Column Tests and Multilevel Well Geochemistry to Explain Contaminant Plume Persistence Issues Downgradient of a Former Uranium Mill Site ✆

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Abstract

At a former uranium mill site in Riverton, Wyoming, USA, legacy mill tailings were moved to a nearby disposal cell. However, groundwater below and downgradient of the former tailings impoundment had already been contaminated. The compliance strategy is 100-year natural flushing, but the downgradient movement of plume contaminants has stalled near the groundwater discharge point. To better understand why this is occurring, core samples of alluvium were collected for analyses of solid-phase uranium concentrations and laboratory column testing. Multilevel wells were completed in the same core holes and monitored for almost 2.5 years. The resulting data indicate that a silt layer provides a storage mechanism for uranium, sulfate, and chloride. Uranium is more concentrated in the silt layer over the uranium plume. This stored uranium can later be released during flooding or high recharge events and creates a persistent uranium plume.

Keywords: uranium, Riverton, plume persistence, secondary contaminants

Introduction

Milling activities at a former uranium mill site near Riverton, Wyoming, USA, processed uranium ore from 1958 to 1963. Uranium mill tailings were removed and surface reclamation was completed by the U.S. Department of Energy (DOE) in 1989. However, shallow groundwater beneath and downgradient of the site is still contaminated with greater than 2 mg/L uranium. Detailed groundwater sampling was completed in 2012 (DOE 2013) using direct-push, temporary, piezometer sampling to create a detailed map of the uranium plume (fig. 1). Other contaminants are relatively coincident with the uranium plume, with some offsets for different contaminants (DOE 2016).

Groundwater modeling predicted that natural flushing of the groundwater aquifer to a nearby river would achieve compliance with applicable groundwater protection standards by the year 2097 (DOE 1998). Initial data from 1989 to 2009 indicated that overall, contaminant concentrations below the former mill site and downgradient of it were declining steadily. However, local flooding in 2010 mobilized stored contaminants in the downgradient floodplain and resulted in an increase in groundwater contaminant concentrations, including uranium (fig. 2). Thus, we define plume persistence as contaminant concentrations that remain at concentrations higher than originally predicted due to previously unrecognized contaminant sources. These stored contaminants or persistent secondary contaminant sources were not considered in the original conceptual site model and groundwater modeling predictions with natural flushing (Dam et al. 2015).
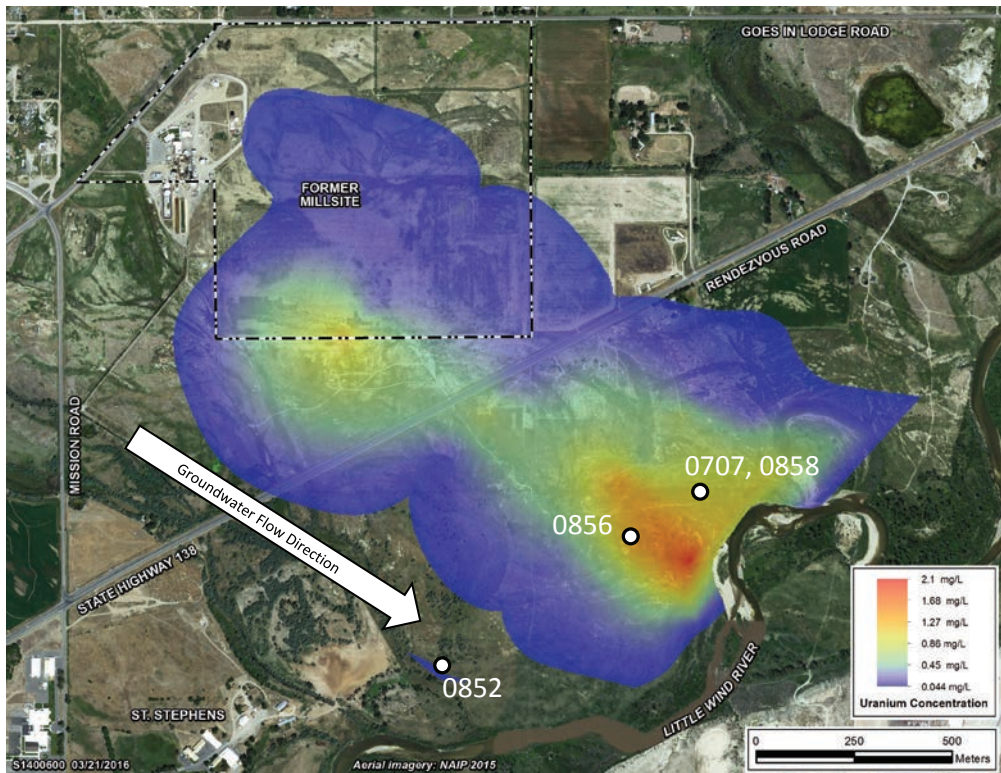
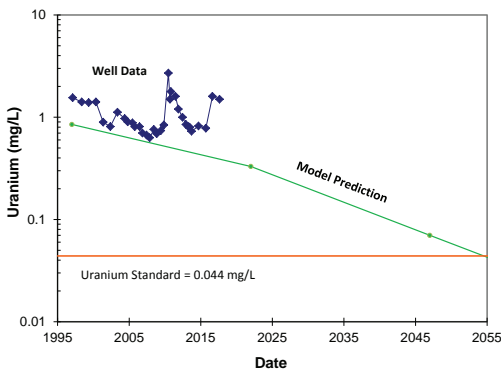*Figure 1 Uranium plume at the Riverton site in 2012 with key sample locations (red is higher concentration).*

