

MOBILIZATION OF NATURALLY OCCURRING URANIUM IN SEDIMENT INTO
GROUNDWATER

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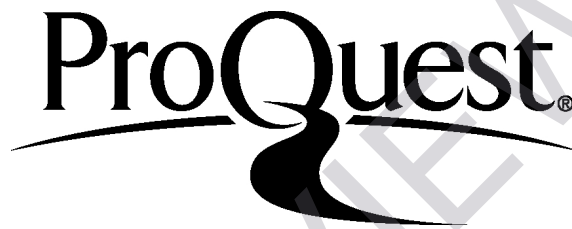
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MOBILIZATION OF NATURALLY OCCURRING URANIUM IN SEDIMENT INTO GROUNDWATER

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University of Nebraska, 2016

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Almost 10% of wells in the USA tested through the USGS National Water-Quality Assessment program produce water with uranium (U) in excess of the drinking water Maximum Contaminant Level (MCL) of $30\mu\text{g/L}$. Uranium exists primarily in two oxidation states in the environment, as oxidized U(VI), and reduced U(IV). Under natural conditions U(VI) readily complexes with ligands such as calcium and carbonate forming highly mobile aqueous species. Whereas, U(IV) primarily exists as an immobile solid-phase mineral. Biological and abiotic reactions can impact the mobility of naturally occurring U. One such reaction is the oxidation of U(IV) by oxidants such as molecular O_2 and nitrate producing U(VI). A strong correlation was identified between nitrate and U in two major USA aquifers, the High Plains and Central Valley aquifers. However, in regions with solid-phase U(VI)-bearing minerals, e.g. schoepite or U(VI) co-precipitated with calcite could account for immobilization of U(VI) in sediment. Desorption alone may not account for concentrations of groundwater U. Uranium(VI) adsorption is traditionally quantified under oxic conditions. In the presence of U(IV) minerals, oxic conditions can stimulate the oxidative dissolution of U(IV), leading to an underestimation of actual U(VI) sediment adsorption. Thus an anoxic method for determining U(VI) sediment adsorption in the presence of U(IV) minerals in

suboxic aquifers should be used. Even where U is present in sediment as an oxidized mineral, nitrate reduction reactions still have the potential to drive U mobilization by local acid production (H^+) production at mineral surfaces, thus causing mineral dissolution. The mobility of naturally occurring U is dependent on U valence state, sediment adsorption, and the availability of ligands for complexation or precipitation. As an influx of an oxidant such as O_2 or nitrate into aquifers occur, the potential for mobilization of naturally occurring U from sediments exist.

PREVIEW

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PREVIEW

CHAPTER 1

INTRODUCTION

Understanding the mobility of naturally occurring uranium (U) in aquifers is critical to the water security of the USA. Recently, U concentrations exceedingly over the US EPA maximum contaminant level (MCL) of $30\mu\text{g/L}$ (US EPA 2012) have been identified in groundwater across the US. These values are particularly high in shallow, unconsolidated aquifers (Ayotte et al., 2011; Nolan and Weber, 2015). Uranium mobility has been studied extensively, primarily in the context of nuclear legacy sites (milling, mining, and waste sites) where anthropogenic contamination has been identified (Campbell et al., 2015; Janot et al., 2016; Moon et al., 2009). The geochemical factors governing the mobility naturally occurring sedimentary U, or U in sediment and soil environments outside of primary deposits that is below ore grade, are poorly understood. Here I expand on some of the potential variables known to influence the mobility of naturally occurring sedimentary U in unconsolidated alluvial aquifers that are used heavily for irrigation and drinking water.

Uranium is a ubiquitous element in soils and sediments and on average is found at concentrations between 2 to 4mg kg^{-1} . Uranium is a naturally occurring radionuclide ultimately derived from the weathering of igneous rocks, and subsequently precipitated during the diagenesis of sediments that contain abundant organic carbon (Adler, 1974). The element typically exists mostly in one of two oxidation states in nature: as either as oxidized U(VI), or as reduced U(IV) (Langmuir, 1978). Uranium(VI) oxides are highly soluble whereas U(IV) oxides are not. The majority of U in soils and sediment, thus, occurs as insoluble reduced U(IV) minerals (e.g., uraninite) (Langmuir 1978). Uraninite

is typically associated with volcanic rocks, black shales, or granites (Adler, 1974). It has long been recognized that exposing reduced U(IV) minerals to oxidizing groundwater leads to oxidative dissolution and mobilization of U(VI) species (Langmuir, 1978).

Adsorption of U(VI) to sediment is typically considered the dominant mechanism inhibiting U mobility once oxidized, maintaining low U levels in oxic aquifers (Jurgens et al., 2010). But, a mix of both U(VI) and U(IV) in U minerals or co-precipitated as U(VI) minerals have also been identified at or near ore deposits (Finch and Ewing, 1992; Plasil, 2014; Reeder et al., 2001). Nevertheless, the mobility of natural U into groundwater is still not well understood.

