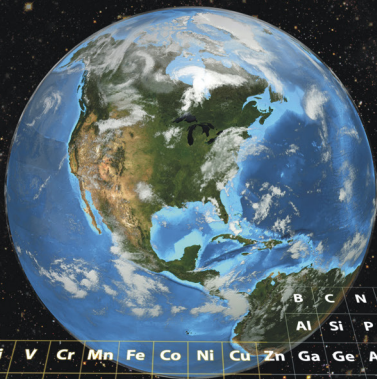


Environmental Sciences Laboratory

Literature Review of Documented Persistent Secondary Uranium Sources at DOE LM Sites

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U.S. DEPARTMENT OF
ENERGY

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Abbreviations

AEC	U.S. Atomic Energy Commission
ARCO	Atlantic Richfield Company
bgs	below ground surface
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CFR	<i>Code of Federal Regulations</i>
COPC	contaminant of potential concern
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
GMA	Great Miami Aquifer
km	kilometers
KNTZ	Kayenta/Navajo Transition Zone
LM	Office of Legacy Management
mg/kg	milligrams per kilogram
mg/L	milligrams per liter
µg/L	micrograms per liter
NRZ	naturally reduced zone
OU	operable unit
pCi/g	picocuries per gram
Ra	radium
RCRA	Resource Conservation and Recovery Act
SAG	San Andres Glorieta
U	uranium
UMTRA	Uranium Mill Tailings Remedial Action
UMTRCA	Uranium Mill Tailings Radiation Control Act
V	vanadium

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Executive Summary

The U.S. Department of Energy Office of Legacy Management (LM) has the responsibility to protect human health and the environment at 101 legacy uranium sites across the nation by implementing long-term surveillance and maintenance requirements. Included in these responsibilities is to provide for the safe and environmentally sound disposal, long-term stabilization, and control of uranium mill tailings in a manner that minimizes or eliminates radiation health hazards to the public at 21 Uranium Mill Tailings Radiation Control Act (UMTRCA) Title I sites; 6 UMTRCA Title II sites; and 9 Comprehensive Environmental Response, Compensation, and Liability Act/Resource Conservation and Recovery Act sites. These sites are former uranium mills (processing sites), or sites with wastes from processing sites encapsulated in engineered disposal cells (disposal sites), or sites where components of atomic weapons were manufactured. During site characterization, groundwater contamination was often discovered and evaluated or modeled for future movement with the inclusion of uranium sorption in the groundwater plume. Much of this original work did identify uranium processing wastes as the original contaminant source. However, more recent work has identified the existence and influence of persistent secondary uranium sources that may prevent uranium in groundwater from attenuating as quickly as initial evaluations or modeling indicated. By understanding these processes, better decisions can be made to choose the appropriate remediation and closure strategies, identify long-term surveillance and maintenance activities, and more accurately estimate life-cycle costs.

For this report, persistent secondary uranium sources are defined in three categories as (1) uranium that is precipitated or sorbed in the unsaturated or saturated zone below former tailings, former evaporation ponds, or existing disposal cells in concentrations and forms (e.g., uranium mineral precipitates, uranium sorption to excess precipitated iron) not typical of sorption to aquifer solids within the main groundwater uranium plume; (2) uranium that is retained in downgradient areas that is precipitated or sorbed in the unsaturated or saturated zones in concentrations and forms (e.g., evaporites with uranium in the unsaturated zone, sorption in high organic carbon zones, precipitation due to geochemical changes) not typical of sorption to aquifer solids within the main groundwater uranium plume; and (3) uranium with rate-limited mass transfer or nonequilibrium processes, such as diffusion out of a clay or other lower-permeability material. While these same processes can create persistent secondary sources for other contaminants, the scope of this literature review is limited to uranium only.

Sites with documented persistent secondary uranium sources under category 1 related to uranium nontypically precipitated or sorbed in the unsaturated or saturated zones below source areas include:

- Grand Junction, Colorado, Site
- Grand Junction, Colorado, Processing Site
- Gunnison, Colorado, Processing Site
- Monticello, Utah, Processing Site
- Monument Valley, Arizona, Processing Site
- Naturita, Colorado, Processing Site
- Rifle, Colorado, Old Processing Site

- Riverton, Wyoming, Processing Site
- Tuba City, Arizona, Disposal Site

In addition to the category 1 documented persistent secondary uranium sources, for the Bluewater, New Mexico, Disposal Site, a mineralized zone with secondary uranium below the disposal cell is theorized but was not directly measured. At the Fernald Preserve, Ohio, Site, residual uranium after source zone removal has not been directly measured, but remedial levels for solid-phase uranium were above background values. For the Shiprock, New Mexico, Disposal Site, a mineralized zone below the disposal cell is also likely but has not been directly measured.

Sites with documented persistent secondary uranium sources under category 2 related to downgradient evaporation and evaporite deposition include:

- Green River, Utah, Disposal Site
- Monticello, Utah, Processing Site
- Riverton, Wyoming, Processing Site
- Shiprock, New Mexico, Disposal Site

Sites with documented persistent secondary uranium sources under category 2 related to high organic carbon zones include:

- Grand Junction, Colorado, Site
- Naturita, Colorado, Processing Site
- Rifle, Colorado, Old Processing Sites
- Riverton, Wyoming, Processing Site
- Shiprock, New Mexico, Disposal Site

Sites with persistent secondary uranium sources under category 3 of rate-limited mass transfer or nonequilibrium processes include:

- Bluewater, New Mexico, Disposal Site
- Fernald Preserve, Ohio, Site
- Rifle, Colorado, Old Processing Site
- Tuba City, Arizona, Disposal Site

Rate-limited mass transfer or nonequilibrium (dual domain) processes likely occur at all LM sites to varying degrees based on hydraulic conductivity variations, which can create uranium concentrations that take much longer to decline than predicted. The sites listed in category 3 all had documents that specifically mentioned these processes. However, it is difficult to quantify and separate nonequilibrium processes from other persistent secondary uranium sources. Thus, no specific documents with the quantification of rate-limited mass transfer are available.

Prior LM site conceptual and numerical models did not typically include persistent secondary uranium sources. Slower natural flushing of uranium in groundwater than expected or predicted

by these prior models indicates the need to include persistent secondary uranium sources. Thus, this report is intended to provide a useful reference when evaluating other LM sites where processes that create persistent secondary uranium sources are possible but not documented.

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1.0 Introduction

Congress passed the Uranium Mill Tailings Radiation Control Act (UMTRCA) in 1978 (Public Law 95-604), which assigned the U.S. Department of Energy (DOE) the responsibility to provide for the safe and environmentally sound disposal, long-term stabilization, and control of uranium mill tailings or uranium processing wastes in a manner that minimizes or eliminates radiation health hazards to the public. At some of these uranium processing sites, groundwater was contaminated with uranium and other chemicals. DOE created the Uranium Mill Tailings Remedial Action (UMTRA) program to decommission the uranium mills and dispose of their residual mill tailings in accordance with standards promulgated by the U.S. Environmental Protection Agency (EPA) in Title 40 *Code of Federal Regulations* Section 192 (40 CFR 192). Subpart B of 40 CFR 192 regulates the cleanup of contaminated groundwater at the processing sites.

Currently, the U.S. Department of Energy Office of Legacy Management (LM) has the responsibility to protect human health and the environment at 101 legacy uranium sites across the nation by implementing long-term surveillance and maintenance requirements. Specifically, LM manages 21 UMTRCA Title I sites; 6 UMTRCA Title II sites; 5 DOE Defense Decontamination and Decommissioning program sites; and 9 Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA)/Resource Conservation and Recovery Act (RCRA) sites. UMTRCA Title I and II sites are inactive uranium-ore processing sites. Title I sites were inactive when UMTRCA was passed, and Title II sites became inactive after UMTRCA was passed. CERCLA sites are also known as Superfund sites, under a federal government program designed to fund the cleanup of sites contaminated with hazardous substances and pollutants. RCRA is a public law passed by Congress that gives EPA the authority to control hazardous waste from the “cradle-to-grave,” including its generation, transportation, treatment, storage, and disposal.

1.1 Purpose and Scope

During site characterization, groundwater contamination was often discovered and evaluated or modeled for future movement. Much of this original work did identify tailings impoundments, evaporation pond seepage, or other uranium processing wastes as the original contaminant sources. In addition, uranium (U) sorption/desorption within groundwater contaminant plumes was often identified and modeled as a process that can retard uranium movement to be slower than ambient groundwater flow velocities. However, work done primarily at the Riverton, Wyoming, Processing and Rifle, Colorado, Old Processing Sites identified the existence and influence of persistent secondary uranium sources that may prevent uranium in groundwater from attenuating as quickly as initial evaluations or modeling indicated. By understanding processes related to persistent secondary uranium sources, better decisions can be made to choose the appropriate remediation and closure strategies, identify long-term surveillance and maintenance activities, and more accurately estimate life-cycle costs.

Persistent secondary uranium sources for this report are defined with the following categories (1) uranium that is precipitated or sorbed below former tailings, former evaporation ponds, or existing disposal cells in the unsaturated or saturated zones and in concentrations and forms (e.g., uranium mineral precipitates, uranium sorption to excess precipitated iron) that are not typical of sorption to aquifer solids within the main groundwater uranium plume; (2) uranium

retained in downgradient areas that is precipitated or sorbed in the unsaturated or saturated zones in concentrations and forms (e.g., evaporites with uranium in the unsaturated zone, sorption in high organic carbon zones, precipitation due to geochemical changes) that are not typical of sorption to aquifer solids within the main groundwater uranium plume; and (3) uranium with rate-limited mass transfer or nonequilibrium (dual domain) processes, such as diffusion out of a clays or other lower-permeability material. Although these same processes can create persistent secondary sources for other contaminant sources, the scope of this literature review is limited to uranium.

The purpose of this report is to summarize the literature available on documented persistence secondary uranium sources at LM sites as part of the Applied Studies and Technology Persistent Secondary Contaminant Sources Project. All UMTRCA Title I, Title II, and CERCLA/RCRA sites that were in the LM program at the time of this report (21 UMTRCA Title I sites; 6 UMTRCA Title II sites; and 9 CERCLA/RCRA sites, <https://www.energy.gov/lm/sites/lm-sites>) were reviewed, along with the Decontamination and Decommissioning program Grand Junction, Colorado, Site. Other LM and DOE sites outside of the LM program (i.e., National Laboratory sites) were not reviewed for this report.

