

## Persistent Secondary Contaminant Sources at a Former Uranium Mill Site, Riverton, Wyoming, USA

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### Abstract

Milling activities at a former uranium mill site near Riverton, Wyoming, USA, contaminated the shallow groundwater beneath and downgradient of the site. Uranium mill tailings have been removed, and 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. From 1989 to 2009, 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. These stored contaminants or persistent secondary contaminant sources were not considered in the original conceptual site model and groundwater modeling predictions with natural flushing.

Additional data collection was performed on the floodplain in 2014 and 2015 to assess the possibility that secondary contaminant sources (specifically uranium) have been retained at the surface and in the subsurface through various processes. Data collection activities included surficial sediment sampling, trenching for direct subsurface observations, sonic drilling for core collection and multilevel well installations, and hand augering for shallow core collection and the installation of pore-water samplers.

Results indicate that uranium is being concentrated and stored in surface and subsurface evaporites and naturally reduced zones (NRZs) with high organic carbon and sulfide content. The evaporites and NRZs contain up to 66 and 140 mg/kg uranium, respectively, with resulting pore-water concentrations up to 2.68 mg/L uranium in the NRZs. These values compare to <1.3 mg/kg uranium in the typical underlying sand and gravel aquifer solids and up to 1.7 mg/L uranium in the current groundwater. It appears likely that the NRZs provide a mechanism to concentrate uranium under reducing conditions. Subsequent oxidation of the NRZs provides a release mechanism for uranium to the overlying unsaturated zone pore waters and the underlying groundwater. Due to a shallow water table and arid conditions that produce high evapotranspiration rates, the uranium-rich pore waters provide a mechanism to form overlying uranium-rich evaporites. The presence and understanding of these persistent secondary contaminant sources can be used for more informed management of the Riverton site.

Key words: uranium, persistent contaminants, evaporites, naturally reduced zones

### Introduction

Milling activities at a former uranium mill site near Riverton, Wyoming, USA, took place 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 (fig. 1) with up to 2.1 mg/L uranium in 2012 and up to 1.7 mg/L uranium currently. 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), which lead to additional site characterization (DOE 2013). 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).

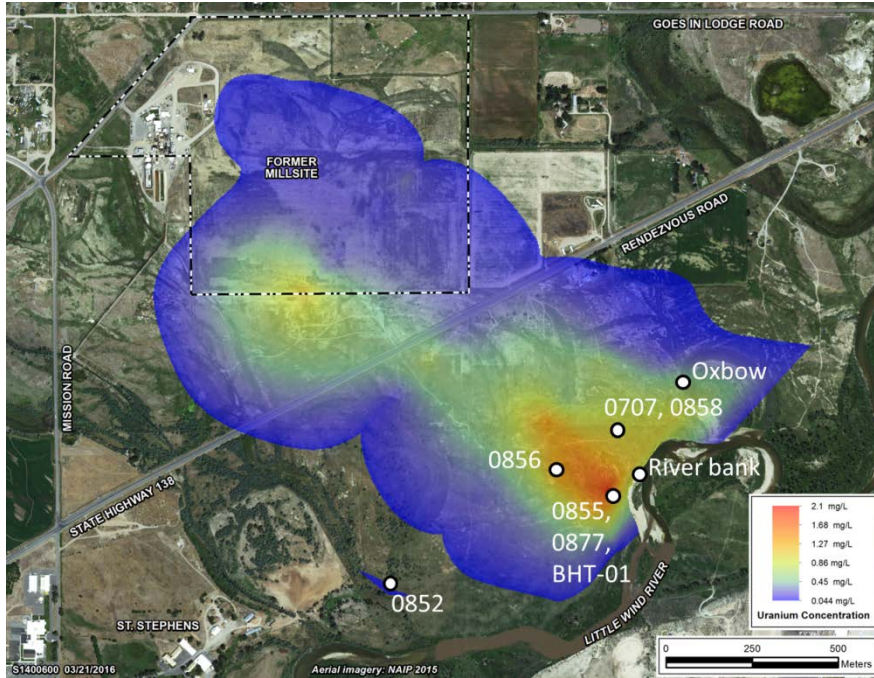


Figure 1 Uranium plume at the Riverton site in 2012 with key sample locations.

