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Mineralogy and geochemistry affecting arsenic solubility in sediment profiles from the shallow basin-fill aquifer of Cache Valley Basin, Utah

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ABSTRACT

Elevated arsenic concentrations have been reported in groundwater samples collected from the semi-arid Western U.S., including the Cache Valley Basin, Utah. The volcanic rock in the basin-fill aquifers underlying portions of the West is considered the primary source of arsenic, but there is debate about the mechanisms that control arsenic solubilization in these semi-arid and arid climates. Sediment cores were collected from a shallow basin fill aquifer in the Cache Valley Basin to systematically determine arsenic mineralogy and solubilization mechanisms in relation to non-redox and redox induced soil processes. Soluble arsenic was present throughout the two studied profiles in varying abundance and oxidation state, with the highest concentration of soluble As(V) and As(III) at the depth of the water table. Sequential chemical extractions of arsenic, with oxidation preservation strategies, revealed mineral sources and sinks of arsenic vulnerable to altering redox conditions down the profile. Weathering of primary arsenic-bearing minerals resulted in soluble arsenic in the vadose zone. Once soluble arsenic was leached to the deeper profile, arsenic solubility was controlled by carbonate minerals that concentrate at the water table. In the zone with alternating oxidizing and reducing conditions, iron oxides became the controlling mineral phase. The association between arsenic and sulfides limited arsenic solubility at depths under permanent water saturation. Arsenic solubility was revealed to be controlled by a sequence of processes that prevail under different geochemical regimes down the profile.

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1. Introduction

The occurrence of elevated arsenic (As) concentrations in groundwater threatens human health. Chronic ingestion of As bearing groundwater causes arsenicosis, which results in skin, bladder and kidney cancers, and blood vessel diseases (Ng et al., 2003). Affected aquifers have been found worldwide, most notoriously in Southern Asia where over a 100 million people have been exposed to As contaminated groundwater. As reviewed by Smedley and Kinniburgh (2002), arsenic entering groundwater is mostly naturally occurring rather than anthropogenic. Natural As sources

include primary and secondary As minerals, and secondarily formed As-containing Fe oxyhydroxides and other oxides. Microbial reductive dissolution of the host Fe oxide minerals is the major mechanism of As solubilization in groundwater systems in Southern Asia (McArthur et al., 2001; Nickson et al., 2000; Ravenscroft et al., 2001; Swartz et al., 2004). Direct microbial reduction of arsenate by dissimilatory arsenate respiring bacteria has also been reported (Ahmann et al., 1997; Weber et al., 2010). These microbial driven mechanisms require reducing conditions and the presence of labile organic carbon (OC) as carbon and energy sources for the dissimilatory reduction of Fe and/or As. The published studies delineating these mechanisms have been conducted in the field and laboratory using aquifer solids from zones with fluctuating redox conditions that are characterized by accumulation of Fe and Mn oxides. Sources and mechanisms that control solubility of As in sediments above and below the redox transition zone, where Fe and Mn oxides may not be the controlling mineral phase, are less

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studied.

Elevated As concentrations derived from near surface materials have been reported at some locations. Polizzotto et al. (2006) and Fendorf et al. (2010) stated that the weathering of primary As containing minerals in the Himalayas results in deposition of these materials in the river basins of Southern and Southeastern Asia. These As–S minerals are oxidized at the soil surface, with sorption of the As(V) to Fe oxides. This provides the continued source of As in deeper sediments where the Fe oxides undergo microbial reductive dissolution and release As to the groundwater. Thus, under oxidizing conditions Fe oxides are a sink for soluble As, but with redox cycling, under reducing conditions they become the source of groundwater As contamination.

Primary As-bearing sulfides are often related to high temperature geological activity (Drahota and Filippi, 2009). Volcanic related minerals were reported to be the primary source of As, including volcanic ash in the Southern High Plains aquifer of West Texas (Scanlon et al., 2009) and volcanic glass in the Rio Dulce alluvial aquifers of Argentina (Bhattacharya et al., 2006). The basin-fill aquifers of the American West, covering parts of New Mexico, Arizona, Southern California, Nevada, Oregon, Idaho, and Utah, contain volcanic rocks. Groundwater As concentrations that exceed the EPA maximum contaminant level (MCL) for As (10 µg/L) were reported in the Safford and San Pedro basins of Arizona (Robertson, 1989), the Socorro Valley of New Mexico (Brandvold, 2001), the Tulare Basin of the San Joaquin Valley of California (Fujii and Swain, 1995), the Willemette Basin of Oregon (Hinkle and Polette, 1999), the Western Snake River Plain Basin of Idaho (Busbee et al., 2009), the Southern Carson Desert of Nevada (Welch and Lico, 1998), and the Cache Valley Basin of Utah (Lowe et al., 2003). The role of primary As-bearing sulfides in the basin-fill aquifers in the West needs to be investigated.

