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The distribution, solid-phase speciation, and desorption/dissolution of As in waste iron-based drinking water treatment residuals

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Abstract

Arsenic concentrations and solid-phase speciation were assessed as a function of depth through Fe-media beds for two commercially available products (Granular Ferric Hydroxide®—GFH and Bayoxide E33®—E33) from pilot-scale water treatment field tests. These results were compared with data from solution (de-ionized water—DI-H_2O) concentrations of As equilibrated with Fe-media in an anoxic environment at 4 °C. The materials had a high capacity for As (GFH media 9620 mg kg^{-1} As, E33 Media 5246 mg kg^{-1}). Arsenic concentrations decreased with bed depth. For E33, X-ray absorption near-edge spectroscopy results showed that As(V) was the dominant solid-phase species. For GFH, As(III) was detected and the proportion (relative to As(V)) of As(III) increased with bed depth. Arsenic concentrations in DI-H_2O equilibrated with the media were low ($\leqslant 35\ \mu g\ l^{-1}$) over a period of 50 d. Arsenic concentrations in the equilibrated solutions also decreased with depth. Results from tests on soluble As speciation show that As in solution is in the form of As(V). Kinetic desorption experiments carried out at different pH values (3, 5, 7, 8, and 9) show that the media exhibit some acid/base neutralization capacity and tend to bind As sufficiently. Concentrations of As in the pH desorption experiments were in the same order of magnitude as the toxicity characteristic leaching procedure extractions (tens of $\mu g\ l^{-1}$) except at low pH values. For the GFH media tested at a pH of three, As increases in solution and is mainly associated with colloidal (operationally defined as between 0.1 and 1.0 μm) iron.
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1. Introduction

The new drinking water maximum contaminant level (MCL) for As ($10\ \mu g\ l^{-1}$) was established in January 2006 in the US. Although not considered as a best available technology due to insufficient data at the time of the As MCL final rule, iron-based sorption media are excellent candidates for the removal of As (especially As(V)) from source waters.

There are many studies on the sorption of As by Fe compounds including goethite (Manning et al., 1998), ferrihydrite (Jain et al., 1999), zero valent iron (Su and Puls, 2001), and hematite (Redman et al., 2002). Iron oxides sorb As(V) more effectively than As(III) at low to neutral pH values (e.g. pH 4–7). This difference in sorption capacity for As(V) and As(III) tends to decrease with increasing pH. The ability of Fe-oxides to sorb As depends on several factors including pH, oxidation/reduction potential, microbial effects, oxide type, and competition for binding sites from other anions. Groundwater sources of drinking water in low redox potential environments will tend to have higher levels of As(III) relative to surface waters, which tend to be dominated by As(V) (if present). To achieve maximum removal efficiency with iron-based media, As(III) should be oxidized to As(V). The oxidation reaction can occur by contact with manganese oxides (Driehaus et al., 1995). The media can then be regenerated by flushing

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with solutions such as caustic soda (USEPA, 2001a) and reused. Depending on results from the toxicity characteristic leaching procedure (TCLP-EPA Method 1311 (USEPA, 1998)), the material may also be disposed in a hazardous waste (40CFR268 Subtitle C) or non-hazardous waste (Subtitle D) landfill. Land disposal will also depend on state and local regulations.

Few studies have focused on the desorption of As from solids. These studies tend to be concentrated in areas involving a high degree of heterogeneity such as mine wastes (Mok et al., 1988), soils (Elkhatib et al., 1984), and sediments (Ahmann et al., 1997). Jing et al. (2005) examined bonding and leaching of As(V) in water treatment residuals and concluded that As(V) was tightly bound by inner-sphere bidentate complexes. They also presented results from models and batch desorption studies that show higher As desorption at higher pH.

The results presented here include data on As desorption/dissolution with As(III) present in the solids. Bench-scale desorption/dissolution studies focused on the use of continuous-stir reactors and provide evidence for the important role that the stabilization of Fe plays in As mobilization from these media. The results presented here are from materials that underwent As saturation in a field pilot plant setting. This is important because additional constituents (usually excluded in laboratory studies) are also in the influent source water and can play a role in the sorption and desorption of As.