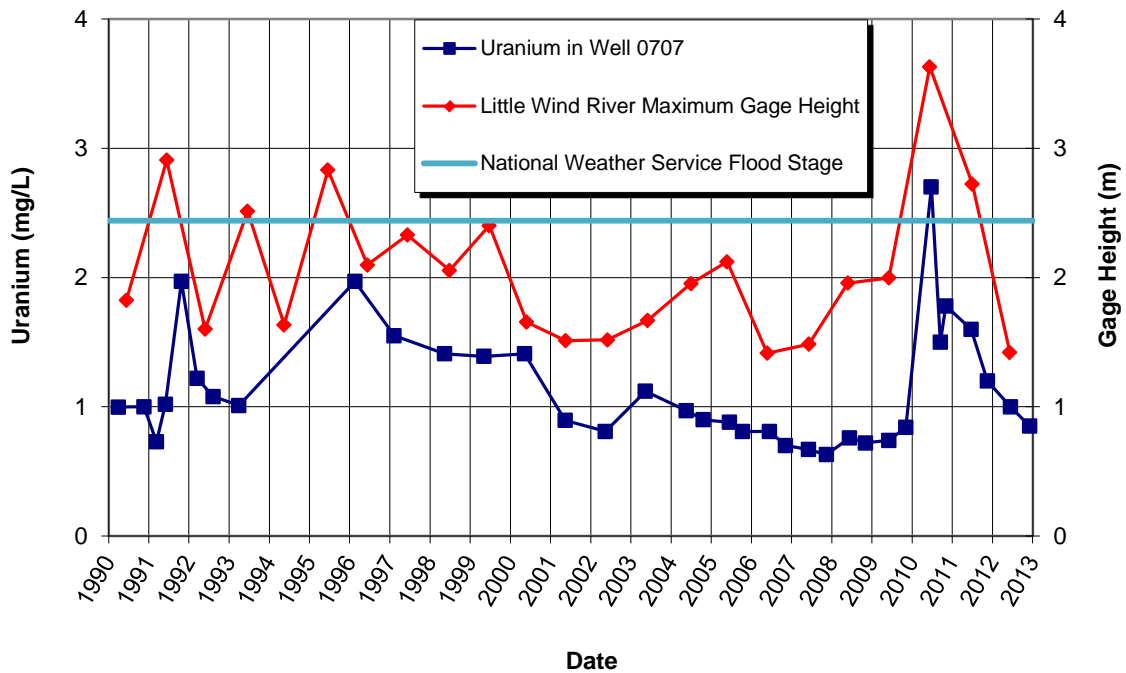


Figure 2 Hydrograph for the Little Wind River compared with uranium concentrations at well 0707 (see location in fig. 1).

This paper focuses on additional data collection performed on the floodplain in 2014 and 2015 to assess the possibility that secondary contaminant sources (specifically uranium) have been retained at the surface and in the subsurface through various processes. Data collection activities included: surficial sediment sampling, trenching for direct subsurface observations, sonic drilling for core collection and multilevel well installations, and hand augering for shallow core collection and the installation of pore-water samplers.

## Methods

Solid-phase samples were analyzed for trace metals and cations using a total acid digestion with subsequent inductively coupled plasma mass spectrometry (ICP-MS). Some solid-phase samples were analyzed using x-ray fluorescence (XRF). Solid-phase anions were analyzed using a water leach and subsequent ion chromatography (IC) analyses.

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 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. Pore water was sampled using hand pumping suction devices with an end filter (suction lysimeter) installed directly into the ground, either in the side of trench walls or subsurface in hand-augered holes.