Figure 2 Model predictions (DOE 1998) compared to measured uranium concentrations for well 0707 (see location in fig. 1).
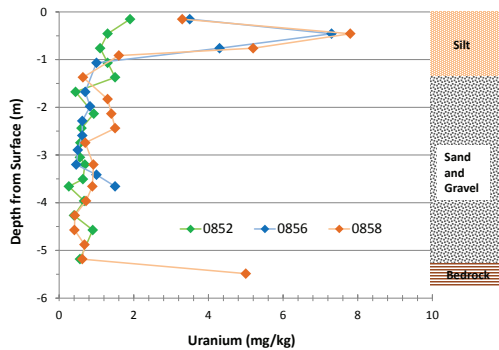

Figure 3 Solid-phase uranium data for locations 0852, 0856, and 0858 with generalized stratigraphy (see fig. 1 for locations).

The zone with higher uranium concentrations near the Little Wind River (fig. 1) was a focus area for solid-phase sampling in 2015 (DOE 2016) to determine the concentrations and locations of secondary contaminant sources. This work identified two main materials with higher uranium concentrations,

which are a silt layer from approximately 0 to 1.5 m below ground surface and an organic-rich zone at approximately 1.5 to 2.5 m below the ground surface (DOE 2016; Johnson et al. 2016). The general stratigraphy with solid-phase uranium concentrations within (0856 and 0858) and outside (0852) of the contami-

nant plume is shown in Figure 3. In general, the uppermost stratigraphic layer is 1.5 m of silt that remains unsaturated most of the year but can become saturated during spring run-off due to flooding or higher water-table conditions. The silt is underlain by about 4 m of sand and gravel, and the organic-rich zones are discontinuous silty layers (old river bank sediments) within the sand and gravel. The Wind River Formation is below the sand and gravel aquifer at about 5.5 m below ground surface and forms a semi-confining layer. While the organic-rich zone is an additional control on contaminant transport, this paper focuses on the release of contaminants from the silt zone. Column tests were performed on material from the silt layer and the underlying sand and gravel. These column results are compared to multilevel groundwater samples collected over almost 2.5 years at the same locations that the sediment for the column tests was collected.

