



Trace metal behavior during in-situ iron removal tests in Leuven, Belgium

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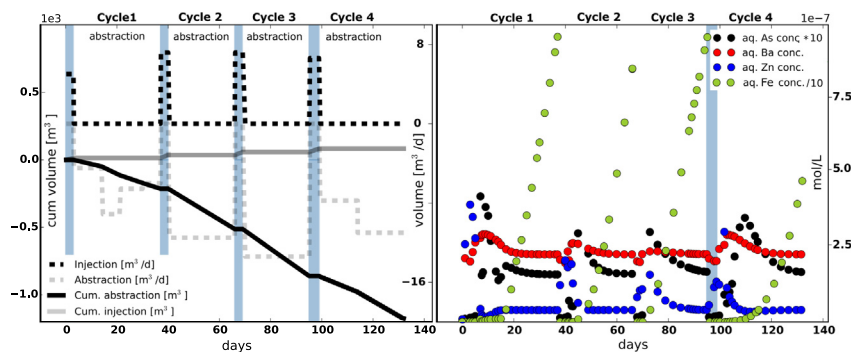
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HIGHLIGHTS

- Arsenic (As) and Barium (Ba) are released during subsurface iron removal operations.
- Sorption through cation exchange retards Ba; complexation lowers total As concentrations.
- As is mobilized despite available sorption surfaces during injection and recovery.
- Study shows high propensity for As mobility during SIR due to desorption.

GRAPHICAL ABSTRACT



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ABSTRACT

Subsurface iron removal (SIR) is an in-situ technique to lower the iron content of extracted groundwater. Through cyclic injection of oxygenated water ferrous iron oxidises and precipitates as iron hydroxide in a zone surrounding the extraction well, enhancing the sorptive capacity of the aquifer. During subsequent pumping phases, groundwater traverses the oxidation zone and ferrous iron sorbs to available and newly formed exchange and sorption sites, thereby retarding the breakthrough of dissolved iron. The process is well-understood in regards to the retardation of iron. Less well understood, however, is the behavior of a number of trace metals and metalloids during SIR operations, foremost arsenic (As). In this study, we analyse major and minor ion and trace metal concentrations from a number of SIR tests in a sand aquifer near Leuven, Belgium. We use reactive transport modelling to evaluate conceptual models of trace metal release and arrest. The test data, underpinned by model results, show that metal release, namely arsenic and barium (Ba), occurs through the oxidation of trace amounts of sulphide minerals during the injection phase. Sorption through cation exchange retards Ba while complexation lowers dissolved As concentrations. Arsenic is mobilized again during the pumping phase through varying phosphate concentrations in the native groundwater, despite available sorption surfaces, while Ba remains adsorbed. Concentrations, however, remain below WHO guideline values for As and Ba (10 µg/l and 0.7 mg/l), respectively. The developed conceptual model of As fate reveals a high propensity for As mobility during SIR due to desorption reactions and delivers an explanation as to why many SIR operations fail to show substantial As removal, despite efficient iron removal. Other monitored trace elements showed no mobilisation, including Zn, Al, Cd, Cr, Cu, F, Hg, Ni, Pb, Sb and Se.

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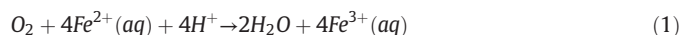
1. Introduction

Subsurface iron removal (SIR) is an effective technique for lowering the ferrous iron (Fe^{2+}) content in groundwater pumped for the production of drinking water or industrial applications (Appelo and de Vet, 2003; Hallberg and Martinell, 1976; Rott and Lamberth, 1993; Van Halem et al., 2011). The technique consists of the periodic injection of oxygenated water into an anoxic aquifer followed by a period of extraction of groundwater with lowered iron concentrations. Subsurface aeration thereby oxidises ferrous iron in-situ by homogenous and heterogeneous oxidation, creating a precipitation zone around the SIR well. Homogeneous oxidation involves the oxidation of dissolved Fe^{2+} to ferric iron (Fe^{3+}) (Eq. (1)) during the injection phase and its precipitation as an iron oxide phase (Eq. (2)). Heterogeneous oxygenation of Fe^{2+} adsorbed on surfaces of iron oxide phases then adds to the built-up of iron oxide precipitates in the aquifer. This provides new sorption sites for dissolved iron, manganese (Mn) and other trace elements such as arsenic (As) or phosphate (PO_4) (e.g. van Halem et al., 2011). During abstraction, ambient groundwater with elevated Fe^{2+} concentrations flows back toward the SIR well traversing the precipitation zone. Adsorption of Fe^{2+} to the available surface sites permits water with reduced iron concentrations to be abstracted. With time, the capacity of available surface sites becomes exhausted and breakthrough of ambient Fe^{2+} concentration occurs at the abstraction well. The efficiency of the SIR scheme is thereby determined by the ratio of the extracted, low-iron groundwater volume to the volume of injected oxygenated water.