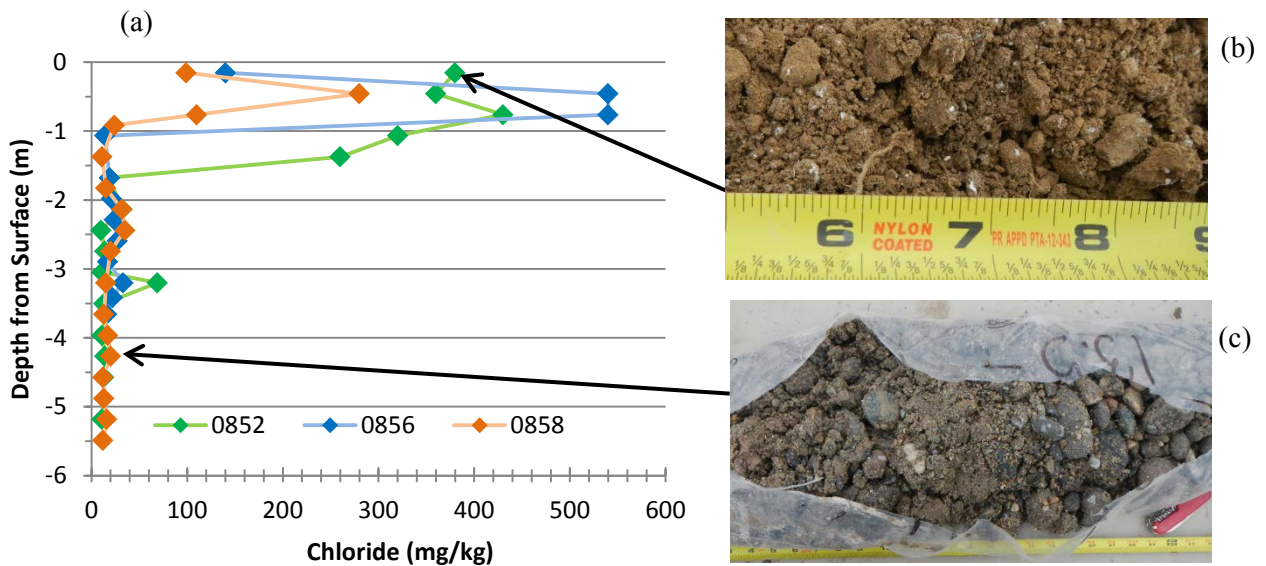
## Evaporites

Surficial evaporite deposits or efflorescent salt crusts (fig. 3) can be found along the river banks. Background concentrations of uranium in these deposits are approximately 1.4 mg/kg uranium, but the uranium concentrations in surficial evaporites over the plume are as high as 66 mg/kg uranium (DOE 2014).

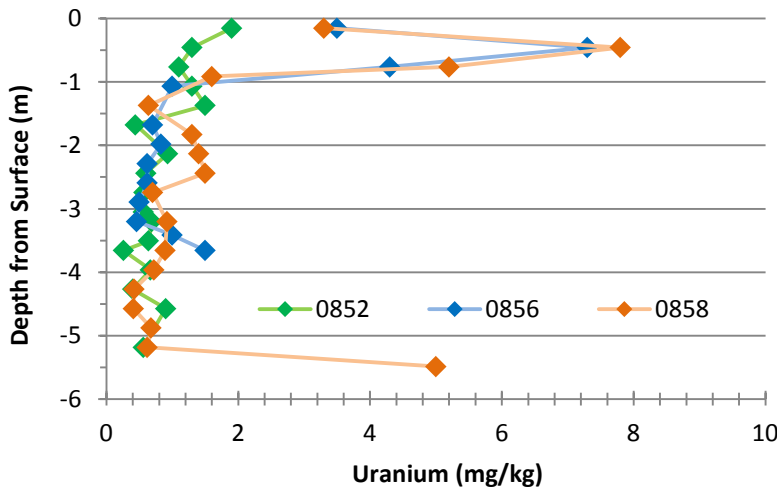


*Figure 3* Evaporite deposits found along the Little Wind River.

The general stratigraphy at the Riverton site is a 0.76 to 1.37 m silt layer (fig. 4b) underlain by 1.92 to 4.63 m of sand and gravel (fig. 4c), which is then underlain by lower-hydraulic-conductivity bedrock (Wind River Formation). The silt layer is generally above the water table but can be within the capillary fringe depending on the time of year and the water-table stage. This silt layer appears to be concentrating a variety of constituents (not shown) in subsurface evaporites. The presence of subsurface evaporites was visually confirmed in the sides of trench walls, hand-augered material, and in sonic drilling cores (fig. 4b). The concentrations of elements that occur naturally, such as chloride, appear to be independent of mill-related plume concentrations (fig. 4). However, the anthropogenic source of uranium does appear to concentrate in the subsurface evaporites in areas over the uranium plume (fig. 5), with up to 8 mg/kg uranium in the silt over the plume compared to approximately 1.5 mg/kg uranium from the silt in borehole 0852 outside of the plume.