Fluctuation of the water table not only alters redox conditions within aquifers, it also causes the formation and dissolution of carbonate minerals. Although carbonate minerals sequester much less As than oxides on a weight basis (Smedley and Kinniburgh, 2002), they are substantial soil components and are widely distributed in semi-arid subsurface systems. Calcite has been reported to sequester As through inner-sphere chemisorption, substitution, and surface complexation (Alexandratos et al., 2007; Bardelli et al., 2011; So et al., 2008). In addition to elevated concentrations of As in groundwater in the Cache Valley Basin, the groundwater and geology is highly carbonaceous. A microcosm study, using sediments collected from the depth of the water table in the shallow aquifer in this basin, showed that the solubilization of As from carbonate minerals, not Fe oxides, caused the elevated As concentration in the aqueous phase (Meng et al., 2016).

The solubilization of As cannot be attributed to a single process or mechanism from the soil surface to depth of groundwater; different processes will dominate as redox regimes alter with depth. The overall aim of this study was to determine the redox-induced variation in mechanisms of As solubilization in sediment profiles collected from a shallow aquifer in the Cache Valley Basin in Northern Utah. Elevated As concentrations, that exceed the MCL, were observed in 23 out of 157 wells sampled throughout Cache County, Utah (Lowe et al., 2003). The selected study area (41°44'03 N and 111°52'22 W) is located at the center of the southern portion of the Cache Valley. This site provided a unique opportunity to explore the role of carbonate minerals in controlling As solubilization. Two continuous sediment cores were collected from the ground surface into the permanently water saturated, reducing layers. Understanding the behavior of geologic arsenic in this location is important because these processes also affect other regions in the Western U.S. and around the world that have similar basin-fill geology and climate.

1.1. Geological settings

Cache Valley Basin is located in northeastern Utah and extends into southeastern Idaho with an area of about 1710 km² (Fig. 1). The elevation of the valley floor ranges from 1340 to 1650 m. The eastern portion of the valley is located on top of an ancient alluvial fan, caused by the withdrawal of Lake Bonneville, with steep slopes that reach into the rest of the basin and to the Logan River bottom (Hintze, 2005; USGS, 1962). The southern half of the basin is bounded by the western slopes of the Bear River Range and the eastern slopes of the northernmost branch of the Wasatch Range, the Wellsville Mountains. The Salt Lake Formation, which consists of Tertiary sedimentary and volcanic rocks, is exposed to the atmosphere along the foothills surrounding Cache Valley (Evans and Oaks, 1996), especially at the southern tip (Dover, 2005). Hintze (2005) attributed the volcanic rocks within the Salt Lake Formation to the migration of the Yellowstone “hotspot” from Northern Nevada, beginning 17 million years ago, to its current location. The valley floor is underlain by more than 300 m of unconsolidated basin-fill deposits consisting of Late Tertiary and Quaternary lacustrine sediments, due to ancient Lake Bonneville, and fluvial deposits (Brunner, 1991). Below that, lies up to 2100 m of the Salt Lake Formation.

1.2. Climate and hydrology

Climatic information (1981–2010) for this study was obtained from the weather station (Station ID: 425,182) nearest to the study site, belonging to the Global Historical Climatology Network. Temperature varied from a minimum of –34.4 °C to a maximum 39.4 °C. During the same time period, annual precipitation ranged from 217 to 833 mm, while annual evapotranspiration ranged from 987 to 1190 mm. Cache Valley is therefore classified as a steppeland in the Koppen–Geiger Climate Classification System, characterized by a cold semiarid climate (Kottek et al., 2006). Cache Valley basin is the easternmost extension of the Great Basin. The underlying aquifer consists mainly of unconsolidated basin-fill deposits of quaternary age from Lake Bonneville and older lakes, and the rest is younger alluvium at the margins of the valley (Kariya et al., 1994; USGS, 1962). The basin-fill aquifer is recharged by infiltration of precipitation, streams, irrigation water, and runoff from the surrounding mountains. In the center of the valley, groundwater is typically confined below depths of about 15 m because the intercalated clays act as confining layers that impede the upward flow of water. The horizontal hydraulic gradients decrease toward the center of the valley. In the shallow zone the water table is expected to be an approximate replica of the valley topography. The water table in the unconfined shallow aquifer ranged from less than 1 m to about 6 m (Kariya et al., 1994). Utah Water Research Laboratory (UWRL) personnel installed a total of 13 sampling wells at the center of the valley into the shallow aquifer. Groundwater depth was monitored from January 2009 to January 2010 by the UWRL. The highest water table was at the ground surface, while the lowest water table was 260 cm below ground surface (bgs); the highest and lowest water tables occurred in April and July, respectively. The groundwater has fluctuated up to 300 cm at a nearby U.S. Geological Survey groundwater monitoring site 41472111590001 (http://nwis.waterdata.usgs.gov/nwis/gwlevels/?site_no=41472111590001).

2. Materials and methods

2.1. Soil collection and processing

Continuous cores were collected in October 2012 from the areas

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