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# Efficacy of acetate-amended biostimulation for uranium sequestration: Combined analysis of sediment/groundwater geochemistry and bacterial community structure



Jie Xu <sup>a, b, d, \*</sup>, Harish Veeramani <sup>e</sup>, Nikolla P. Qafoku <sup>f</sup>, Gargi Singh <sup>b, c</sup>,  
 Maria V. Riquelme <sup>b, c</sup>, Amy Pruden <sup>b, c</sup>, Ravi K. Kukkadapu <sup>g</sup>, Brandy N. Gartman <sup>f</sup>,  
 Michael F. Hochella Jr. <sup>a, b, f, \*\*</sup>

<sup>a</sup> Department of Geosciences, Virginia Tech, Blacksburg, VA 24061, USA

<sup>b</sup> The Virginia Tech National Center for Earth and Environmental Nanotechnology Infrastructure, Virginia Tech, Blacksburg, VA 24061, USA

<sup>c</sup> Department of Civil and Environmental Engineering, Virginia Tech, Blacksburg, VA 24061, USA

<sup>d</sup> Department of Geological Sciences, University of Texas at El Paso, El Paso, TX 79968, USA

<sup>e</sup> Infrastructure and Environment, School of Engineering, University of Glasgow, Glasgow, G12 8QQ, Scotland, United Kingdom

<sup>f</sup> Geosciences Group, Earth Systems Science Division, Pacific Northwest National Laboratory, 902 Battelle Blvd., Richland, WA 99354, USA

<sup>g</sup> Environmental Molecular Science Laboratory, Pacific Northwest National Laboratory, 902 Battelle Blvd., Richland, WA 99354, USA

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## ABSTRACT

Systematic flow-through column experiments were conducted using sediments and ground water collected from different subsurface localities at the U.S. Department of Energy's Integrated Field Research Challenge site in Rifle, Colorado. The principal purpose of this study is to gain a better understanding of the interactive effects of groundwater geochemistry, sediment mineralogy, and indigenous bacterial community structures on the efficacy of uranium removal from the groundwater with/without acetate amendment. Overall, we find that the subtle variations in the sediments' mineralogy, redox conditions, as well as contents of metal(loid) co-contaminants showed a pronounced effect on the associated bacterial population and composition, which mainly determines the system's performance with respect to uranium removal. Positive relationship was identified between the abundance of dissimilatory sulfate-reduction genes (i.e., *drsA*), markers of sulfate-reducing bacteria, and the sediments' propensity to sequester aqueous uranium. In contrast, no obvious connections were observed between the abundance of common iron-reducing bacteria, e.g., *Geobacter* spp., and the sediments' ability to sequester uranium. In the sediments with low bacterial biomass and the absence of sulfate-reducing conditions, abiotic adsorption onto mineral surfaces such as phyllosilicates likely played a relatively major role in the attenuation of aqueous uranium; however, in these scenarios, acetate amendment induced detectable rebounds in the effluent uranium concentrations. The results of this study suggest that immobilization of uranium can be achieved under predominantly sulfate-reducing conditions, and provide insight into the integrated roles of various biogeochemical components in long-term uranium sequestration.

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## 1. Introduction

Uranium bioremediation typically involves the injection of

simple organic substrates such as ethanol or acetate into the subsurface to stimulate indigenous microbial communities that are capable of reducing and immobilizing soluble U(VI) species (Li et al., 2009; Campbell et al., 2012; Bargar et al., 2013; Qafoku et al., 2014). During U(VI) bioremediation, a wide range of organic/inorganic compounds including their intermediate species are competitively utilized as electron donors or acceptors by physiologically diverse groups of microorganisms (Suzuki et al., 2005; Xu et al., 2010; Handley et al., 2012) and via abiotic

\* Corresponding author. Department of Geological Sciences, University of Texas at El Paso, El Paso, TX 79968, USA.

\*\* Corresponding author. Department of Geosciences, Virginia Tech, Blacksburg, VA 24061, USA.

E-mail addresses: [jxu2@utep.edu](mailto:jxu2@utep.edu) (J. Xu), [hochella@vt.edu](mailto:hochella@vt.edu) (M.F. Hochella).

chemical reactions (Jeon et al., 2005; Chakraborty et al., 2010; Hyun et al., 2012). Thus, the total immobilization of uranium is driven by complex series of biological and abiotic pathways.

A variety of microorganisms capable of enzymatically reducing U(VI) have been reported in the laboratory and field studies. These organisms include sulfate/sulfur-reducers, e.g., *Desulfovibrio* spp., *Desulfotomaculum* spp., and *Shewanella* spp., iron-reducers, e.g., *Geobacter* spp., denitrifiers, e.g., *Pseudomonas* spp., and spore-forming species, e.g., *Clostridium* spp., *Cellulomonas* spp., and *Anaeromyxobacter* spp. (Lovley and Philips, 1992; Francis et al., 1994; Abdelouas et al., 1998; Ganesh et al., 1999; Pietzsch et al., 1999; Chang et al., 2001; Suzuki et al., 2003; Beyenal et al., 2004; Wall and Krumholz, 2006; Sharp et al., 2009; Della Vecchia et al., 2010; Handley et al., 2013; Cologgi et al., 2014). The enzymatic reduction of U(VI) by mixed microbial communities can be strongly affected by the presence of competing electron acceptors, however. For example, it had been previously shown that microbial U(VI) reduction was suppressed or even entirely inhibited when nitrate, Fe(III) oxyhydroxides and Fe(III)-containing clays also existed in the environment (Wielinga et al., 2000; Senko et al., 2002; North et al., 2004; Sani et al., 2004; Komlos et al., 2008). Microorganisms can also mediate the reduction of U(VI) indirectly via biogenic solids and soluble species. Several laboratory studies have demonstrated the reduction of U(VI) to nanoparticulate uraninite by sulfide and iron sulfide minerals (i.e., mackinawite) produced during bacterial sulfate reduction (Veeramani et al., 2011, 2013; Hyun et al., 2012). In addition, former work on the interactions of U(VI) with iron-reducing bacteria (i.e., *Geobacter*) indicated that the biogenic Fe(II) could catalyze reduction of U(VI) when both species were adsorbed on clays or (oxyhydr)oxides (Liger et al., 1999; Jeon et al., 2005; Nico et al., 2009; Regenspurg et al., 2009). Overall, the minerals that have been reported to abiotically reduce U(VI) include framboidal pyrite (Qafoku et al., 2009; Descostes et al., 2010; Yang et al., 2014), magnetite (Missana et al., 2003; Scott et al., 2005), green rust (O'Loughlin et al., 2003), zero-valent iron (Farrell et al., 1999), and Fe(II)-bearing clays (Chakraborty et al., 2010).