The four objectives of this dissertation are:

- A. Thoroughly discuss recent literature relating to abiotic and biotic geochemical mechanisms known to contribute to U mobility in aquifers.
- B. Identify regions in two unconsolidated aquifers, High Plains (HP; Colorado, Kansas, Nebraska, New Mexico, Oklahoma, South Dakota, Texas, and Wyoming) and Central Valley (CV; California) aquifers, where U has been measured over the MCL and to determine which groundwater constituents or parameters influence U mobility.
- C. Geochemically characterize sediment and U mobility in an oxic ($> 5\text{mg/L O}_2$) aquifer where U was measured below the MCL.
- D. Develop a technique to quantify U(VI) adsorption to sediment. Specifically, by surface complexation in a suboxic, in the presence of reduced U(IV) minerals in suboxic aquifers.

Following this introduction, the dissertation begins by presenting the abiotic and biotic geochemical mechanisms known to contribute to U mobility in the subsurface. While the bulk of the work I discuss is based on studies conducted in environments anthropogenically contaminated with U, the mechanisms presented are also relevant to natural systems. One known oxidant of U, nitrate, I find shows a moderately strong correlation to the presence of nitrate in a metadata analysis. In fact, I also find nitrate is present in 78% of the locations where U is over the MCL. This indicates the potential link between nitrate additions to aquifers and the mobilization of U in groundwater, causing contamination. Levels U in groundwater up to 2,674 $\mu\text{g/L}$ (89 times the MCL) in one aquifer were identified. This chapter was published in *Environmental Science and Technology Letters* (Nolan and Weber, 2015) and was selected as a Science Editor's Choice (Science, 2015).

But in some areas, even in the presence of oxidants, U groundwater levels remain low, below the MCL. To explore this finding, I then present a comprehensive study of U mobility at a study site in Hastings, NE, where U levels below the MCL in groundwater. Oxidants (molecular $\text{O}_2 > 7.0\text{mg/L}$ and nitrate $> 9\text{mg/L}$) were present in groundwater, but U concentrations remain below the MCL. Using an anoxic method of intact core collection, I find the U in sediment was mostly oxidized, but remained immobile in sediment (85% \pm 10% by X-ray Adsorption Near-Edge Structure analysis). But equilibrium adsorption experiments and surface complexation modeling indicate levels of U(VI) associated with sediment, cannot be retained in sediment by adsorption alone. I then find that the U(VI) in sediment, is likely a solid-phase U(VI) mineral or linked to the presence of calcite.

But while equilibrium adsorption and surface complexation modeling are widely used to describe U(VI) sediment adsorption under oxic conditions, I then detail the results of an anoxic approach to these methods. Under suboxic or anoxic conditions, the presence of reduced phase U(IV) minerals can cause an interference in adsorption quantification due to oxidation of U(IV) in sediment. I therefore describe a method of equilibrium adsorption experiments and surface complexation modeling for U(VI) in suboxic sediment in the presence of U(IV). In previous studies, if reduced U(IV) did exist in sediment, it would present a substantial underestimation of sediment adsorption capacity for U(VI). I then conclude this dissertation by discussing some of my key findings and describe how ongoing influxes of oxidants can destabilize both reduced U(IV) minerals in sediment as well as oxidized solid-phase U(VI) minerals. Lastly, I discuss how this dissertation has contributed to this field of research.

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CHAPTER 2

NATURALLY OCCURRING URANIUM IN GROUNDWATER: A REVIEW

Abstract

Natural occurring U is recognized as a contaminant in groundwater throughout the USA (US EPA MCL of 30µg/L) with known negative health effects. This review summarizes results relevant to naturally occurring subsurface sedimentary U and present an understanding of the potential for mobilization of naturally-occurring U. Uranium in groundwater can come from the dissolution or weathering of U rich silicate minerals, desorption of adsorbed phases, or oxidative dissolution of U-rich minerals in sediment. Numerous biologically mediated and abiotic mechanisms have been thoroughly linked to these processes. Dissolved oxygen can directly increase U mobility through both oxidation of U directly or oxidation of organic carbon. Nitrate, a common co-contaminant with U in shallow subsurface aquifers, has been recognized to be a stronger oxidant of U than molecular oxygen at some legacy sites, where U was subsequently reduced. Further mechanisms that may play a role in U mobilization include the biologically-mediated reduction of nitrate or nitrite to NO_x species, directly, or coupled oxidation of reduced iron or manganese minerals. As well, complex interactions between U and sedimentary and dissolved organic/inorganic carbon can further explain ongoing disequilibrium and the potential for biogeochemical cycling. Thus it is likely a complex system of linked biological and abiotic mechanisms control natural U mobilization and a full understanding of these processes are needed given the potential for widespread U contamination of groundwater.

