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Implementation of zero-valent iron (ZVI) into drinking water supply – Role of the ZVI and biological processes

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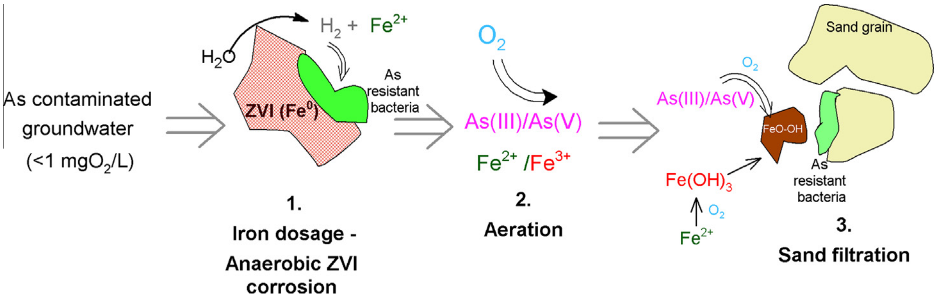
HIGHLIGHTS

- A new ZVI-based treatment for arsenic removal was monitored for period of 45 m.
- The 6 m period, a lag phase, was found to be necessary to achieve maximum As removal.
- The lag phase is linked with biological processes in ZVI and sand media.
- Bacterial found at each stage of treatment have high As resistance.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic in drinking water is concerning millions of people around the world, even though many solutions to the problem have come up in recent years. One of the promising solutions for removing arsenic from water is by implementation of a zero-valent iron (ZVI) in the drinking water production. The purpose of this work was to study a treatment of As pollution based on the ZVI, aeration and sand filtration that was monitored for period of 45 months. In applied configuration and conditions ZVI was not able to remove arsenic alone, but it worked as a source of ferrous ions that during its oxidation enabled to co-precipitate arsenic compounds in the sand filter. The results show that after a lag phase of about 6 months, it was possible to achieve water production with an As content from 20 μg L^{-1} to below 5 μg L^{-1}. The treatment also enabled to remove phosphates that were present in groundwater and affected As uptake by hindering its co-precipitation with Fe compounds. Determination of colony forming units on As amended agar helped to find arsenic resistant bacteria at each stage of treatment and also in the sand filter backwash sludge. Bacterial communities found in groundwater, containing low concentration of As, were found to have high As resistance. The results also indicate that the lag phase might have been also needed to initiate Fe ions release by corrosion from elemental Fe by help of microbial activity.

1. Introduction

Many solutions for arsenic problem in drinking water, which is concerning millions of people around the world, have come up in the recent years. It is still a major challenge in the developing countries, where a sustainable As remediation technology is needed (Sarkar et al., 2012). One of the promising solutions has appeared with introduction of a zero-valent iron (ZVI) that can be implemented by making usage of simple materials available on the local market, i.e. iron nails from industry.

The role of the ZVI, or rather corrosion products that are produced on a water/ZVI surface, can be dual; it directly adsorbs arsenic species on the corroded surface, or it is used for iron compounds supplement if the raw water content is low. Efficient

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adsorption of As from aqueous solutions has been found with synthesized nanosized particle zero-valent iron (nZVI) (Rahmani et al., 2010). Application of the nZVI might be difficult in the water treatment, especially in rural areas, due to costs and limited availability of the material. Additionally, the application of nZVI in the public water systems may be limited due to its tiny particle size (Zhu et al., 2009). Hence, more advantageous is the use of bulk ZVI, where the main mechanism of arsenic removal is based on sorption to partially oxidized iron compounds, such like magnetite or green rust, which result from corrosion of metallic iron (Khan et al., 2000; Manning et al., 2002).