**Figure 4** Solid-phase chloride concentrations with depth (a) with photographs of silt with white evaporites flecks (b) and underlying sand and gravel (c). Borehole 0852 is outside of the uranium plume, and boreholes 0856 and 0858 are within the uranium plume (see locations in fig. 1).

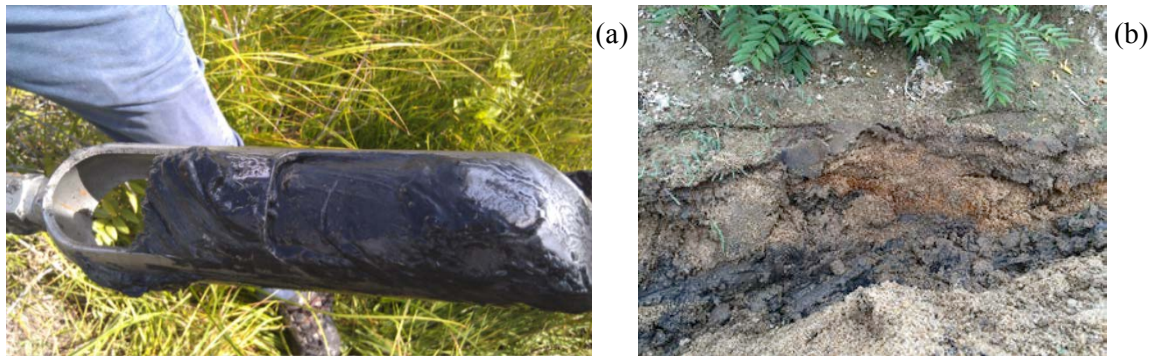


**Figure 5** Solid-phase uranium concentrations with depth. Borehole 0852 is outside of the uranium plume and boreholes 0856 and 0858 are within the uranium plume (see locations in fig. 1).

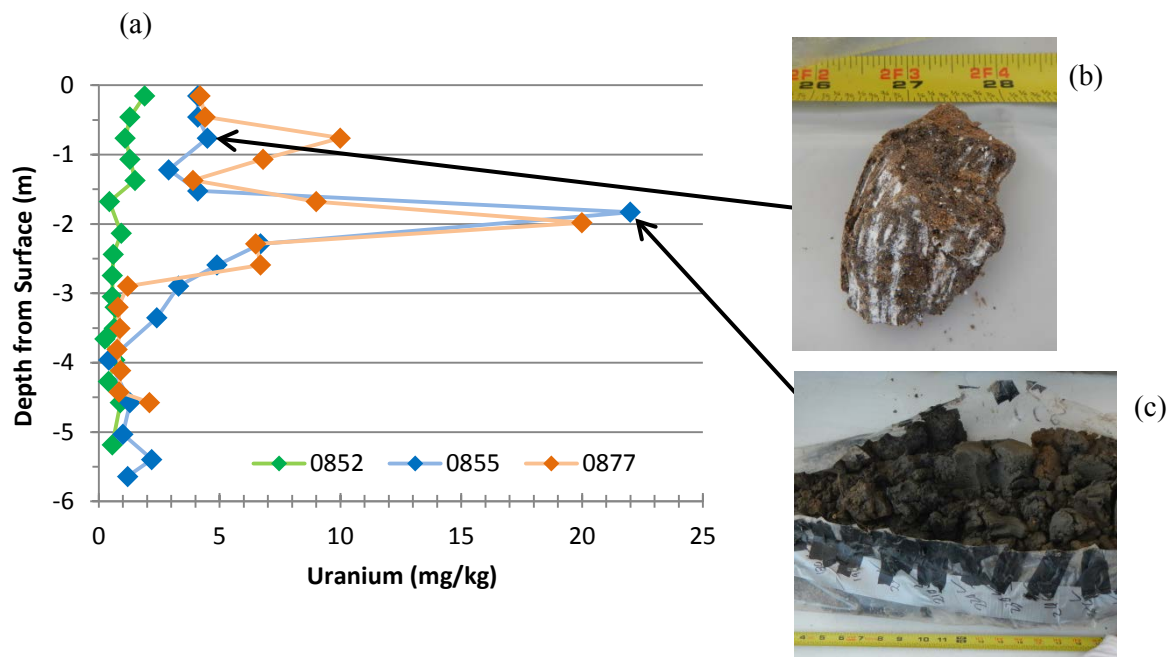
### Naturally Reduced Zones (NRZs)