1. Introduction

Naturally occurring U is ubiquitous in sediments and soils across the United States, but typically occurs at higher concentrations in the western states due to the weathering of primary deposits in the Rocky Mountains (Adler, 1974; Langmuir, 1978). Uranium is a naturally occurring radionuclide, occurring in multiple minerals (e.g. uraninite, apatite, autinite) and at levels on average of $2\text{-}4\text{mg}\cdot\text{kg}^{-1}$ in soil and sediment (Cowan and Burnett, 1994). When U is mobilized in groundwater, it poses a public health hazard as both chemotoxicological (Kurtio et al., 2005; Vicente-Vicente et al., 2010) and a carcinogen if ingested (US EPA, 2012). Uranium contamination of groundwater is commonly associated with nuclear legacy mining, mill tailings, test sites, or nuclear fuel leaks. But U is a widespread natural constituent of groundwater across the USA (Nolan and Weber, 2015) and a noted increased reporting of U over the US EPA MCL of $30\mu\text{g/L}$ in groundwater may be due to increased groundwater testing, or anthropogenic influences on mobilization. The sources and mechanisms of mobilization of U into groundwater have chiefly been studied in the context of legacy sites (Campbell et al., 2013), leading to numerous studies that can be used as proxies for naturally occurring U mobility. The recent realization of widespread U contamination (Figure 1), as well as the potential of significance of climate change impacts on aquifers, has reinvigorated interest in U mobility (Jurgens et al., 2010; Nolan and Weber, 2015). Here, I review recent and fundamental research on the sources and mechanisms of mobilization of naturally occurring U in the environment.

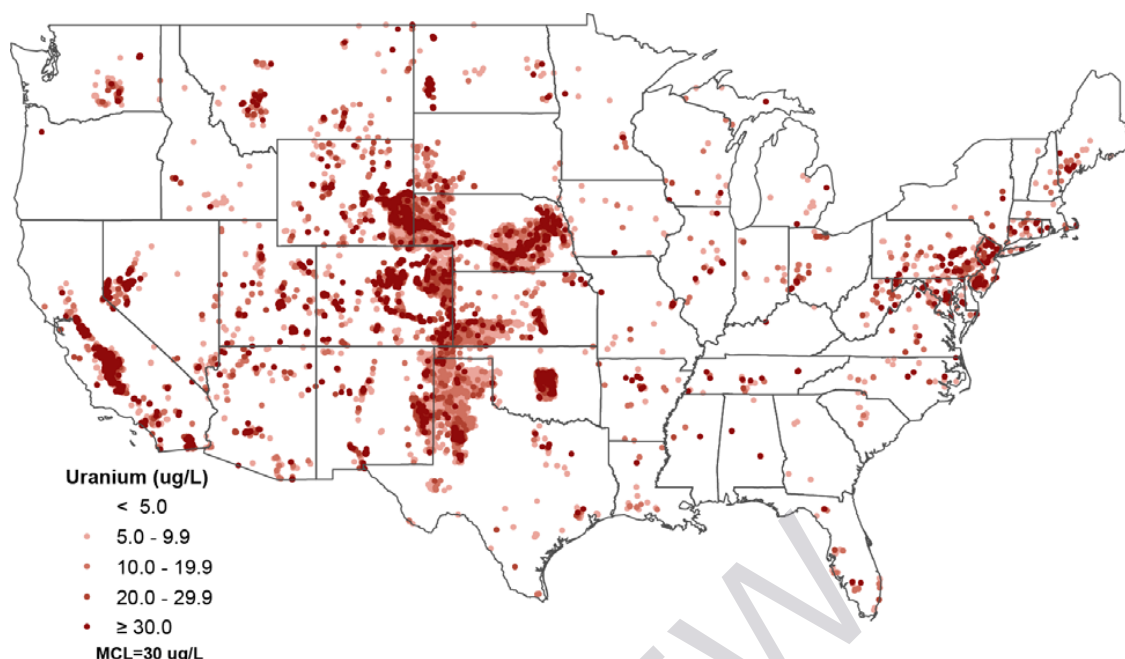


Figure 1. Uranium concentrations in groundwater across the United States using USGS National Water Quality Assessment (NWIS database). Modified from Nolan and Weber, 2015.

The most recent USGS National Water Quality Assessment (NAWQA) data retrieved from the National Water Information System (NWIS) indicates a trend in U mobilization into groundwater in vast regions of the USA (Ayotte et al., 2011). Around 10% of wells in the High Plains and Central Valley aquifers, the source of drinking water for 6 million people, have associated U levels that exceed the MCL of 30 μ g/L (Figure 1) (Federal Register, 2000; Nolan and Weber, 2015). Contamination of aquifers is a major threat to the groundwater security in the USA as increased depletions occur.

Groundwater is currently the primary source of drinking water for 46% of the nation (DeSimone et al., 2009), and 98% of drinking water used in rural communities (DeSimone et al., 2009). Under the current federal Safe Drinking Water Act (SDWA), domestic or “household wells”, and small rural systems that supply water to less than 25 people year round are exempted from testing for U contamination (DeSimone et al.,