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In situ treatment of arsenic-contaminated groundwater by air sparging

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ABSTRACT

Arsenic contamination of groundwater is a major problem in some areas of the world, particularly in West Bengal (India) and Bangladesh where it is caused by reducing conditions in the aquifer. In situ treatment, if it can be proven as operationally feasible, has the potential to capture some advantages over other treatment methods by being fairly simple, not using chemicals, and not necessitating disposal of arsenic-rich wastes. In this study, the potential for in situ treatment by injection of compressed air directly into the aquifer (i.e. air sparging) is assessed.

An experimental apparatus was constructed to simulate conditions of arsenic-rich groundwater under anaerobic conditions, and in situ treatment by air sparging was employed. Arsenic (up to 200 μg/L) was removed to a maximum of 79% (at a local point in the apparatus) using a solution with dissolved iron and arsenic only. A static "jar" test revealed arsenic removal by co-precipitation with iron at a molar ratio of approximately 2 (iron/arsenic). This is encouraging since groundwater with relatively high amounts of dissolved iron (as compared to arsenic) therefore has a large theoretical treatment capacity for arsenic.

Iron oxidation was significantly retarded at pH values below neutral. In terms of operation, analysis of experimental results shows that periodic air sparging may be feasible.

1. Introduction

Arsenic, widely acknowledged as biologically harmful, is a contaminant in groundwater in many areas of the world, including Cambodia, Argentina, Chile, China, Hungary, Laos, Mexico, Mongolia, Nepal, Pakistan, Taiwan, Thailand, Vietnam, and the USA (Ahmed, 2003). However, the most widespread and serious groundwater arsenic levels are evident in West Bengal (India) and Bangladesh. The Bangladeshi arsenic problem has been described as "the largest poisoning of a population in history, with millions of people exposed" (Smith et al., 2000, pg. 1093).

The current World Health Organization guideline for inorganic arsenic in drinking water is 10 μg/L (WHO, 2008). However, not all jurisdictions follow this guideline, including Bangladesh and India, where a guideline of 50 μg/L is used (Chakraborti et al., 2009; Flanagan et al., 2012; WQAA Government of India). Chronic arsenic exposure may result in severe health effects with skin lesions, hyperkeratosis, and increased risk of cancers.

Although anthropogenic sources of arsenic exist (e.g. smelting operations), the most widespread problems are of natural geochemical origin. Groundwater arsenic concentrations reported in the literature range from <0.5 μg/L to 5000 μg/L under natural conditions (Smedley and Kinniburgh, 2002). Oxides of iron, aluminum, and manganese are likely the most important sources and sinks for arsenic in aquifer sediments (Stollenwerk, 2003).

Arsenic may be mobilized from soil as a result of reducing conditions in groundwater, as occurs in West Bengal and Bangladesh. The reducing conditions are the result of oxidation

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Abbreviations: DI, deionized; DO, dissolved oxygen; HFO, hydrous ferric oxide; HRT, hydraulic retention time; ORP, oxidation–reduction potential; RmV, relative milli-volts.

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of buried organic matter in sediments. This causes mobilization of arsenic by reduction of iron oxyhydroxides, reductive dissolution, and change in structure of iron oxide minerals (BGS and DPHE, 2001; Nickson et al., 1998; Smedley and Kinniburgh, 2002).

Treatment of arsenic-impacted groundwater is feasible, and many options exist including oxidation and sedimentation, coagulation and filtration, sorption techniques, and membrane techniques. While being capable of effectively removing arsenic, many treatment technologies have drawbacks due to requirements for use of chemicals, disposal of arsenic-rich wastes, and/or technological complexity.

As an alternative option, in situ treatment (by creation of oxidizing conditions) is a fairly simple procedure, does not require chemicals, and does not require disposal of arsenic-rich wastes. The basis for in situ treatment of arsenic is the same as that for in situ treatment of iron and manganese, which has been utilized in Europe for decades. Oxidation–reduction potential (ORP) conditions can be changed in the subsurface by introducing dissolved oxygen (DO), causing oxidation of ferrous iron and other metals (in solution and on the soil grains). This process creates ferric iron oxyhydroxides capable of adsorbing ferrous iron and other oxyanions such as arsenic (Appelo and de Vet, 2003; Rott, 1985; Rott and Lamberth, 1993; van Beek, 1985; van Halem et al., 2010a). These oxidation processes are enhanced by autocatalytic effects from oxidation products (Rott and Friedle, 2000; Sung and Morgan, 1980; Tamura et al., 1980).