Methods

Column tests were completed in 5.1 cm × 45.7 cm Plexiglas tubing packed with air-dried, <2 mm size sediment in 5.1 cm lifts, with tamping between lifts, until the column was filled with ≈22.9 cm of solid material. The solids were topped with a fabric filter and then with ≈6.4 cm of 5 mm clean glass beads to prevent material movement during column filling. A tube was placed at the top filter (top of the column) and attached to a 60 mL syringe to obtain the effluent pore fluid for analyses. The influent water was added to the column at the rate of ≈3 mL/min from the bottom. Each column was allowed to equilibrate for 24 ± 2 hours before another pore volume of influent was put through the column, and the effluent water was collected for analyses each day. The influent water for the columns was deionized water with 0.001 M HCl (provides 31 mg/L of chloride), unless otherwise noted. The small amount of acid was added to represent the slightly acidic conditions produced by soil gas carbon dioxide in the subsurface. Sample analyses were completed using ion chromatography (IC) and inductively coupled plasma mass spectrometry (ICP-MS) techniques.

Sediment core was collected using a sonic drilling rig with well installations completed in the same core hole. Multilevel well installations were completed using continuous multichannel tubing (CMT) that was cut in the field based on the geology to provide three sampling ports with depth. The CMT was attached to a traditional plastic well riser pipe with a bottom screen (0.3 m) to provide a fourth groundwater sampling interval. Water sampling was done using a peristaltic pump, and water analyses were completed using IC and ICP-MS techniques.

Column Test Results

Column test results are shown below for location 0852 silt (fig. 4), which is a location outside of the uranium plume and for location 0858 silt (fig. 5), which is within the uranium plume (fig. 1). Data from 0852 show that the release of uranium, sulfate, and chloride from the silt can occur in areas outside of the uranium plume and that uranium can be released above the maximum concentration limit of 44 µg/L. Likewise, the location 0858 silt layer also releases uranium, sulfate, and chloride, but with much higher uranium concentrations. The solid-phase data from the silt over the plume (fig. 3) indicate higher uranium concentrations, which are subsequently released in column flushing. In both columns, chloride flushes within the first pore volume whereas uranium and sulfate are released more slowly. In the 0858 silt column (fig. 5), 12 pore volumes are required to reach the uranium standard of 44 µg/L and 6 pore volumes are required to reach the sulfate secondary standard of 250 mg/L.

A column test of the sand and gravel aquifer material with background groundwater as the influent was also completed (fig. 6). These data show limited retention of uranium on the main aquifer material, similar to the flushing of sulfate and chloride (fig. 6). Both uranium and sulfate are flushed to below standards in less than 3 pore volumes after accounting for constituent release as a difference from the influent concentration.
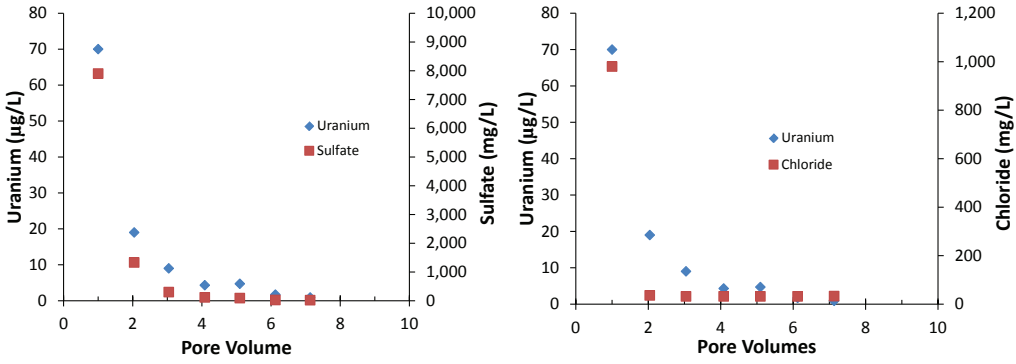
Figure 4 Column test: Effluent data from silt material at location 0852 collected 0.3–0.76 m below ground
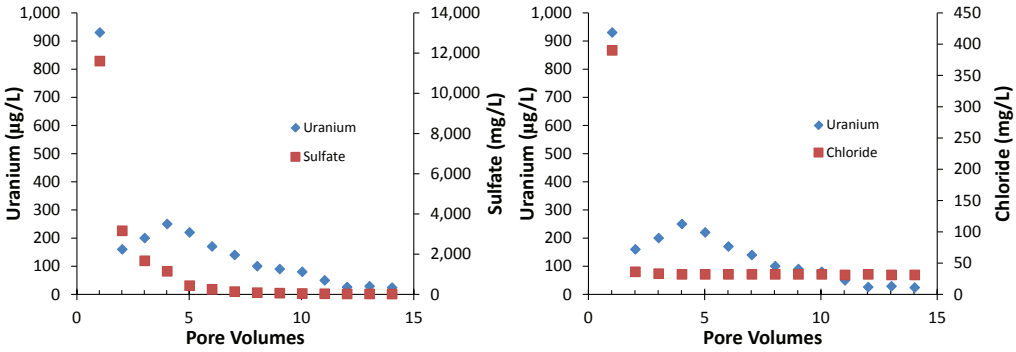