1.2 Site Summary and Report Organization

Conclusive documentation of persistent secondary uranium sources was found for the following sites:

- Bluewater, New Mexico, Disposal Site
- Fernald Preserve, Ohio, Site
- Grand Junction, Colorado, Site
- Grand Junction, Colorado, Processing Site
- Green River, Utah, Disposal Site
- Gunnison, Colorado, Processing Site
- Monticello, Utah, Processing Site
- Monument Valley, Arizona, Processing Site
- Naturita, Colorado, Processing Site
- Rifle, Colorado, Old and New Processing Sites
- Riverton, Wyoming, Processing Site
- Shiprock, New Mexico, Disposal Site
- Tuba City, Arizona, Disposal Site

In Section 2.0 a summary of each of these sites is provided, which includes a brief site history, description of site hydrogeology, and the literature describing the existence of persistent secondary uranium sources. Other LM sites may have persistent secondary uranium sources that have not been documented. However, this report focuses only on directly documented persistence secondary uranium sources. A separate summary section (Section 3.0) describes hydrogeochemical processes that can cause persistent secondary uranium sources, with a listing of LM sites where those processes have been documented (references are provided in

Section 4.0). Section 3.0 provides a useful summary to use when evaluating these processes at other LM sites where persistent secondary uranium sources are possible but not documented.

2.0 LM Sites with Documented Persistent Secondary Uranium Sources

2.1 Bluewater, New Mexico, Disposal Site

The Bluewater, New Mexico, Disposal Site is the location of a former uranium-ore processing mill that operated from 1953 to 1959 using a carbonate-leach process and from 1957 to 1982 using an acid-leach process (DOE 2016a). The site is in Cibola County, approximately 9 miles northwest of Grants, New Mexico (Figure 1). Anaconda Copper, which became a subsidiary of the Atlantic Richfield Company (ARCO) in 1977, constructed the mill and used the carbonate-leach mill to process uranium ore mined from the Todilto Limestone in the vicinity of the site (DOE 2016a; DOE 1997). The acid-leach mill was constructed to process sandstone uranium ore from the Jackpile-Paguate mine located north of the Laguna Pueblo (DOE 2016a). The Jackpile-Paguate mine was the largest open-pit uranium mine in North America at the time (DOE 2016a).

Decommissioning of the mill began in 1989 by ARCO, and site reclamation began in 1991 (DOE 2016a). By 1995, all mill tailings, contaminated soils, demolished mill structures, and contaminated vicinity property materials were encapsulated in a main tailings disposal cell, a carbonate tailings disposal cell, an asbestos disposal area, a disposal area that contains a polychlorinated biphenyl disposal cell, and two small landfills (DOE 2016a). The main tailings disposal cell contains more than 90% of the total tailings material, covers 354 acres, and contains an estimated 23 million tons of tailings and other contaminated materials (DOE 2016a).

The Bluewater site fact sheet and related site documents can be found at <https://www.energy.gov/lm/bluewater-new-mexico-disposal-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=BLU>.

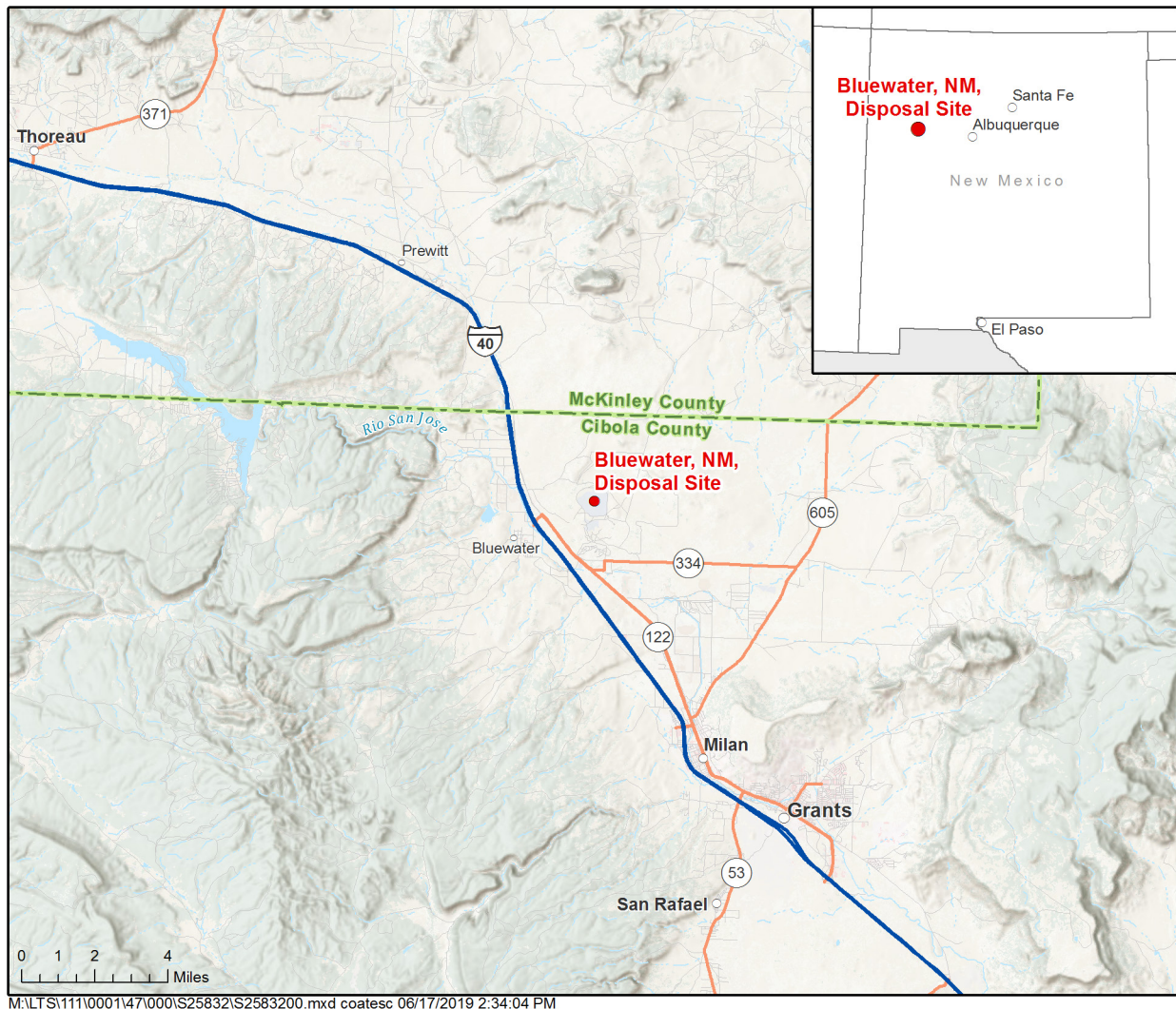


Figure 1. Site Location Map of the Bluewater, New Mexico, Disposal Site

2.1.1 Hydrogeology

The site occupies 3300 acres, one-third of which (the southern and western parts) is covered by basalt lava flows that filled a former river valley and covered alluvial sediments in the valley. Much of the rest of the site consists of sedimentary rock outcrops and large areas of wind-deposited fine-grained sediments (DOE 2016a). Groundwater occurs in the ancestral Rio San Jose alluvium and the San Andres Glorieta (SAG) aquifer.

The ancestral Rio San Jose alluvium was deposited by the ancestral Rio San Jose and cut into beds of the lower part of the Chinle Formation during the middle and late Pleistocene (DOE 2014a). The alluvium is not exposed at the site because it was buried by the Bluewater Basalt flows. The source of the Bluewater Basalt is a small shield volcano about 4 miles north of the main tailings disposal cell that erupted approximately 68,000 years ago (DOE 2014a). The thickness of the Bluewater Basalt at the site ranges from 70 to 130 feet. The ancestral Rio San Jose alluvium ranges in thickness from a few feet to approximately 25 feet and consists mostly of sand and gravel with coarse sand and gravel in the base of the channels and finer-grained

material (medium- and fine-grained sand and silt) in the shallower parts of channels and laterally away from channels (DOE 2014a). The ancestral Rio San Jose alluvium rests on the Chinle Formation, which is considered to be an aquitard and is not in direct connection with the SAG aquifer.

The San Andres Glorieta aquifer consists of the San Andres Limestone and the Glorieta Sandstone and is referred to as the SAG because these formations function as a single, prolific regional source of groundwater (DOE 2014a). The Lower Permian-aged San Andres Limestone was deposited in a marine environment and consists of limestone, dolomite, sandstone, and chert. The Lower Permian-aged Glorieta Sandstone was deposited in a marginal marine environment and consists of light gray, fine- to medium-grained sandstone with well-sorted, rounded to subrounded quartz grains. The Glorieta Sandstone does not crop out at the site but intertongues with the overlying San Andres Limestone.

Groundwater flow in the aquifer beneath the site is in the east-southeast direction toward the Homestake site and continues in an east-southeast to southeast direction toward areas north of Grants. The greatest amounts of groundwater within the SAG aquifer are transmitted in solution channels, cavernous zones, and fractures that have developed in the San Andres Limestone. Groundwater in the Glorieta Sandstone mainly flows through the porous matrix, with some component flowing in fractures within the sandstone.

2.1.2 Evidence of Persistent Secondary Uranium Sources

DOE 2014a identifies a persistent secondary uranium source as the dissolution of solid-phase uranium in a thick mineralized zone beneath the main tailings disposal cell (category 1). This mineralized zone was formed as the acidic tailings fluids were neutralized, as they seeped into underlying aquifers, causing contaminants to precipitate and adsorb to aquifer sediments, and likely extends more than 100 feet deep below the base elevation of the cells and laterally along fault zones (DOE 2014a). The dissolution of this continuous, constant-concentration source of uranium is expected to continue in the aquifer for decades to hundreds of years (DOE 2014a). However, as indicated in Table 17 of DOE 2014a on uncertainties of conclusions, “Sampling of materials under the disposal cells has never been conducted to confirm the presence or extent of the mineralized zone.”

A second persistent secondary uranium source at the Bluewater site is the occurrence of dual-domain transport processes (category 3) in the Rio San Jose alluvium and SAG aquifer (DOE 2014a). The coarse-grained sands and gravels in paleochannel portions of ancestral Rio San Jose alluvium act as preferential pathways for dissolved uranium, but a mixture of coarse- and fine-grained sediments occurs in the former overbank areas due to reworking of those sediments by river flows in years preceding the lava incursion. Low velocities of groundwater are expected in these floodplain deposits due to the presence of abundant silts and clays. Thus, the paleochannel is a preferential flow path conveying groundwater and contamination downstream at a high velocity, but the finer-grained deposits in non-channel parts of the alluvium retain and slowly release relatively immobile contamination to the preferential pathways (DOE 2014a). In the SAG aquifer, rapid groundwater velocities occur in the preferential pathways consisting of solution channels and fractures in limestone, and slower groundwater velocities occur in the adjacent limestone and sandstone rock matrixes. The exchange of dissolved contamination between the fractures and the matrix occurs through either

molecular diffusion in the matrix or very slow advection (DOE 2014a). The occurrence of dual-domain groundwater transport may cause uranium contamination to persist for many years in the SAG aquifer (DOE 2014a).