NRZs at the Riverton site occur in sediments within the area of a river cutoff meander that has formed an oxbow lake (fig. 6a), in sediment near the riverbank (fig. 6b), and in subsurface sediments away from the river (figs. 7a and c). The defining characteristics of these NRZs are fine-grained sediments with enough organic carbon to produce sufficient reducing capacity to maintain anoxic conditions when saturated. Under these conditions, uranium is less soluble in water and tends to sorb and/or precipitate on the solid phase. As a result, XRF data for the oxbow and riverbank NRZs range from 22 to 140 mg/kg uranium. Subsurface NRZs have concentrations up to 22 mg/kg (fig. 7a) compared to generally less than 2 mg/kg uranium in the underlying sand and gravel. Uranium concentrations in the subsurface evaporites that occur above the NRZs (fig. 7b) range from about 4 to 10 mg/kg uranium (fig. 7a), which is similar to those of evaporites that do not occur over an NRZ, which range from 3 to 8 mg/kg uranium (fig. 5).





**Figure 6** Hand-auger sample of oxbow sediments (a) and a cross-sectional view of an NRZ at the riverbank (b). For sample locations see fig. 1. Note the oxidized top portion in (a) with iron hydroxide staining.



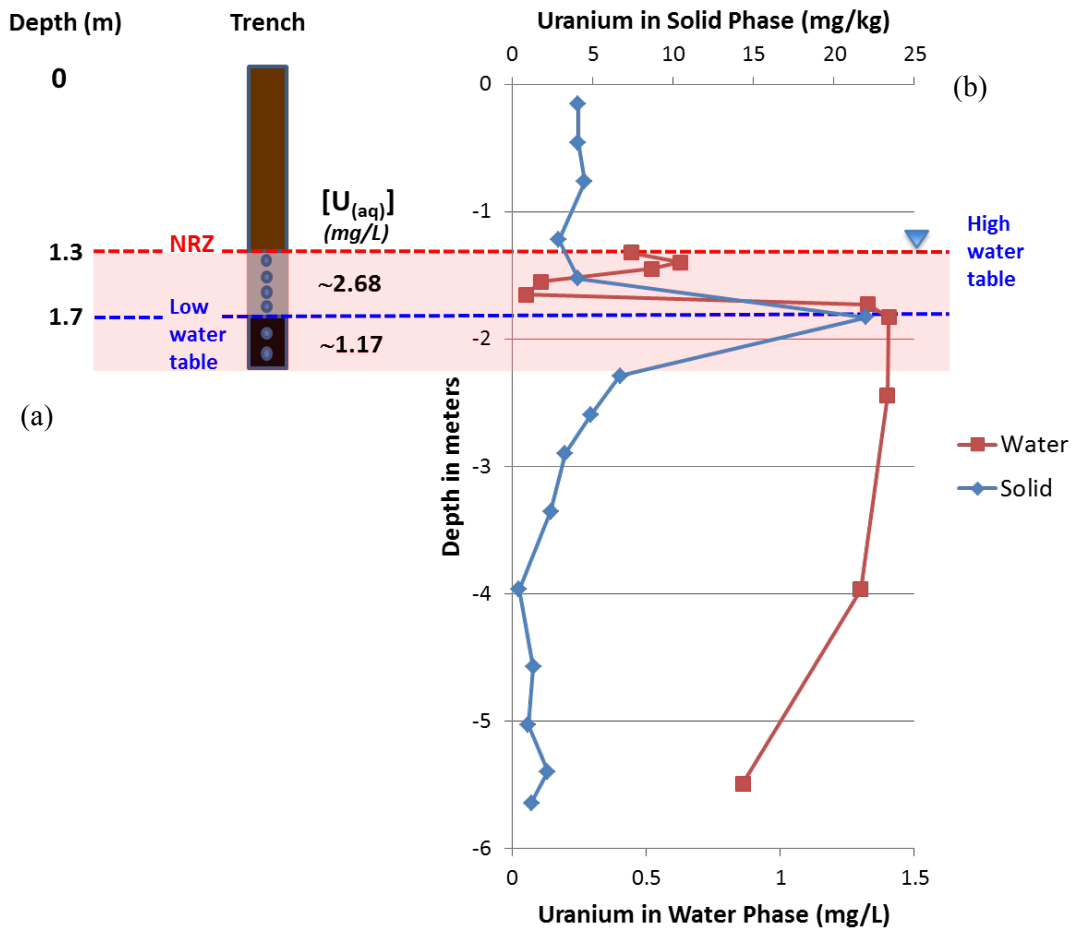
**Figure 7** Solid-phase uranium concentrations with depth (a) from borehole 0852 outside of the uranium plume and boreholes 0855 and 0877 within the uranium plume (see locations in fig. 1) along with photographs of evaporites (b) and an NRZ zone (c).