One method of in situ treatment for arsenic in groundwater involves injection of aerated water into the aquifer. This method has met with moderate success to date (Rott and Friedle, 1999, 2000; Sen Gupta et al., 2009; van Halem et al., 2010a, 2010b, 2010c). One possible concern regarding in situ treatment is the possibility that pore spaces in the aquifer may become clogged. However, this is not a significant problem in reality. Subsurface treatment for iron in groundwater (by method of injection of aerated water) has been used in Europe, and clogging has not been found to be an issue, even after more than a decade of operation (van Halem et al., 2011). Iron may initially precipitate as hydrous ferric oxide (HFO, of low crystallinity), but in the subsurface it ages and changes to thermodynamically more stable and less voluminous crystallized forms such as goethite (Mettler, 2002; Rott and Friedle, 2000; Smedley and Kinniburgh, 2002; Stollenwerk, 2003), and this prevents clogging.

Another option for in situ treatment of arsenic in groundwater is direct air sparging in the saturated zone. However, besides literature regarding treatment on a deep well (Miller, 2006, 2008) as well as clean-up from a lead smelter site (Miller et al., 2002), there appears to have been limited research on this option. Grombach (1985) suggests that introduction of air directly into the aquifer is the easiest method but did not undertake experimental observations.

The investigation described herein is a lab-scale study to investigate the potential of in situ treatment of arsenic-contaminated water by air sparging. As described below, the experiments utilized a simple solution of dissolved inorganic arsenic and dissolved ferrous iron in order to demonstrate the concept, potential, and key factors of this type of treatment, as a precursor to possible field trials.

2. Materials and methods

The experimental apparatus is illustrated in Fig. 1. Dimensions and sampling port labels are illustrated in Fig. 2.

A large, sealable, food-grade polyethylene barrel (Fig. 1) was used for the inlet solution, which was subjected to nitrogen sparging to remove dissolved oxygen. The main apparatus was made of plexiglass. A small precision-flow peristaltic pump (Fisher model CON3386) was used to transport solution from the reservoir barrel to the inlet column. Rotameters of appropriate size were used for measuring gas flow.

The apparatus had both an inlet and an outlet reservoir, fitted with a fine stainless steel mesh to retain the sand medium. A uniformly graded (rounded, nominal size 0.40 mm) sand was rinsed with deionized (DI) water and used as aquifer soil for purposes of the laboratory simulation. In chemical composition, the sand was as follows: SiO_2 >99.5%; TiO_2 ~0.10%; K_2O ~0.10%; CaO ~0.03%; Fe_2O_3 ~0.03%; Al_2O_3 ~0.01%; Loss on ignition ~0.12%

The outlet height was used to adjust the hydraulic gradient. Air sparging was accomplished using an aquarium aeration pump (Hagen, model Maxima-R) with tubing attached to a rotameter. The outlet from the rotameter ran to a small (2.5 cm) alumina diffuser stone (Fisherbrand model ME46944C) at the sparging point in the apparatus, as shown in Fig. 1.

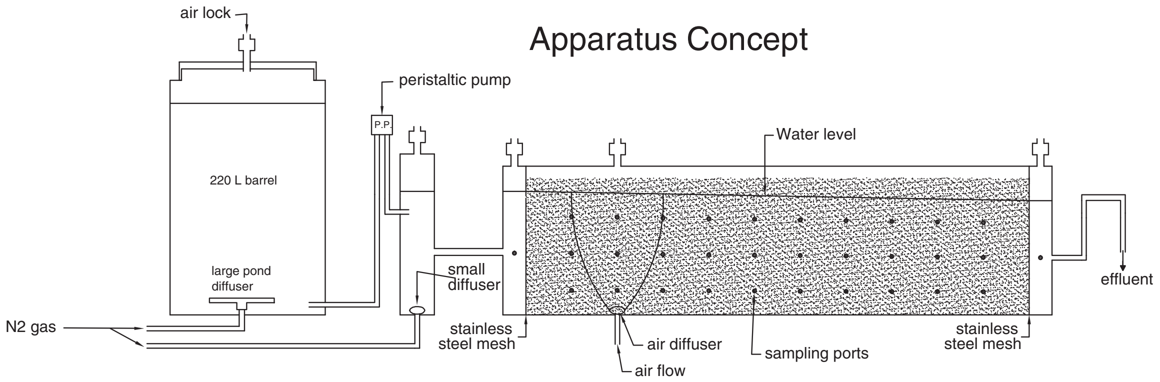

Fig. 1. Apparatus conceptual design.