2.2 Fernald Preserve, Ohio, Site

The Fernald Preserve, Ohio, Site is on the site of the former Feed Materials Production Center in southwestern Ohio, approximately 18 miles northwest of Cincinnati (Figure 2). The site is just north of Fernald, Ohio, and lies on the boundary between Hamilton and Butler Counties. The Feed Materials Production Center processed uranium feed materials to produce high-purity uranium metal between 1951 and 1989. Processed uranium was then shipped to other DOE or U.S. Department of Defense facilities for use in the nation's nuclear weapons program (DOE 2016b). Uranium used in the reactors at the Hanford site in the state of Washington was recycled at this site, and it also served as the nation's key federal repository for thorium-related nuclear products (DOE 2016b). The recycled material was the source of technetium-99, a radiological contaminant that was prevalent at the Fernald site.

The facility produced nearly 500 million pounds of uranium metal products, and the production operations caused releases to the surrounding environment, which resulted in the contamination of soil, surface water, sediment, and groundwater on and around the site (DOE 2017a). In 1991, as a result of the contamination, the mission of the site changed from uranium production to environmental cleanup under CERCLA (DOE 2017a). Various operations at the site generated liquid and solid wastes between 1952 and 1989. Before 1984, solid and slurried processing wastes were deposited in the on-property Waste Storage Area. After 1984, wastes produced from operations were containerized for offsite disposal. "Contaminants from material processing and related activities were released into the environment through air emissions, wastewater discharges, storm water runoff, leaks, and spills" (DOE 2016b).

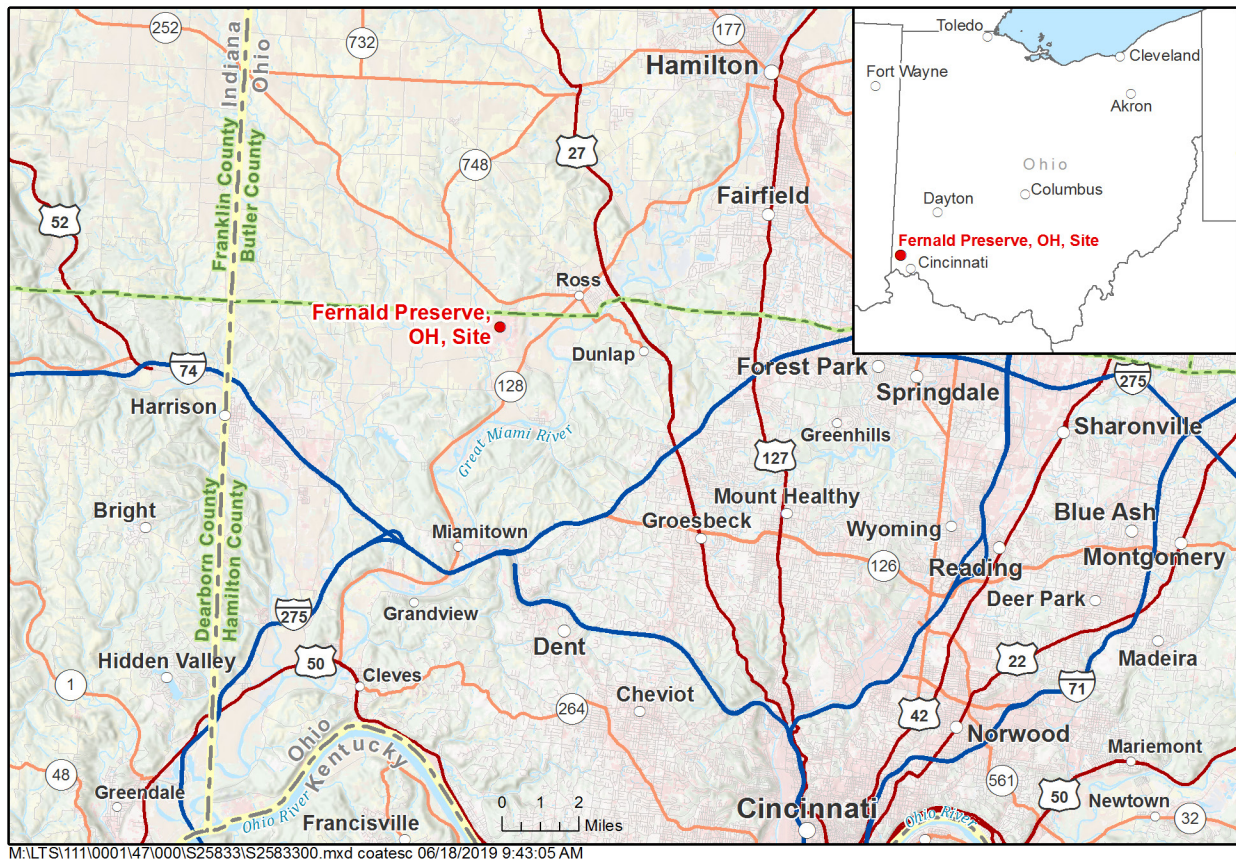


Figure 2. Site Location Map of the Fernald, Ohio, Site

Remediation activities generally occurred between 1986 and October 2006, which included the removal of contamination sources during soil remediation (DOE 2016b). Uranium is the principal contaminant of concern for the aquifer. As part of the remediation effort, the site was organized into five operable units (OUs) to organize site components by geographical location and by the potential for similar technologies to be used for environmental remediation (DOE 2016b). This review will address only OU 5, Environmental Media, which includes groundwater, surface water, and all soil not included in the definitions of OUs 1–4, sediment (DOE 2016b).

In 2007, the name of the site was changed to the Fernald Preserve to reflect the completion of site cleanup (with the exception of groundwater) and the fact that the site had been opened to the public as a nature preserve (DOE 2016b; DOE 2017a). The site restoration activities have made the Fernald Preserve attractive to a diversity of wildlife, including a large number of nesting and migrating birds and locally rare species (DOE 2017a).

The Fernald Preserve, Ohio, Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/ferald-preserve-ohio-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=FER>.

2.2.1 Hydrogeology

The Fernald Preserve is in the Great Miami River drainage basin. Drainage from the site to the Great Miami River occurs primarily via Paddys Run, an intermittent stream that flows from north to south along the site's western boundary (DOE 2017a). Paddys Run loses water to the underlying sand and gravel aquifer south of the former Waste Pits Area and empties into the Great Miami River 1.5 miles (2.4 kilometers [km]) south of the site.

The site is situated on glacial deposits consisting of a low-permeability mixture of clay and silt with minor amounts of sand and gravel that overlie the sand and gravel of the Great Miami Aquifer (DOE 2017a) (GMA). The site occupies a 2- to 3-mile wide (3–5 km) buried valley known as the New Haven Trough. The buried valley was created when river valleys up to 200 feet deep were filled with sand and gravel from melting glaciers. Groundwater is restricted to the sand and gravel within the buried valley by the impermeable shale and limestone bedrock that defines the edges and bottom of the New Haven Trough (DOE 2017a).

2.2.2 Evidence of Persistent Secondary Uranium Sources

Persistent secondary uranium sources mentioned in DOE 2016b are sorbed uranium in the unsaturated zone of the aquifer (category 1) and matrix diffusion from lower-permeability material (category 3). Uranium sorbed to soils in the unsaturated zone of the GMA beneath former contamination source areas is considered a contributing mechanism for prior modeling not predicting actual aquifer remediation cleanup times (DOE 2016b). No measured solid-phase data are provided in DOE 2016b, but DOE 2016b states that “Early indicators include rising uranium concentrations in groundwater beneath former source areas when water levels are high.” In addition, Table 1-4 in DOE 1998a is part of an excavation work plan that provides final remediation concentrations for uranium. These solid-phase concentrations range from 28.0 to 210 milligrams per kilogram (mg/kg) depending on the site area and sampled material (e.g., different operable units and soil or sediment). Background uranium concentrations are from 3.00 to 3.73 mg/kg (DOE 1998a).

Rate-limited mass transfer processes are thought to be occurring because the GMA is both heterogeneous and anisotropic. Indirect evidence that these processes are occurring is the asymptotic trend of decreasing uranium concentrations in the extraction wells and the relatively stable extent of the uranium plume (DOE 2016b). As stated in DOE 2016b, “coarser-grained aquifer material flushes contamination more effectively than the finer-grained aquifer material because more water is moving through the coarser material. Contamination sorbed to the finer-grained aquifer material slowly leaches out into the more active flow paths. Over time, this ineffective flushing of the finer-grained material results in reduced cleanup efficiency and prolonged cleanup times.”

2.3 Grand Junction, Colorado, Site

The Grand Junction, Colorado, Site is a Decontamination and Decommissioning program site near the city of Grand Junction, Colorado, about 26 miles from the border with Utah (Figure 3). The site is within an accretionary bend of the Gunnison River approximately 0.5 mile (800 meters) upstream from its confluence with the Colorado River (DOE 2006). The U.S. War Department operated a refinery onsite from 1943 to 1946 to concentrate uranium oxide for use

by the Manhattan Engineer District (DOE 2016c). The refinery produced an estimated 2.36 million pounds of uranium oxide and a comparable amount of vanadium oxide, which were shipped offsite for further processing (DOE 2016c). The U.S. Atomic Energy Commission (AEC) operated a uranium concentrate sampling plant and assay laboratory onsite until 1974. Beginning in 1953, AEC conducted a research program in a small pilot mill at the site to test experimental uranium-ore milling techniques, and in 1954, a larger pilot mill was constructed at the south end of the property. Approximately 30,000 tons of ore were processed before milling operations ceased in 1958.