The concepts behind SIR are well understood (Appelo et al., 1999; Appelo and de Vet, 2003; van der Laan, 2008). Less well understood, however, is the behavior of a number of trace metals and metalloids during SIR operations, foremost arsenic. Trace metals and metalloids could be shown to be removed in conjunction with iron during SIR operations through adsorption or co-precipitation onto newly formed iron or manganese oxides, e.g. in the case of arsenic (e.g. Rott et al., 2002) or lead (Pb), vanadium (V), nickel (Ni), fluoride (F), strontium (Sr) and zinc (Zn) (e.g. van Halem et al., 2011). In fact, SIR technology has been proposed as a cost-effective option for subsurface arsenic removal (SAR) (van Halem et al., 2010a). However, other SIR schemes reported insufficient adsorption of trace metals or even mobilisation. Van Halem et al. (2010b) reported on an in-situ iron removal trial close to Dhaka, Bangladesh, where aerated water was injected into an aquifer of elevated iron (1 mg/l) and arsenic (145 $\mu\text{g/l}$). Although ferrous iron levels were significantly reduced, only negligible amounts of arsenic were removed from the groundwater. In addition, while the efficiency of iron removal increased with successive cycles (cycles consist of one injection and pumping phase), the same was not observed for arsenic, nor did increased oxygen levels in the injectant lead to increased As retention. The authors speculated that either the contact times with freshly formed iron oxides was not sufficient or competing ions such as phosphate prevented sufficient As sorption. However, Rahman et al. (2015a, 2015b) reported from SAR trials in Bangladesh that, despite low phosphate concentrations, arsenic in the extracted water never dropped below the WHO guideline of 10 $\mu\text{g/l}$. The authors speculated that competitive sorption of silica (SiO_2) and bicarbonate (HCO_3^-) hindered As retention. Appelo and de Vet (2003) noted variable arsenic concentrations during successive cycles of an SIR system in the Netherlands and speculated that As sorbed to colloidal iron oxides could be the cause of observed As peaks. Multicomponent effects were also proposed to influence As concentrations during the SIR test.

In this study, a series of field scale tests in a sand aquifer are used to explore the fate of trace metals and metalloids during subsurface iron removal operations. We provide a data-constrained quantitative analysis of the relative importance of individual geochemical processes, and their interplay with hydraulic conditions, to mechanistically explain observed trace metal behavior at a potential site for commercial scale SIR operations for potable water production in Leuven, Belgium. The chosen

field site is well-characterised and benefits from strictly controlled hydraulic flow conditions that are induced through injection and extraction cycles. Data on abstraction and injection volumes and rates, hydraulic conductivities, mineralogy, ambient water chemistry and detailed temporal data on the water chemistry evolution of the extracted water provide effective constraints for the development of plausible conceptual and numerical models on trace metal behavior under field scale heterogeneity. The coupled flow and reactive transport simulations provide a framework which assists in the analysis of the field data and provides a process-based explanation for the observed trace metal behavior at the site. This quantitative understanding is essential for the sustainable operation of SIR operations and allows the prediction and elimination of potential operational risks (e.g. clogging). At the field site itself, it provides the fundamental basis to support a decision whether or not to implement SIR at the scale of a groundwater extraction for potable water production.



2. Materials and methods

2.1. Field site

The SIR trial site is located near the city of Leuven (Belgium) where the drinking water company De Watergroep operates the 876,000 m^3/year groundwater abstraction plant Kessel-Lo Vlierbeek (Fig. 1). Here, groundwater is withdrawn from the Brussels Sands aquifer using a well battery with screen locations at a depth between 15 and 30 m below surface level. The Brussels sands (Formation of Brussel) is a regionally important aquifer consisting of sediments from the Middle-Eocene. The formation is inclined toward the north and is phreatic at the location of the test site, covered by maximum 7 m of moderate to coarse grained Quaternary sand. The Brussels Sands are bounded at depth by the clay of the Formation of Kortrijk.

Regionally, the Brussels Sands is considered heterogeneous, consisting of quartz sands with variable percentages of feldspar, flint, glauconite and carbonate minerals, often cemented by calcite, silica and/or iron oxides and occasionally sulphides to form sandstone layers and concretions (Laga et al., 2001; Houthuys, 2011; Peeters, 2010; Lagrou et al., 2004). While the predominant mineral component of the Brussels Sands is quartz, followed by calcite, petrographical analysis by Lagrou et al. (2004) confirmed the presence of 0 to 1.7 weight percent (wt%) glauconite and trace amounts of opaque minerals in some samples, which were interpreted as pyrite. XRD analysis by Peeters (2010) identified trace amounts of goethite in ferruginous sandstone in the Brussels Sands.

At the test site, the upper 14 m of the Brussels Sands are composed of brown unconsolidated moderate to coarse sands intercalated with sandstone horizons. The lower 23 m consist of grey median to coarse sands with a 4 m thick very coarse sand layer at the base. The groundwater chemistry in these lower grey sands, which constitute the target aquifer for the SIR trial, has been monitored in the well battery of the Kessel-Lo Vlierbeek abstraction plant since 2007. The ambient water is fresh ($\text{EC} \approx 700 \mu\text{S/cm}$) and, because of the presence of calcite cement of calcium-bicarbonate type (Table 1). Median concentrations of calcium (0.3 mmol/l) and bicarbonate (0.35 mmol/l) indicate equilibrium to slight oversaturation in respect to calcite ($\text{SI}_{\text{calcite}} 0.2$), with an average pH of 7.35. The redox state of the groundwater is reducing with elevated Fe^{2+} and Mn^{2+} concentrations (2–6 mg/l and 0.07–0.13 mg/l, respectively). Oxygen and nitrate concentrations are below detection limit, in accordance with the redox state, while sulphate concentrations range between 120 and 130 mg/l.

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