Figure 5 Column test: Effluent data from silt material at location 0858 collected 0–0.76 m below ground surface.
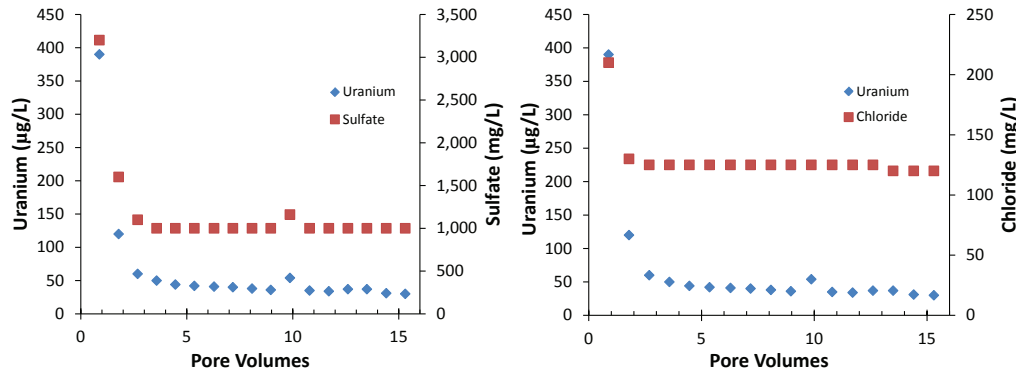

Figure 6 Column test: Effluent data from sand and gravel material at location 0858 (note that influent solution was background groundwater that had 28 µg/L uranium, 1,000 mg/L sulfate, and 125 mg/L chloride).

The column for the location 0858 silt has an additional geochemical control on the release of uranium based on the concentration curve reaching a secondary peak at 4 pore volumes. On the basis of a preliminary data evaluation, this appears to be due to changes in calcite solubility, which then influences uranium mobility. Geochemical modeling and detailed interpretations of the column data are still ongoing.

Multilevel Groundwater Sampling Results

Multilevel groundwater sampling has been ongoing at locations 0852 and 0858 for almost 2.5 years. Both locations show significant spikes in uranium concentrations in May of 2016 (figs. 7 and 8), especially at the shallower sampling ports. The solid line in Figures 7 and 8 indicates a large flooding event on May 8, 2016, due to heavy rainfall locally and upstream. Similarly, a dashed line in Figures 7 and 8 indicates another large rain event on March 31, 2017, which caused minor flooding. Additional flooding occurred from June 9 through 19, 2017 during runoff from mountain snow melt. Sulfate and chloride trends (not presented) are similar to the uranium results. The post-flooding (May 2016) sulfate concentration increases at 0852-1 and 0858-1 relative to 0852-4 and 0858-4 are similar at 5,200 and 5,400 mg/L, respectively, and chloride increases are 660 and 280 mg/L, respectively. The same comparisons for uranium increases are 120 and 2,520 µg/L, respectively (figs. 7 and 8). The April 2017 concentrations changes were more subtle. In May 2016, post-flood constituent increases start at the top of the aquifer, until mixing throughout the aquifer occurs with time (figs. 7 and 8). Over time, these increased concentrations decline as less contaminated groundwater from upgradient flushes through the sand and gravel aquifer.