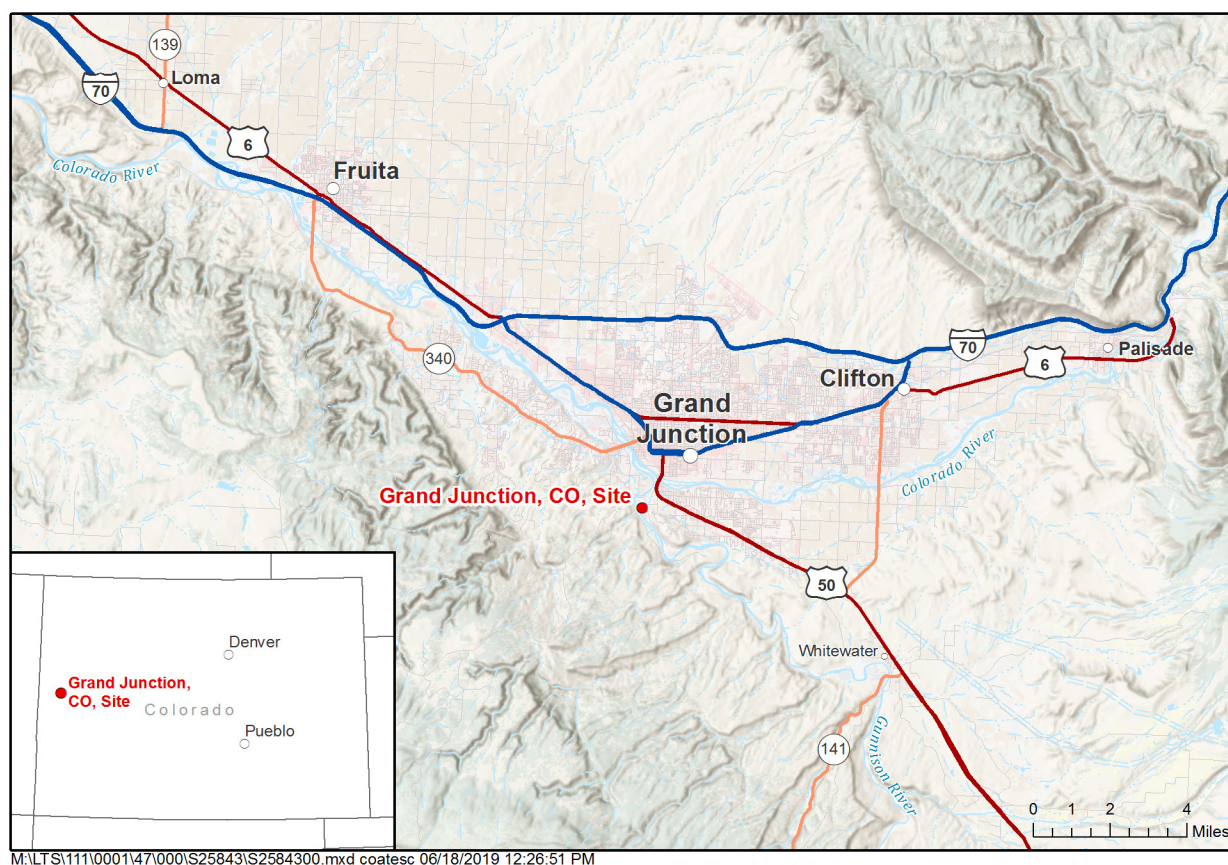


Figure 3. Site Location Map of the Grand Junction, Colorado, Site

Remediation began in 1989 and consisted of the removal of uranium mill tailings and contaminated soil, with most of the contaminated soil removed by 1994 (DOE 2016c). Other small deposits of contaminated soil and material were removed during remedial action conducted from 1998 through 2014 (DOE 2016c). The total volume of uranium mill tailings and tailings-contaminated material removed was approximately 256,340 cubic yards (DOE 2016c). The tailings and related materials occupied approximately 33.3 acres. The primary locations of remediation included the two onsite pond areas, areas located on the north and northwest of the property, and a dike along the Gunnison River (DOE 2016c).

The Grand Junction, Colorado, Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/grand-junction-colorado-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=GJO>.

2.3.1 Hydrogeology

Groundwater occurs in an unconfined alluvial aquifer (uppermost aquifer) beneath the site. The alluvial aquifer consists of two facies, a silty sand unit overlying a basal unit of poorly sorted, unconsolidated sands, and gravels that are laterally consistent across the site (DOE 2006). The Quaternary river alluvium is as much as 32 feet (9.8 meters) thick and was deposited on top of the Brushy Basin Member of the Morrison Formation (DOE 2006). The alluvial aquifer is bounded on the east by the silts, shales, and sandstones of the Brushy Basin member of the Morrison Formation and on the west and north by the Gunnison River. Depth to groundwater ranges from 5 to 10 feet (1.5–3 meters), and the general flow direction beneath the site is to the north (DOE 2006). The alluvial aquifer is in hydraulic connection with the Gunnison River (DOE 2006). Hydraulic conductivity within the aquifer ranges from 30 and 45 feet (9.1–13.7 meters) per day (DOE 2006). The Brushy Basin Member of the Morrison Formation acts as an aquitard beneath the alluvial sediments.

2.3.2 Evidence of Persistent Secondary Uranium Sources

In 2012, solid-phase sampling for uranium was completed at the Grand Junction site in an area where mill tailings had been removed (DOE 2017b). As stated in DOE 2017b, “The selective extraction data identify higher uranium concentration zones at the site that have secondary uranium deposited due to the uranium mill processes (1) above the water table (most common), (2) below the water table in limited areas and likely associated with gypsum, and (3) near and below the water table in association with organic carbon.” Detailed mineralogy work in DOE 2017b identified secondary uranium sources associated with mineral coatings and cements composed of iron oxyhydroxides and/or clays (items 1 and 2, category 1) and sorbed to the organic carbon (item 3, category 2). Column work indicated release of uranium up to 7 mg/L to the dissolved phase from these secondary uranium sources (DOE 2017b). Modeling of the column data evaluated dual porosity (category 3) as a mechanism that was needed for the column test data; however, simulations at slower groundwater flow rates did not show this as a major process. Column modeling did conclude that for item 3, uranium desorption influenced by gypsum dissolution was a key process in controlling uranium release (DOE 2017b).

Evidence of high organic carbon zones with higher solid-phase uranium concentrations is also documented in Noël et al. (2017). Solid-phase uranium information for the Grand Junction, Colorado, Site are provided as supplemental data with Noël et al. (2017).

2.4 Grand Junction, Colorado, Processing Site

The Grand Junction, Colorado, Processing Site, formerly known as the Climax Uranium Mill, is on the north bank of the Colorado River in Grand Junction, Colorado (Figure 4). The site was originally a sugar beet mill that began operations in 1899. In 1950, the Climax Uranium Company transformed the site into a uranium and vanadium mill that eventually processed more than 2 million tons of ore, producing about 12 million pounds of uranium oxide (U_3O_8) and 46 million pounds of vanadium oxide (V_2O_5), before it closed in 1971 (DOE 2002a; DOE 2017c). Ore was crushed, ground, salt roasted, and water leached to remove vanadium; uranium was extracted with sulfuric acid (DOE 2002a). The mill tailings were available to private citizens and contractors from 1950 to 1966 for use as fill material and as a component of concrete and mortar (DOE 2017c). Sampling of the tailings by the Colorado Department of

Public Health and Environment for radon-222 in 1966 indicated elevated levels and caused Climax to stop releasing the tailings from the site, but 300,000 tons of tailings had already been removed by 1966 (DOE 2017c).

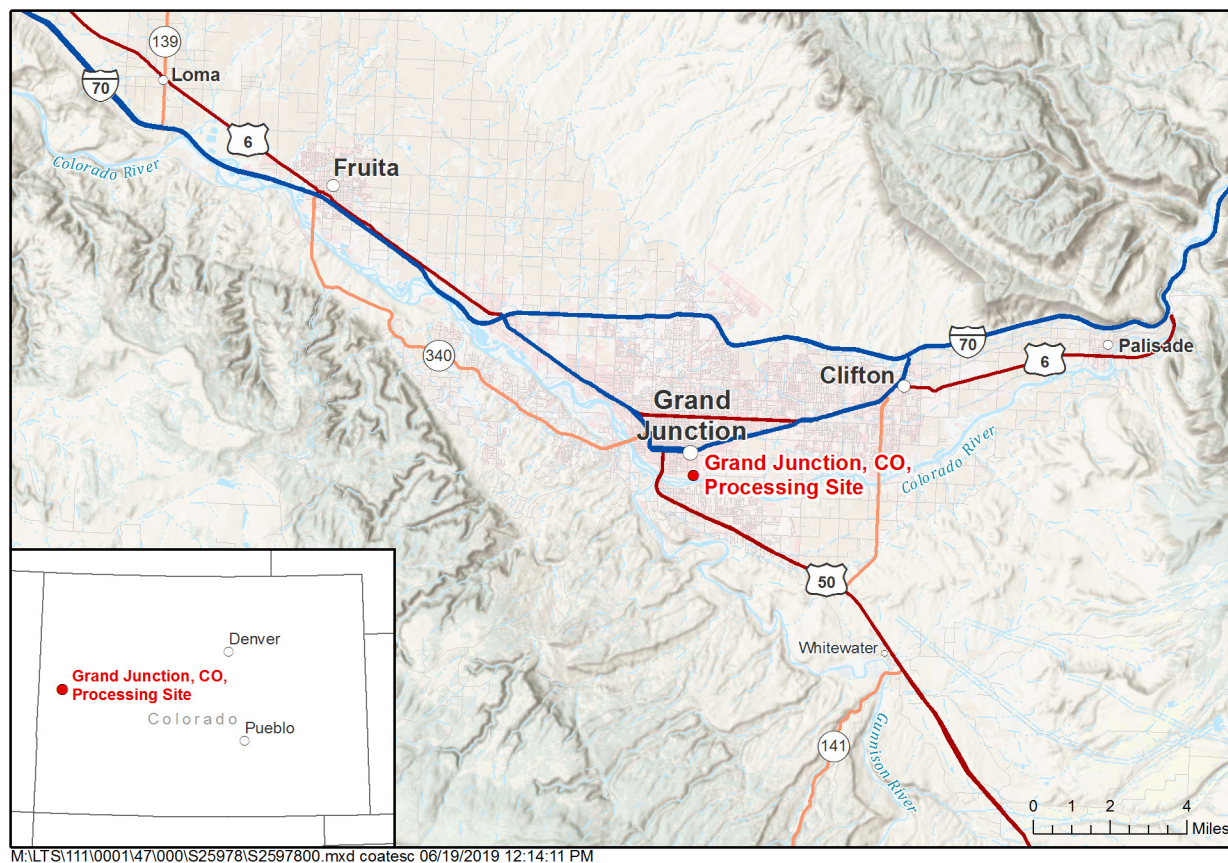


Figure 4. Site Location Map of the Grand Junction, Colorado, Processing Site

Surface remediation of the processing site and contaminated private and commercial properties (i.e., vicinity properties) began in the mid-1980s and initially consisted of storing tailings and other contaminated materials removed from vicinity properties temporarily at the processing site (DOE 2017c). By the end of 1994, approximately 4.4 million cubic yards of contaminated processing site materials, including vicinity property materials in temporary storage, were transported to the former Cheney disposal cell, now referred to as the Grand Junction, Colorado, Disposal Site, about 18 miles southeast of Grand Junction (DOE 2017c). DOE completed vicinity property cleanup in 1998 (DOE 2017c).