The TCLP has been critically examined for many contaminants, especially As. The TCLP is the test by which a particular spent water treatment media is deemed hazardous or non-hazardous. This classification plays a critical role in the ultimate cost of a particular media-based treatment technology as disposal in a hazardous waste landfill may be 5–10 times more expensive than disposal in a municipal solid waste (MSW) landfill. Approximately 4000 water treatment systems in the US (including community water systems and non-transient non-community water systems) serving 25 people or more will need to implement some sort of treatment strategy for As (USEPA, 2001b). The vast majority of these systems will be small systems serving 10000 people or less. If several simplifying assumptions are made (g residuals l^{-1} H_2O treated = 0.003 (USEPA, 2001a), 4000 utilities, flow = 2.45 million $l\,d^{-1}$ corresponding to a system serving 5000, all utilities use Fe-based media for treatment and all dispose of spent media), annual production of spent media is approximately 10000 t. The total amount of waste placed in US MSW landfills for 2001 was approximately 116120 t (out of a total of 207745 t of MSW) (USEPA, 2003). Thus, 10000 t would represent 0.009% of all US MSW landfilled annually.

To further put this in perspective, compare the estimated volume of spent treatment media with another As-bearing waste; chromate–copper–arsenate (CCA) treated lumber. It has been predicted that, in the US, by the year 2020, 1.6×10^7 m^3 of CCA treated lumber will be taken out of service (and presumably placed in landfills) (Cooper, 1991). If we assume a bulk density of 1.3 $g\,cm^{-3}$ for spent water treatment media, 10000 t of waste equates to 7692 m^3. Thus, the volume of Fe-media waste potentially occupying landfill space would be four orders of magnitude less than the volume occupied by CCA treated wood.

Hazardous waste landfills can cost 5–10 times more for disposal alone, and the costs can escalate enormously depending on proximity and access to hazardous waste landfills. The classification of spent treatment media as hazardous would place an even larger financial burden on small systems that can ill-afford it. Thus, it is essential for us to accurately characterize risks from arsenic desorption/dissolution from these media.

Though the TCLP has been criticized for not characterizing As leachability accurately (Hooper et al., 1998; Ghosh et al., 2004), the fact remains that research to date on As desorption/dissolution in complex environments gives us little information for predicting the short, medium and long-term fate of As bearing wastes in landfill environments. It will be critical in the near future to improve our understanding of the speciation, bonding mechanisms and biogeochemistry of As in landfill environments in order to conclude that one particular extraction procedure is better than another for risk assessment/management purposes.

The objectives of this work were to (1) assess the total As and As speciation on the solid phase as a function of treatment bed depth; (2) examine the desorption (total and species) of As from the media in static leaching experiments; (3) provide data on the kinetics of As desorption from the media; (4) compare the desorption (total, species, and colloidal) of As from the media in different pH environments. Microbial effects were minimized for the static experiments by keeping the samples at 4 °C.

2. Materials and methods

Two Fe-based media were obtained from a pilot-scale water treatment plant. Granular Ferric Hydroxide® (GFH) is manufactured by US Filter with the iron phase made up of ferric hydroxide ($Fe(OH)_3$) and akaganeite (β-FeOOH). The Bayoxide E33® (E33) media manufactured by Bayer for Severn Trent contains approximately 90% FeOOH (presumably goethite from X-ray diffraction studies). Details on the materials, their properties, and intended use may be found at www.usfilter.com (GFH) and www.severntrentservices.com (E33). The emphasis in this research was not on comparing the water treatment capabilities of the media. The focus was on examining the solid-phase speciation of As and the desorption/dissolution of As from GFH and E33. The media were sampled, after As breakthrough occurred ($[As]_{source\ water} = 60.0$ $\mu g\,l^{-1}$), as a function of depth in each of the bed reactors (0–3 cm, 3–10 cm, 10–18 cm, and 18–25 cm). The columns used in the field studies were designed to allow As breakthrough on the order of weeks to months for testing purposes. Full-scale system tests would have required multiple months to years for As breakthrough, which

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