**Groundwater**

Uranium in pore water within and above an NRZ zone was sampled from a trench in May 2015 (fig. 8a) and then again in August 2015 using pore-water samples installed in a hand-augered hole (fig. 8b). Uranium concentrations in groundwater from the multilevel well installations (three points at greater than 2 m depth) were sampled in September 2015 and are plotted along with the August 2015 pore-water data and compared to uranium concentrations in the solid phase (fig. 8b). In August/September 2015 the uranium concentrations in water are highest in the bottom half of the NRZ and in the underlying groundwater compared to the above pore water and the groundwater below 5 meters (fig. 8b). The water table in this area likely reaches the top of the NRZ in the spring at approximately 1.3 m in depth but was close to 1.6 m in depth in August/September 2015. With a lower water table in May 2015 at above 1.7 m, before the spring snowmelt and high river stage, the upper part of the NRZ is unsaturated and has likely been oxidizing though the fall and winter months. This results in higher uranium concentration in pore water (fig. 8a) in the upper part of the NRZ, up to 2.68 mg/L. By August/September 2015 the pore water in the upper part of the NRZ was likely flushed to the

underlying groundwater, thereby decreasing the uranium in the upper zone to less than 0.7 mg/L and increasing the uranium in the bottom part of the NRZ from 1.17 mg/L to 1.4 mg/L (fig. 8).

Overall, it appears likely that the NRZ zone can maintain a groundwater uranium concentration of 1.4 mg/L compared to the underlying uranium concentration of 0.86 mg/L in the sand and gravel just above the bedrock interface, which may represent upgradient plume water. Seasonal measurements of uranium in the pore water and groundwater in and above this NRZ zone are ongoing. These seasonal measurements should assist in better understanding the contributing amount of uranium to the groundwater from overlying evaporites compared to the underlying NRZs. Release of uranium from the evaporites does require a higher water table and/or flood conditions such that these evaporites become saturated, with a resulting release of uranium to the groundwater. As such, uranium release from the evaporites may not occur every year, while uranium release from the NRZs may occur on a more seasonal basis.



**Figure 8** Pore-water uranium concentrations in May 2015 from trench BHT-01 (see location in fig. 1) with an NRZ (a) and pore water and groundwater uranium concentrations in August/September 2015 compared to solid-phase uranium concentrations (b) from the same general location.

### Conclusions

Measured solid-phase uranium concentrations in evaporites and NRZs indicate higher concentrations of uranium and provide potential secondary contaminant sources that can delay natural flushing at the Riverton site. Due to a shallow water table and arid conditions that produce high evapotranspiration rates, the underlying uranium-rich plume waters provide a mechanism to form overlying uranium-rich evaporites (with or without NRZs). The reducing conditions produced by NRZs provide a mechanism to concentrate uranium; however, subsequent oxidation of the NRZs provides a release mechanism for uranium to the overlying unsaturated zone pore waters and the underlying groundwater. Seasonal

measurements of pore water and groundwater at the site near the evaporites and NRZs are ongoing in order to better understand the amount and rates of uranium release. Overall, the presence and understanding of these persistent secondary contaminant sources and how they relate to plume persistence can be used for more informed management of the Riverton site.

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