The Grand Junction, Colorado, Processing Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/grand-junction-colorado-disposal-and-processing-sites>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=GJT>.

2.4.1 Hydrogeology

Groundwater occurs in an unconfined alluvial aquifer and a confined aquifer in the sandstone members of the underlying Dakota Sandstone (DOE 2002a). The shale members of the

Dakota Sandstone act as an aquitard between the two aquifers. The alluvial aquifer is considered the uppermost aquifer at the site. Surface water at the site includes the Colorado River that flows along the southern border of the site and irrigation canals and drainage ditches north of the site (DOE 2002a).

The alluvial aquifer is composed of unconsolidated clays, silts, sands, gravels, and cobbles. Depths to water in the alluvium ranges from zero near the river to approximately 20 feet at the northern end of the site, with a saturated thickness of 5–20 feet (DOE 2002a). Groundwater generally flows southwest toward the Colorado River at a horizontal gradient of 0.004. Hydraulic conductivity ranges from 20 to more than 200 feet per day (DOE 2002a). Lateral and vertical facies changes typical of alluvial deposits drives the variability in conductivity. The average linear groundwater velocity is 2.0 feet per day based on an estimated average hydraulic conductivity of 100 feet per day, a hydraulic gradient of 0.004, and an effective porosity of 20% (DOE 2002a).

The depth to the top of the shale aquitard in the Dakota Sandstone ranges from less than 10 to 20 feet below ground surface (bgs) and may be as much as 50 feet thick (DOE 2002a). Wells completed in the shale member indicate that it is saturated with water, and the horizontal hydraulic conductivity is variable depending on the degree of weathering, but the lower end of the range for the unweathered material may be as low as 0.02 feet per day (DOE 2002a). An upward vertical hydraulic gradient is present, with a few exceptions during high water levels in the alluvial aquifer (DOE 2002a).

2.4.2 Evidence of Persistent Secondary Uranium Sources

Persistent secondary uranium sources at this site were evidenced by an analysis of subpile soils (category 1). The conclusion from the analysis was “mill-related radionuclides and non-radionuclides remaining in place after remediation to surface cleanup standards may pose a potentially unacceptable source of groundwater contamination” (DOE 1999a). Uranium concentrations in subpile soils are high enough and the K_d (distribution coefficient) value low enough that significant amounts of uranium could partition to groundwater (DOE 1999a). The maximum uranium concentration was 45.2 mg/kg, which is above background values of 0.44 to 1.1 mg/kg (DOE 1999a).

It is unknown if the subpile soils contained evaporite or precipitated uranium minerals. Evaporite minerals do occur along the floodplain of the Colorado River throughout the year and are more prevalent in dry months (DOE 2016d). Two evaporite samples contained thenardite (Na_2SO_4), blödite ($\text{Na}_2\text{Mg}(\text{SO}_4)\cdot 4(\text{H}_2\text{O})$), and watevillite ($\text{Na}_2\text{Ca}(\text{SO}_4)_2\cdot 4(\text{H}_2\text{O})$), and one sample contained halite (NaCl) (DOE 1999a). These samples were not analyzed for trace elements.

2.5 Green River, Utah, Disposal Site

The Green River, Utah, Disposal Site is a former upgrading facility for uranium ore approximately 1.5 miles southeast of the City of Green River in Grand County, Utah (Figure 5). The site is just south of Browns Wash, which is an ephemeral stream, and approximately 0.5 mile east of the Green River. The mill was constructed by Union Carbide Corporation in 1957 and processed 183,000 tons of uranium ore and generated an estimated 114,00 cubic yards of radioactive tailings from March 1958 through January 1961, with the ore concentrate shipped

to a uranium mill in Rifle, Colorado, for further processing (DOE 2002b; DOE 2017d). The State of Utah acquired ownership of the processing site property from Union Carbide in 1988, and DOE owns the disposal site (DOE 2017d).

The abandoned uranium mill tailings, all residual radioactive material, and contaminated material from 17 vicinity properties were stabilized in a disposal cell onsite from November 1988 through September 1989 (DOE 2017d). The disposal cell occupies 6 acres on the 21.5-acre site and contains about 382,000 cubic yards of contaminated material with a total activity of 30 curies of radium-226 (^{226}Ra) (DOE 2017d). The areas occupied by the former tailings pile and all areas disturbed during remedial action were backfilled, graded to promote surface drainage, and revegetated (DOE 2017d).

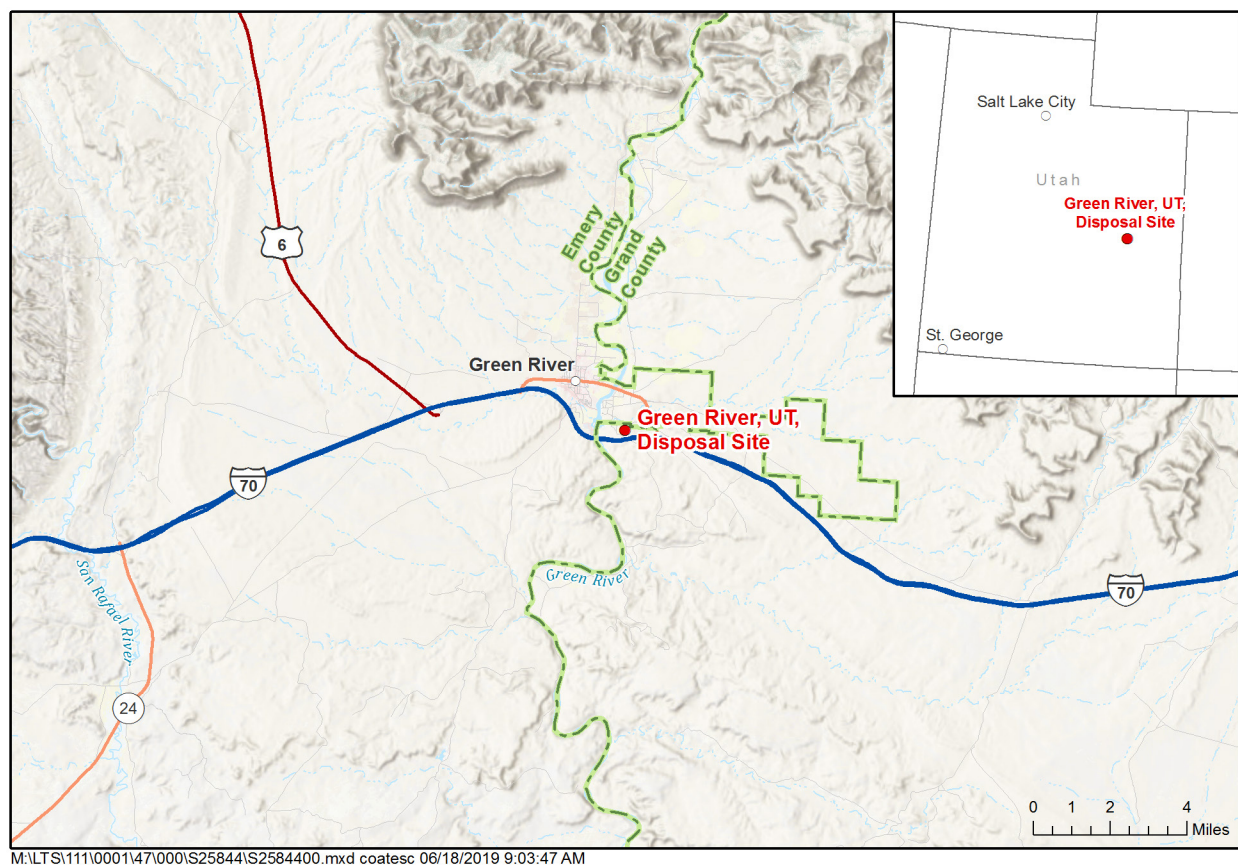


Figure 5. Site Location Map of the Green River, Utah, Disposal Site

The Green River, Utah, Disposal Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/green-river-utah-disposal-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=GRN>.

2.5.1 Hydrogeology

Groundwater occurs in the Browns Wash alluvium and in the middle sandstone unit of the Cedar Mountain Formation. The upper unit of the Cedar Mountain Formation acts as an aquitard between the Browns Wash alluvium and the middle sandstone unit of the Cedar Mountain

Formation (DOE 2002b). Bedrock underlying the site consists of Cretaceous age Mancos Shale, Dakota Sandstone, and the Cedar Mountain Formation and Jurassic age Brushy Basin member of the Morrison Formation.

The alluvium along Browns Wash extends approximately 400 feet on either side of it and varies in thickness from 0 to 35 feet. Alluvial material consists of a mixture of silt, sand, gravel, and some small cobbles. Shallow groundwater occurs in the Browns Wash alluvium under unconfined conditions and is limited by the lateral extent of the alluvium. Depth to groundwater varies from 8 to 17 feet below land surface (DOE 2002b). In July 2002, the saturated thickness of the Browns Wash alluvium was between 0 and 3 feet, with the maximum thickness near Browns Wash (DOE 2002b). The groundwater flow direction is to the southwest toward the Green River with an estimated seepage velocity that ranges from 1.2 to 2.3 feet per day (DOE 2002b).

The middle sandstone unit of the Cedar Mountain Formation consists of siltstone to coarse-grained sandstone with minor conglomerates and ranges in thickness from 15 to 40 feet (DOE 2002b). Groundwater occurs under confined to semiconfined conditions and is approximately 60 feet bgs but has fluctuated over 5 feet since 1991 (DOE 2002b). Groundwater flows from the northeast to the southwest with the hydraulic conductivity range from 0.09 to 3.1 feet per day (DOE 2002b).

2.5.2 Evidence of Persistent Secondary Uranium Sources

The formation and subsequent dissolution of evaporite minerals (category 2) is a potential persistent secondary uranium source at this site. Browns Wash is considered a groundwater discharge point with little surficial drainage and mainly discharges through evapotranspiration (DOE 2002b). Evaporite deposits have been observed along the stream bank and channel of Browns Wash (DOE 2016d). Sediment samples collected in Browns Wash in 1993, 1994, and 1995 had a maximum uranium concentration of 27.7 mg/kg in September 1993 (DOE 2002b). The mineralogy of the evaporite deposits is not known, but they are assumed to be composed of sodium sulfates like other sites with areas of natural evaporite formation (DOE 2016d). Because of the occurrence of the Mancos Shale in the area, which likely contributes high concentrations of dissolved constituents to the groundwater, it is not clear if some of the uranium in the evaporites is naturally occurring. Background solid-phase uranium concentrations in the evaporites are not available (DOE 2002b).