Depending on the treatment design the ZVI can be used at aerobic or anaerobic conditions, resulting in different removal mechanisms. In case of oxygen rich water the ZVI reacts with dissolved oxygen (DO) and forms intermediates that not only adsorb arsenic species, but also helps in oxidizing As(III) to As(V), but the mechanisms were the subject of a wide discussion (Katsoyiannis et al., 2008, 2009; Pang et al., 2009; Noubactep et al., 2012a). The lack of oxygen, that is used as an electron acceptor in the corrosion process, can be substituted by As(V) compounds. Arsenates presence was reported to generate a Fe(III) compounds from iron fillings resulting in removal of arsenic compounds by adsorption on iron oxides adsorption sites (Melitas et al., 2002). However, in case of naturally contaminated groundwater low level of DO and reductive conditions cause that arsenic occur in form of As(III) and abstracted water is hardly performing chemical ZVI oxidation due to lack of electron acceptors. Contrary a possible reduction of As(III) to As(0) could happen, but with much lower removal rate compared to oxygenated conditions (Kanel et al., 2005; Klas and Kirk, 2013). In such a case some lessons might be learnt from studies on permeative reactive barriers containing ZVI. Microbial activity, determined as an increase in bacteria counts around the permeable reactive barrier, where ZVI promotes anaerobic microbial growth in the surrounding area by an increasing pH, decreasing redox potential, production of dihydrogen gas, and the releasing ferrous iron result in the finding of several bacteria that could utilize H_2 produced during anaerobic ZVI corrosion (Zhang, 2003; Da Silva et al., 2007). Therefore, the role of bacteria may be dual, they can help in releasing the iron cations and enhance the processes of arsenite oxidation that was found to be performed by many strains of As(III)-oxidizing bacteria (AOB). However, studies in their performance were limited to laboratory and pilot scale treatments (Katsoyiannis et al., 2002; Katsoyiannis and Zouboulis, 2004; Ike et al., 2008; Gottinger et al., 2010; Liao et al., 2011a). Hence, most of the studies on applied techniques for arsenic removal based on ZVI neglect microbial activity solely focusing on the mechanism for the removal of As by sorption to iron (hydr)oxides, such as magnetite or green rust, which results from corrosion of ZVI and cover iron surface (Manning et al., 2002; Melitas et al., 2002; Klas and Kirk, 2013). Although based on the studies on the ZVI a number of point-of-use methods have been applied around the world for arsenic removal thanks to their simplicity and the minimum maintenance required, they still face issues like disposal of As-bearing sludge (Meng et al., 2001; Ngai et al., 2007; Giles et al., 2011; Cornejo et al., 2012). The above mentioned and other issues might be overcome by up-scaling the point-of-use treatment to fully treatment plants, where better process optimization and organization may be applied. The subject of this study is technology needs, i.e. improving reliability of small drinking water systems, in places where the arsenic problem is a serious threat, like rural areas in the Bengal Delta region.

The purpose of this work was to present and describe a treatment based on the zero-valent iron, aeration and sand filtration. Regarding the limited number of studies about long term implementation of the ZVI based treatment it can help to obtain knowledge of a steady state treatment. Additionally, an objective was to find a role of microbial activity in the treatment that might bring an understanding of how the methods might contribute to developing a technology for small scale rural waterworks that can bring a real change in developing countries. The treatment has been put into operation in a small waterworks treating naturally contaminated groundwater in Denmark, and had been monitored for period of 45 months.

2. Material and methods

2.1. Description of the treatment plant

The water treatment plant is placed in the eastern part of Jutland, Denmark. The quality of its raw water is presented in Table 1, where the fluctuations of compound concentrations resulting from changes in the period of the monitoring are taken into account. The speciation of As and Fe species reveal that they were in forms of dissolved As(III) and Fe(II).

The As removal treatment plant was based on the patented MicroDrop Aqua (MDA) system containing the ZVI source, followed by an aeration unit and a sand filter (Sønderby et al., 2008; Lebech, 2011). The flow sheet of the treatment process with the sampling taps is presented in Fig. 1. The core of the system are two parallel iron capsules (IC) with ZVI. A design of the treatment, especially a reactive zone (RZ) containing ZVI, described according to paradigm proposed by Noubactep et al. (2012b) is following:

- the ZVI was in the form of diverse spiral coils with shape of length between 3 and 30 mm and porosity 0.86 ± 0.02 and a bulk density of 0.95 ± 0.05 kg L^{-1};
- the dimensions of the RZ, that contains only ZVI placed in a cylindrical chamber, are height (H_{ZVI}) of 1.2 m and diameter (D) of 0.31 m that results in a IC's volume of 90 L each.

After the groundwater pass through the IC's, with pump 1 (P1) with flow 3 $m^3\,h^{-1}$, it is aerated in the aeration unit (MDA aerator). The aerator contains open grid plastic tubes that like a trickling filter that can increase the water–air contact interface. From the tank placed below the aeration unit, the aerated water is collected and pumped, with pump 2 (P2) with flow 1 $m^3\,h^{-1}$, to a pressurized sand filter, a inert zone (IZ). The sand filter consist of cylindrical container containing quarts sand (0.2–1 mm) with sand bed height (H_{sand}) of 1.4 m and internal diameter of (D_{sand}) 0.65 m. The samples from the waterworks were collected in a period from September 2010 to December 2013, but the system in the presented form was implemented in June 2010. The plant produces about 10000 m^3 per annum, with a flow of 1 $m^3\,h^{-1}$ and an estimated energy consumption of 0.7 kW h m^{-3}.

Table 1
Main features of the raw waters (errors comes from differences in measurements from the period of monitoring at the waterworks).

Parameter	Reading/concentration
pH	7.1–7.6
O_2 (mg L^{-1})	1.5 ± 0.5
Conductivity (μS cm^{-1})	380 ± 10
Total Fe (mg L^{-1})	0.3 ± 0.2
Total As (μg L^{-1})	19 ± 2
Ca (mg L^{-1})	65 ± 5
Mg (mg L^{-1})	5.0 ± 0.5
SO_4^{2-} (mg L^{-1})	18
Cl^- (mg L^{-1})	18 ± 1
NO_3^- (mg L^{-1})	2.00 ± 0.05
PO_4^{3-} (μg L^{-1})	29 ± 4