Conclusions

Solid-phase data (fig. 3) indicate retention of uranium in the silt material over the uranium plume (fig. 1) that can contribute uranium to the underlying sand and gravel aquifer (figs. 7 and 8). Column tests provide uranium release concentrations (figs. 4 and 5) that are similar to those seen in the field (figs. 7 and 8). Column flushing of the silt took 12 pore volumes to get uranium concentrations below standards. A location outside the contaminant
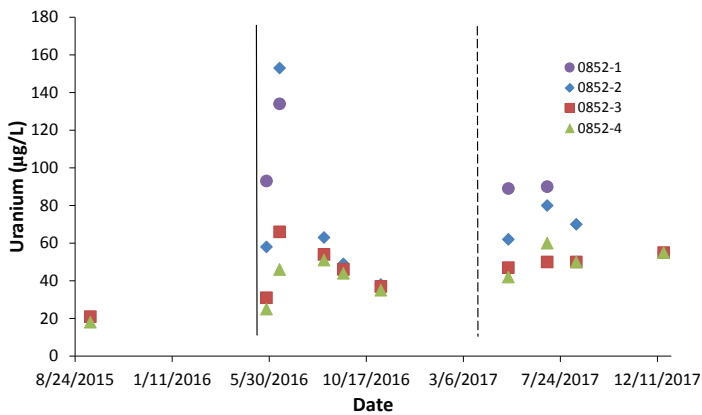

Figure 7 Multilevel groundwater data at location 0852 (outside of the plume). Solid line indicates flooding on May 8, 2015, and dashed line indicates flooding on March 31, 2017. Sample depths for levels 0858-1, -2, -3, and -4 are 1.6, 3.0, 3.7, and 4.4 m, respectively.
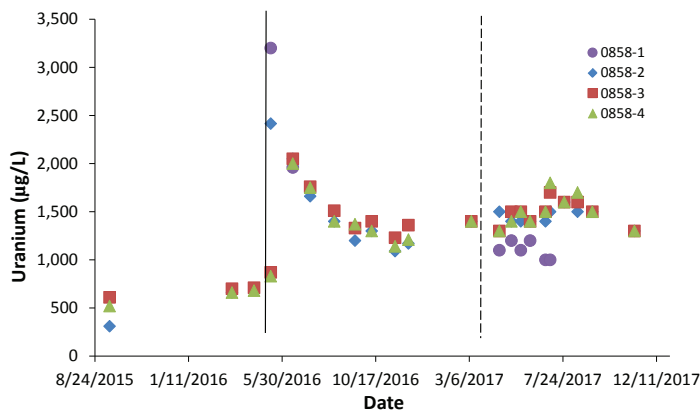

Figure 8 Multilevel groundwater data at location 0858 (within the plume). Solid line indicates flooding on May 8, 2015, and dashed line indicates flooding on March 31, 2017. Sample depths for levels 0858-1, -2, -3, and -4 are 1.1, 2.4, 3.9, and 5.3 m, respectively.

plume (0852) indicates that the release of uranium, sulfate, and chloride after flooding or high recharge events is a naturally occurring process. The sulfate and chloride release concentrations at the location outside of the contaminant (0852) are similar or even greater than from the location within the uranium plume. However, the data within the uranium plume (0858) indicate that the uranium release here is much greater than the uranium release at the location outside of the plume (0852). These results indicate that the silt layer provides a storage mechanism for uranium that creates a plume persistence issue. This storage and release keeps the uranium plume from declining near the Little Wind River and delays natural flushing beyond initial expectations.

The original conceptual model of natural flushing in the sand and gravel aquifer is still valid when looking at contaminant transport within the sand and gravel only. However, the continued inputs of uranium and other contaminants from the silt layer were not considered in the original conceptual model (Dam et al. 2015). As a result, the silt material coupled with flooding or high recharge events creates uranium concentration spikes after these events (figs. 2, 7, and 8). This leads to a new conceptual model that helps explain the plume persistence issues at the site.

Acknowledgements

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