2.6 Gunnison, Colorado, Processing Site

The Gunnison, Colorado, Processing Site is a former uranium-ore processing site occupying 61.5 acres of land adjacent to the Gunnison County airport, one-half mile southwest of the city of Gunnison, Colorado (Figure 6). The site is on an alluvial terrace between the Gunnison River and Tomichi Creek. The mill operated from 1958 to 1962 and processed approximately 540,000 tons of uranium ore that was mined from along the Los Ochos fault near Cochetopa Pass approximately 25 miles southeast of Gunnison (DOE 2001; DOE 2017e).

Contaminated material above the water table and 1 foot below the water table was removed during the surface cleanup from 1992 to 1995. High thorium-230 concentrations were detected in sediments below the water table, so supplemental standards were applied to thorium-230 that

was left in place on the processing site (DOE 2001; DOE 2017e). Most tailings and other contaminated materials were transported to the Gunnison disposal site about 6 miles east of Gunnison and 0.4 mile south of the county solid-waste landfill (DOE 2001). In 1995, DOE completed construction of the disposal cell that covers 29 acres.

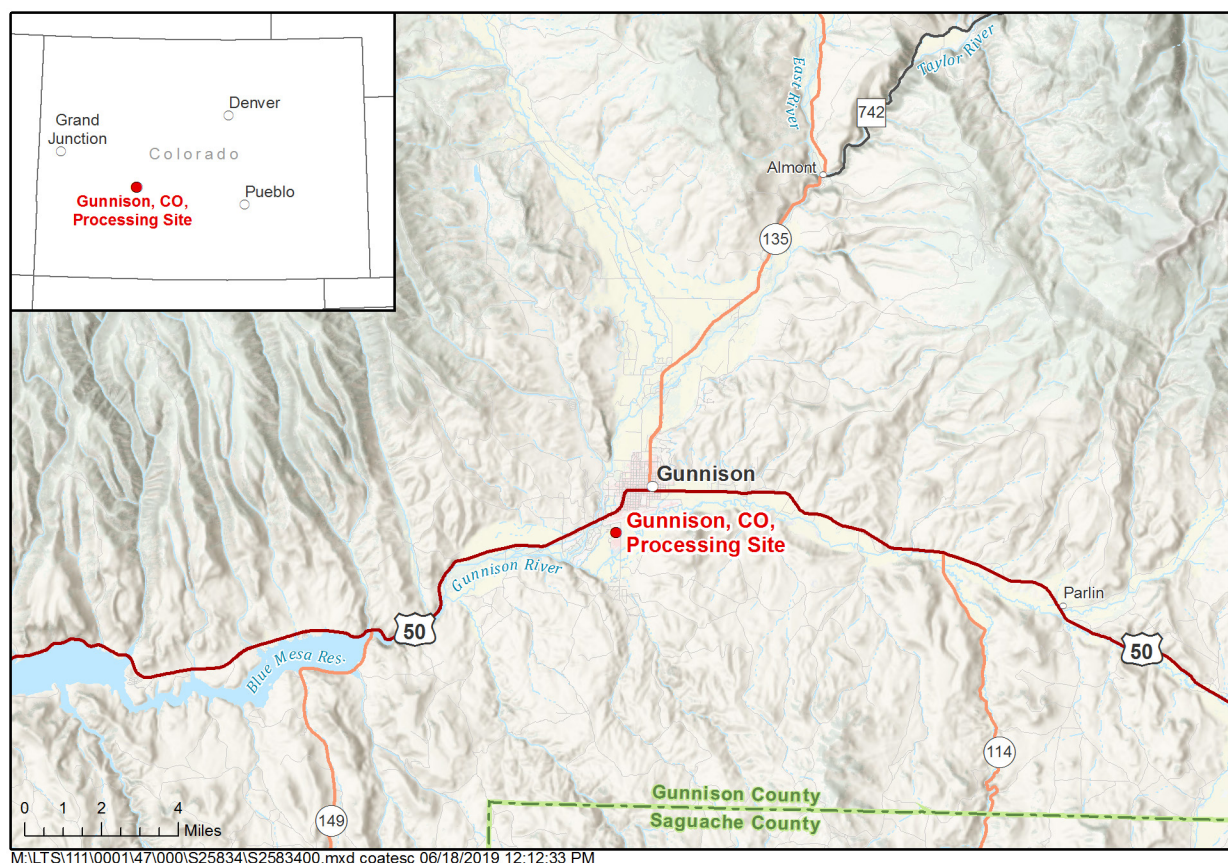


Figure 6. Site Location Map of the Gunnison, Colorado, Processing Site

The Gunnison, Colorado, Processing Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/gunnison-colorado-disposal-and-processing-sites>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=GUP>.

2.6.1 Hydrogeology

Groundwater occurs under unconfined conditions in the alluvial aquifer beneath and downgradient from the site (DOE 2001). The thickness of the alluvium ranges from 70 to 130 feet, and it is composed of poorly sorted sediments that range in size from clay-sized particles to gravel with cobbles and occasional boulders (DOE 2001). The depth to the top of the water table ranges from 2 to 11 feet and may fluctuate more than 10 feet over the course of a year, with highest water levels occurring in the spring and summer (DOE 2001). Groundwater flows to the southwest with an average gradient of 0.005, and hydraulic conductivity ranges from 100 to 170 feet per day (DOE 2002c). The average groundwater velocity, based on a hydraulic gradient of 0.005 and an estimated effective porosity of 0.27, ranges from 1.9 to 3.2 feet per day (DOE 2002c).

2.6.2 Evidence of Persistent Secondary Uranium Sources

Subpile soils (category 1) at the Gunnison site have elevated uranium concentrations and are considered a likely continuing source of groundwater contamination (DOE 2001). Because of high thorium-230 concentrations detected in sediments below the water table during remediation from 1992 to 1995, DOE collected soil samples within and adjacent to the former mill site from November 2–9, 1999. Uranium concentrations in subpile soils within the former mill site are up to 400 times higher than background concentrations (DOE 2001). Two samples with the highest uranium concentrations (locations 545 and 546) were leached with groundwater from background well 0002 in a column test. The initial uranium concentration in the effluent of the 545 column was about 1.50 mg/L and reached a maximum of 1.67 mg/L after about 30 pore volumes (DOE 2001). After 115 pore volumes, the concentration of uranium was 0.296 mg/L. The effluent from the 546 column had lower uranium concentrations than those from the 545 column despite having similar concentrations of uranium in the soil. The first effluent of the location 546 column had a uranium concentration of 0.2 mg/L which decreased to 0.067 mg/L after 26 pore volumes. Groundwater at some onsite monitoring wells had uranium concentrations of about 1 mg/L for several years. Thus, DOE 2001 concludes that soils similar to the 545-location sample, with uranium release over a long period of time, might be the most common type of soil present onsite (DOE 2001).

2.7 Monticello, Utah, Disposal and Processing Sites

The Monticello, Utah, Disposal and Processing Sites are in and near the city of Monticello, Utah, about 250 miles southeast of Salt Lake City (Figure 7). The former mill site (Monticello processing site) occupies 110 acres within the valley of Montezuma Creek. The mill processed approximately 900,000 tons of uranium and vanadium ore from 1942 to 1960 (DOE 2017f). Milling operations generated 2.5 million cubic yards of tailings that were impounded onsite in piles at four locations adjacent to Montezuma Creek (DOE 2016e). Some of the tailings were commonly used in Monticello as fill for open lands; backfill around water, sewer, and electrical lines; sub-base for driveways, sidewalks, and concrete slabs; backfill against basement foundations; and as sand mix in concrete, plaster, and mortar (DOE 2007; DOE 2017f).

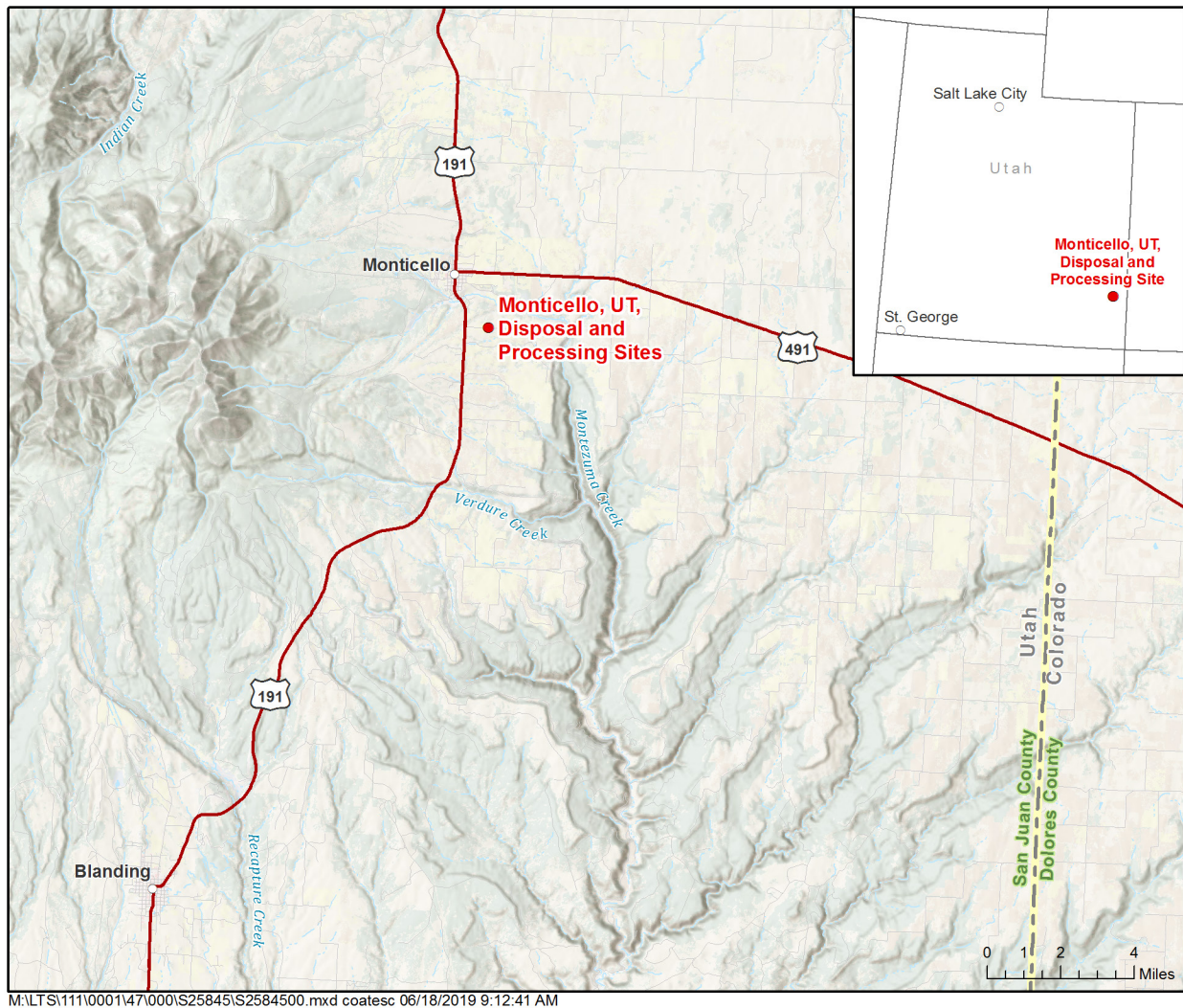


Figure 7. Site Location Map of the Monticello, Utah, Disposal and Processing Sites