Uranium can also be attenuated via a range of other processes and interactions with minerals, including surface adsorption (Arnold et al., 1998; Yusan and Akyil, 2008; Bachmaf and Merkel, 2011; Sun et al., 2014), complexation (Kelly et al., 2008; Singer et al., 2009; Stylo et al., 2013; Long et al., 2015), and co-precipitation (Bruno et al., 1995). A range of clays and oxyhydroxides including kaolinite, smectite, montmorillonite, illite, gibbsite, and goethite, were shown to be capable of immobilizing aqueous U(VI) via surface adsorption, and the actual amount depends on the specific mineral surface structure (i.e., aluminol vs. silanol sites), the solution pH, and the co-existence of trace elements (Arnold et al., 1998; Cheng et al., 2004; Chang et al., 2006; Bachmaf and Merkel, 2011). In treated mine tailings from an active uranium mill at Key Lake, Saskatchewan, Canada, uranium sequestration occurs primarily due to sorption within a complex suite of primary and secondary clay minerals, as well as the precipitation of uranium-bearing carbonate and phosphate nanoparticles (Schindler et al., 2015).

In biostimulated aquifers, the overall efficiency of uranium sequestration is a collective result of all the interactions between groundwater, sediments, and microbial populations that may have positive (i.e., adsorption, reduction and precipitation) or negative (i.e., complexation and mass transport facilitated by groundwater flow) effects on soluble uranium species. More, changes in groundwater pH and compositions have direct control over the stability and thus, mobility of the colloidal forms of uranium in aquifers. As intricate feedback systems also exist among the different biogeochemical components, a comprehensive

understanding of the immobilization/remobilization mechanisms of uranium during bioremediation remains elusive and even less is known regarding the interactive relationships between sediment mineralogy, bacterial population structure, and uranium sequestration kinetics.

To investigate the interactions of various biogeochemical components involved in uranium bioremediation processes, we have carried out systematic, controlled column experiments using field sediments that were collected from different uranium-contaminated and non-contaminated aquifers in Rifle, Colorado (USA). These sediments exhibited subtle variations in their mineralogy and geochemical compositions and presented different redox and ecological conditions. By comparison of the behavior of the various sediment systems in the presence and absence of a biostimulant (acetate), we have furthered our understanding by answering the following overarching questions: (1) what is the relationship between the sediment geochemistry and the original bacterial community composition and what are their combined effects on the removal efficiency of aqueous uranium?; (2) does biostimulation with acetate drive detectable changes in the structure and diversity of bacterial communities in the sediment and does this stimulation effect also show dependence on the sediment type?; and (3) what immobilization/remobilization mechanisms of U(VI) are likely to be involved during the biostimulation of the sediments?

## 2. Materials and methods

### 2.1. Site description and sediment sampling

The U.S. Department of Energy's Integrated Field Research Challenge (IFRC) site in Rifle, Colorado is located on a small flood plain in northwestern Colorado underlain by an aquifer comprised of 6.5 m of unconsolidated sands, silts, clays and gravels deposited by the adjacent Colorado River and underlain by a relatively impermeable aquitard known as the Wasatch formation (Anderson et al., 2003; Williams et al., 2011; Qafoku et al., 2014). This site was contaminated with U, V, As, Se and other contaminants from former milling activities. Groundwater monitoring over the past decade shows varying patterns of dissolved metals behavior, with significant fluctuations in aqueous U, V, As, and Se associated with seasonal variations in water level (Qafoku et al., 2014). The sediments labeled D08, LQ 107 and CD were collected from the uranium-contaminated subsurface at Winchester, La Quinta, and Plot C experimental galleries, respectively, whereas the RABS (Rifle Aquifer Background Sediment) was collected from an area that is not affected by the subsurface U(VI) plume (See the schematic in Supplementary Materials). Average groundwater composition has also been reported previously (Anderson et al., 2003; Yabusaki et al., 2007). In general, the aquifer has elevated concentrations of SO<sub>4</sub> (~10 mM) and alkalinity (~10 meq/L), and is close to equilibrium with calcite. The pH is typically between 7.2 and 7.4. Dissolved Fe(II) can be quite variable across the site (Campbell et al., 2012).

The same method as that described in Qafoku et al., 2014 has been used to collect the sediments. Specifically, sediment cores were collected using a nitrogen gas borehole flush to avoid air (oxygen) contamination, and sediment samples were immediately processed in a nitrogen-filled field-portable glove bag. Double-layered Mylar bags containing oxygen scavengers were used to pack the sediment samples before they were shipped on ice to the laboratory. Upon arrival at the laboratory, the sediment samples were stored at -80 °C prior to the column experiments. It is noted that the subsurface area where the sediment samples were collected had not been impacted by previous organic carbon amendment experiments performed at the Rifle IFRC site.

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