1992. Dewatering was stopped on 30 March 1994 with a drainage pipe being installed to carry the mine water from the gallery outlet to a settling pond.

The hydrogeological configuration, typical of a hard-rock multi-layered aquifer (Wyns et al., 2004), comprises a) unconsolidated weathered rock (saprolite or regolith) with an interstitial porosity and a capacitive role, overlying b) a fissured-weathered layer ensuring the transmissive function of the aquifer, overlying c) fresh basement which is permeable only where deep tectonic fractures are present. The piezometric level reflects the surface topography and thus, following the classical pattern, water will preferentially infiltrate the high-elevation areas (where the piezometric level is seen to be higher in the upper compartment than in the lower) and have its drain several tens to several hundreds of metres farther down. At the study site, however, the piezometric contours are perturbed by the gallery and its drainage pipe.

The region's primary activity is agriculture characterized by intensive livestock (poultry, pigs) and crop (corn and wheat) farming with temporary pastures for dairy production.

2.2. Sample collection

Chemical monitoring of the water at the gallery outlet began in 1991, at the start of the excavation work, in order to check the quality of the waste water. The collected samples were filtered (0.45 µm) and acidified (up to pH 2) before being analysed for their trace- and major-element concentrations.

More recently (May and September 2001) samples were collected for analysis from different points around the gallery (Fig. 1); i.e. a) the gallery outlet equipped with a pipeline reaching the ground (OUT), b) a spring (GUY), c) three exploration drill holes (ART, H2, H4), and d) a farm well (ROL). Two additional samples were collected in November 2001 from a well drilled specifically to attain the gallery (LOP); these were taken with a submersible pump just above and inside the gallery, respectively, once a volume corresponding to three times that of the well had been pumped out. Based on the well equipment and depth, the water from the farm well (ROL) is from the upper aquifer compartment (weathered facies), that of the spring (GUY) and drill hole H4 is from both the weathered and fissured compartments, and that of drill holes H2 and ART is from the deeper (fresh basement) compartment. The gallery was excavated within the fissured and fresh-basement compartments of the aquifer. The water samples were filtered through a 0.45 µm membrane and stored at 4 °C for anion determination, or acidified to pH 2 with ultrapure HNO₃ for cation and trace-element determination. A solid sample was also collected at the gallery outlet. Here, the solid and liquid phases were separated by centrifugation (15 min, 3000 t/min) followed by filtration before freeze drying. The Fe and As contents of the solid phase were determined by ICP-AES after acid digestion.

Additionally, two SIGMA 800 automatic water samplers were installed at the gallery outlet in May 2001 to collect samples at 2–4 day intervals over a one-year period for anion

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