Tailings and other contaminated materials from the processing site and in Monticello were transferred to an engineered disposal cell (Monticello disposal site) approximately 1 mile south of the processing site by June 2000 (DOE 2017f). The disposal cell contains a base liner system that collects water that remained in the waste when it was encapsulated. The water that drains from the waste material is transferred to an adjacent engineered solar evaporation pond (DOE 2017f). Supplemental standards were applied to some areas around the processing site and in the city of Monticello where attempts to reach cleanup standards greatly increased the risk of human injury, could cause excessive harm to the environment, or the cost of cleanup was unreasonably high compared to the long-term benefits to human health and the environment (DOE 2017f).

The Monticello, Utah, Disposal and Processing Sites fact sheet and related site documents can be found at <https://www.energy.gov/lm/monticello-utah-disposal-and-processing-sites>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=MNT>.

2.7.1 Hydrogeology

Groundwater at the Monticello processing site occurs in a shallow alluvial aquifer, which is composed of silty sand and gravel channel fill deposits within the valley of Montezuma Creek, and the underlying Burro Canyon Formation (DOE 2007). The Dakota Sandstone is between these two aquifers and acts as an aquitard. Groundwater contamination occurs only in the alluvial aquifer (DOE 2007). Depth to bedrock is generally within 15 feet of ground surface within the valley floor, and the average saturated thickness of the aquifer is about 5 feet (DOE 2007). Groundwater within the alluvial aquifer flows from west to east along the gradient of the valley of Montezuma Creek (DOE 2007).

2.7.2 Evidence of Persistent Secondary Uranium Sources

Solid-phase uranium data are summarized in DOE 2019. Persistent secondary uranium sources identified in DOE 2019 include:

- Residual uranium below former uranium tailings impoundment, waste ponds, and ore storage areas that remains in the unsaturated zone (up to 54 mg/kg, category 1).
- Uranium in the unsaturated zone above the uranium plume emplaced by seasonal changes in groundwater levels and evaporite deposition through evapotranspiration (up to 10 mg/kg, category 2).

Prior reports indicate background solid-phase uranium concentrations of below 3.2 mg/kg (DOE 2019). New data in DOE 2019 indicate background uranium concentrations as low as 1.4 mg/kg. In DOE 2019 and in prior reports referenced there, the possibility of precipitated uranium/vanadium and iron/vanadium minerals is indicated through indirect data, but such minerals were not directly identified. Column data in DOE 2019 quantify the release rates and concentrations of uranium that occur above the uranium standard of 30 micrograms per liter ($\mu\text{g/L}$) for the above cases for over 10 pore volumes. The transport of these persistent secondary uranium sources to the saturated zone has not been quantified but is part of ongoing reactive transport modeling efforts (DOE 2019).

2.8 Monument Valley, Arizona, Processing Site

The Monument Valley, Arizona, Processing Site is located in the Navajo Nation in northeastern Arizona, approximately 15 miles south of Mexican Hat, Utah (Figure 8), on the west side of Cane Valley. A uranium-ore processing mill operated at the site from 1955 to 1968 on property leased from the Navajo Nation. The mill closed in 1968, and control of the site reverted to the Navajo Nation. Most of the mill buildings were removed shortly thereafter. The uranium ore processed at the site was discovered in 1942 approximately 0.5 mile west of the former mill site (DOE 2005). A total of 767,166 tons of uranium and vanadium ore was mined from the deposit between 1943 and 1968 (DOE 2005).

Ore at the site was processed by mechanical milling from 1955 until 1964 using an upgrader, which crushed the ore and separated it by grain size, which used only minor amounts of flocculent (a substance used to consolidate particles within a liquid) (DOE 2005). The finer-grained material, which was higher in uranium content, was shipped to other mills for chemical processing. Coarser-grained materials were stored on site in the “old tailings pile.”

Some groundwater contamination may have resulted from water draining through the tailings piles during that period (DOE 2005).

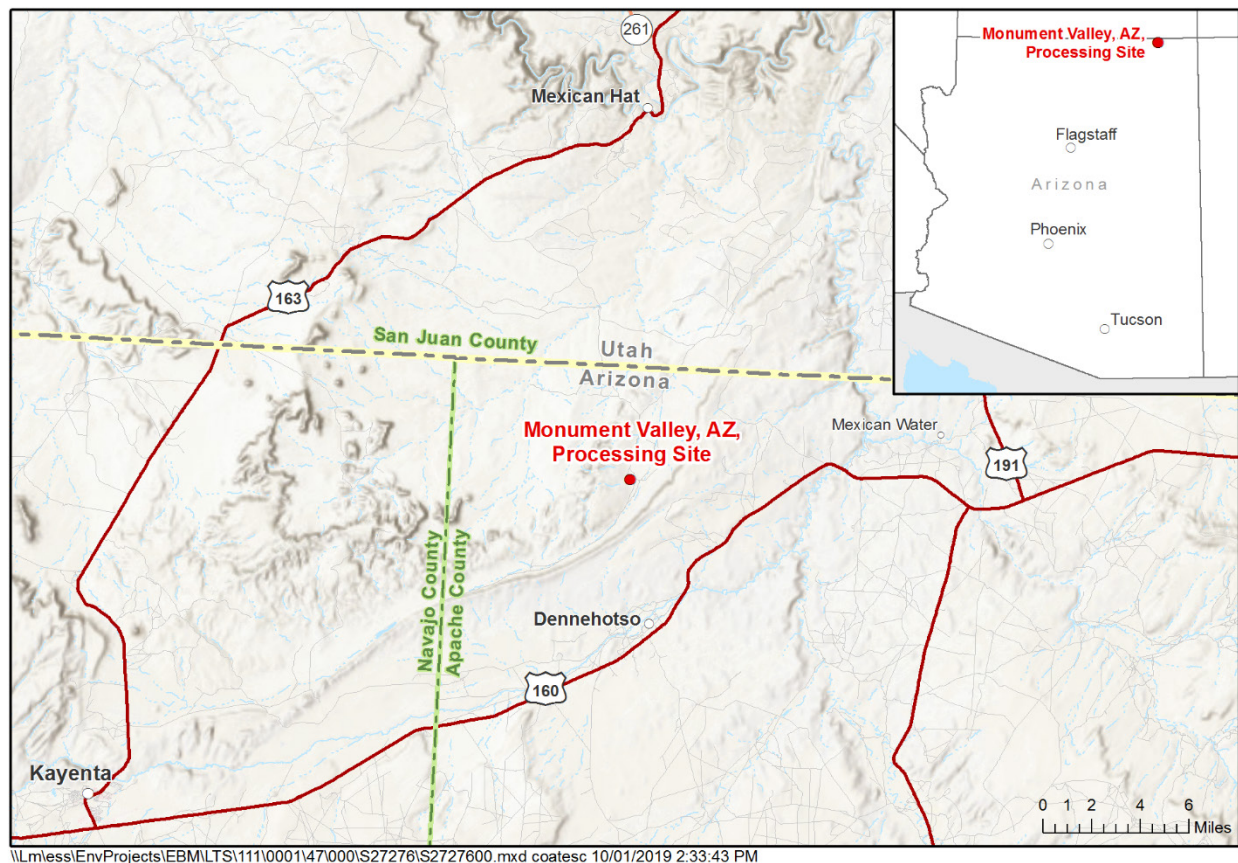


Figure 8. Site Location Map of the Monument Valley, Arizona, Processing Site

Batch leaching and heap leaching were used from 1964 until 1968 to process an estimated 1.1 million tons of tailings and low-grade ore at the site (DOE 2005). In the batch-leaching process, sandy tailings were placed in lined steel tanks, and uranium and vanadium were leached by an upward flow of sulfuric acid solution. Heap leaching consisted of placing crushed, low-grade ore on polyethylene sheeting and percolating a sulfuric acid solution through the ore. Chemical solutions used in ore processing are believed to have been discharged to the “new tailings pile.” An evaporation pond was on the east side of the new tailings pile (DOE 2005).

Source materials and other site-related contamination were removed during surface remediation at the site from 1992 through 1994. All contaminated materials from the Monument Valley processing site were transported north (approximately 15 miles) and encapsulated in the Mexican Hat, Utah, disposal cell (DOE 2017g).

The Monument Valley, Arizona, Processing Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/monument-valley-arizona-processing-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=MON>.

2.8.1 Hydrogeology

The Monument Valley site is in Cane Valley and is drained by Cane Valley Wash (DOE 1999b). Underlying the site is unconsolidated material of Quaternary age that consists of dune sand, alluvial material (sand and gravel), and fine-grained sediments that are probably lake bed deposits (clay or sandy clay) (DOE 1999b). Bedrock beneath the Quaternary alluvium consists of several members of the Triassic Chinle Formation, the Triassic Moenkopi Formation, and the Permian Cutler Formation.

The three main aquifers onsite are the alluvial, Shinarump, and De Chelly aquifers, with the Shinarump and De Chelly separated by the Moenkopi Formation and its lowermost Hoskinnini Member (DOE 1999b). The western portion of the former old tailings area is underlain by a buried paleovalley approximately 120 feet deep that has eroded the Shinarump Member of the Chinle Formation and the Upper Moenkopi Formation. At this location, Quaternary material rests on the Hoskinnini Member of the lower Moenkopi Formation and the medium- to coarse-grained Hoskinnini sandstone provides a hydrologic connection between the alluvial and the underlying De Chelly aquifer (DOE 1999b).

The alluvial aquifer is composed of Quaternary eolian deposits consisting mainly of fine- to medium-grained ranging in thickness from 0 to 120 feet (DOE 1999b). The alluvial aquifer is predominantly unconfined and is underlain by the unconfined and leaky confined Shinarump aquifer. The main confining unit is the Upper Moenkopi, which overlies the leaky confined Hoskinnini and De Chelly aquifers. The Hoskinnini and De Chelly appear to be hydrologically connected and are described as a single unit in some of the earlier boring logs.

Alluvial groundwater generally flows north in the site vicinity, and groundwater velocity ranges from 0.6 to 1.0 foot per day (DOE 1999b). Recharge to the alluvial aquifer is from infiltration of precipitation and from upward leakage from the underlying aquifers (DOE 1999b). The area receives approximately 6.4 inches of precipitation annually, with the majority of the precipitation resulting from isolated thunderstorms during the late summer and early fall. Discharge from the alluvial aquifer is primarily the result of evapotranspiration. An estimated 1.6 inches of the annual 6.4-inch total is available for recharge and runoff on a yearly basis (DOE 1999b).

2.8.2 Evidence of Persistent Secondary Uranium Sources

The Monument Valley site was identified as having evaporites/mineral precipitates within a former evaporation pond area (classified as category 1 only, since this occurs only at the former evaporation pond area). Measured uranium concentrations are available from multiple dates and reports, which are summarized in DOE 2016d. Several solid-phase uranium concentrations were in the range of 400–450 mg/kg with a maximum value of 1500 mg/kg (DOE 2016d). The surficial soils at the former evaporation pond also had high vanadium concentrations. The formation of this persistent secondary uranium source is summarized in DOE 2016d as:

The surficial discolored soils at Monument Valley are unique, as these materials are probably not a direct result of the evaporation of shallow groundwater. The high vanadium content suggests that the soils are uranium/vanadium minerals such as carnotite ($K_2(UO_2)_2(VO_4)_2 \cdot 3(H_2O)$) or tyuyamunite ($Ca(UO_2)_2V_2O_8 \cdot (5-8)H_2O$) that form because of their low solubility in water when high vanadium

concentrations are present. No mineral identification has been completed on these samples to date, but evaporative concentration at the surface would assist in the formation of these minerals. The formation of these low-solubility minerals can maintain high uranium/vanadium concentrations in the soils but limit their mobility in groundwater. Thus, this is a possible reason for the observation of low uranium and vanadium concentrations in groundwater below the former evaporation pond.

2.9 Naturita, Colorado, Processing Site

The Naturita, Colorado, Processing Site is a former uranium- and vanadium-ore processing mill that operated intermittently between the 1930s and 1958 (DOE 2016f). The site occupies 79 acres on the floodplain of the San Miguel River about 2 miles northwest of the city of Naturita in Montrose County, Colorado (DOE 2016f) (Figure 9). The ore processed at the mill was mined predominantly from the Salt Wash Member of the Morrison Formation from the Uravan Mineral belt (DOE 2002d). Carnotite and tyuyamunite were the primary uranium and vanadium minerals (DOE 2002d). The mill processed approximately 704,000 tons of ore, which produced 24 acres of tailings on site. The tailings pile was purchased in 1976 and moved off-site for additional processing, leaving approximately 126 acres of soil around the tailings pile contaminated from ore storage and windblown tailings (DOE 2016f).

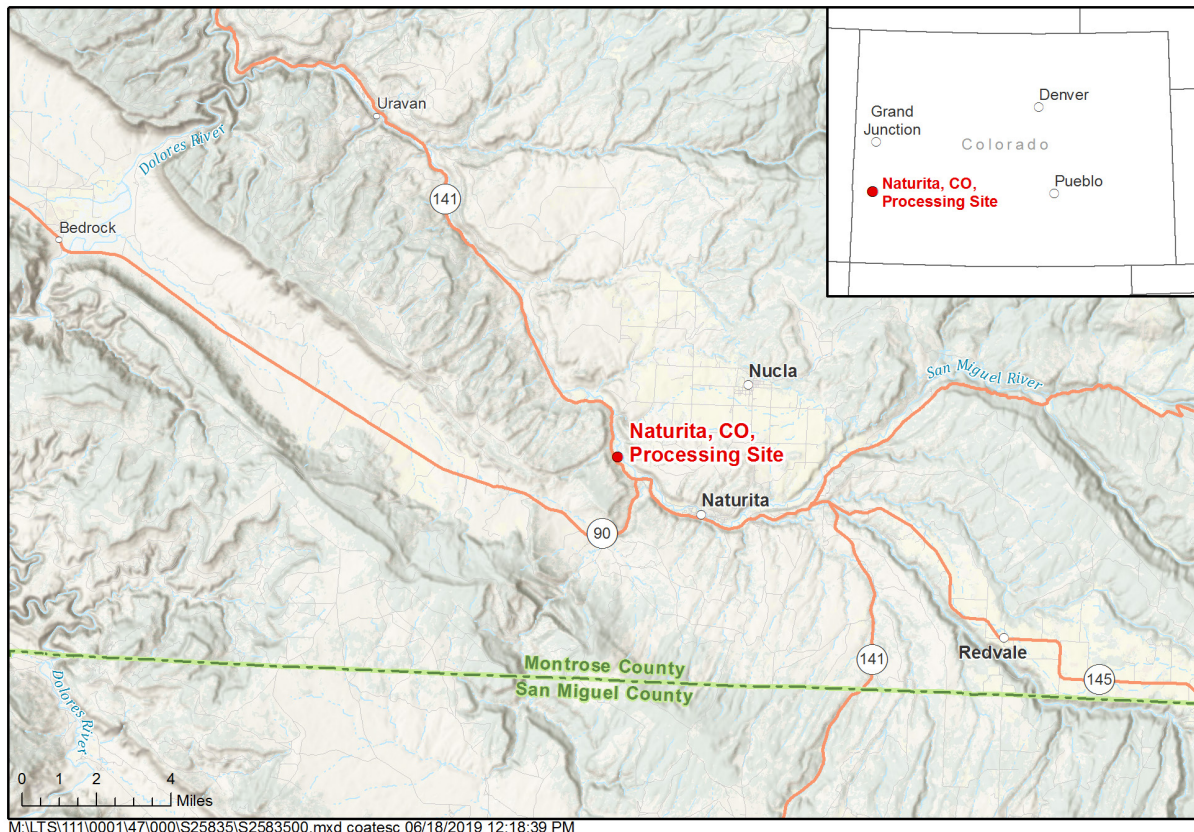


Figure 9. Site Location Map of the Naturita, Colorado, Processing Site

Remediation of the site occurred from 1993 to 1997 and resulted in the removal of 793,000 cubic yards of contaminated soil and other contaminated materials from the site. This material was

stabilized in an engineered disposal cell (Naturita, Colorado, Disposal Site) near the former town of Uravan, Colorado, 15 miles northwest of Naturita (DOE 2016f). The cell has produced no detectable soil or groundwater contamination (DOE 2016f).

Not all of the contaminated material was removed from the site during the surface cleanup. Some contaminated material was left in place in five areas of supplemental standards covering 11 acres and in another 11 acres on adjacent downgradient property (DOE 2002d; DOE 2016f). More than 1 acre of contaminated soil was left in place because the ^{226}Ra concentration standard was exceeded even though soil had been excavated to 1 foot below the water table. The other contaminated areas on the site and on the downgradient property were left in place because compared with the existing low radiological hazard, removing the material would have produced excessive environmental harm and increased risk to workers who would have had to remove it (DOE 2016f). On the downgradient floodplain property, contamination was left on approximately 5 of the 11 acres because the property owner did not want the area disturbed, and the harm to the environment outweighed the benefit of removing it (DOE 2002d).

The Naturita, Colorado, Processing Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/naturita-colorado-disposal-and-processing-sites>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=NAP>.

2.9.1 Hydrogeology

Groundwater at the Naturita site occurs primarily within a surficial aquifer composed of alluvium that was deposited by the San Miguel River (DOE 2002d). The alluvium covers the valley floor and is composed of clayey gravel to sandy cobbles, interbedded with layers of silty to sandy clay that pinch out against the bedrock on the west side of the site (DOE 2002d). Bedrock underlying the alluvium consists of the Brushy Basin and Salt Wash Members of the Jurassic Morrison Formation that are up to 820 feet thick in the Naturita area (DOE 2002d).

The alluvial aquifer varies from 5 to 23 feet in thickness along the San Miguel River over most of the site. Estimates of hydraulic conductivity were made from falling-head slug tests in 13 wells in November 1999 and May 2000 (DOE 2003). Values of hydraulic conductivity ranged from 19 (MAU-04) to 333 feet per day (NAT-09).

2.9.2 Evidence of Persistent Secondary Uranium Sources

As stated in DOE 2002d, “Contaminated soils and surface materials were removed from the site in 1997, and the excavated areas were backfilled with clean, clay-sized material. At that point, no radioactive materials were left. However, it is possible that contaminants have leached into the soils below the depth of remediation. These soils could contaminate infiltrating ground water and prolong the cleanup effort.” A 5% nitric acid leach of one onsite sample indicated a background value of 1.0 mg/kg uranium. The same extraction technique on two contaminated subpile soil samples in the former tailings area had values of 6.0 and 6.5 mg/kg uranium (category 1). As summarized in DOE 2002d, “This indicates that significant leachable uranium is still present in subpile soils and could represent a future source of uranium contamination.”

Zones that are high in organic carbon are not discussed in the DOE literature but are identified as occurring at the site in Noël et al. (2017). These high organic carbon zones can concentrate

uranium due to stronger sorption characteristics. Often, these high organic carbon zones are found near or below the groundwater table; thus, microbial degradation of the organic carbon zone consumes oxygen in a groundwater environment that is already low in oxygen. These high organic carbon zones are typically referred to as naturally reduced zones (NRZs). Solid-phase uranium data in these NRZs (category 2) for the Naturita, Colorado, Processing Site are provided as supplemental data with Noël et al. (2017).

2.10 Rifle, Colorado, Old Processing Site

The Rifle, Colorado, Old Processing Site is the location of a former uranium- and vanadium-ore processing mill that operated from 1924 to 1932 and from 1942 to 1958 and was idle in the interim (DOE 2017h). The site occupies 22 acres of a crescent-shaped floodplain in an erosional meander of the ancestral Colorado River approximately 0.3 mile east of the city of Rifle, Colorado (Figure 10). Vanadium and uranium ores were stockpiled in the east end of the site, and the tailings produced by the milling were placed across a large area that covered most of the site's west half. Ponds designed to contain mill-process waters were often constructed over large portions of the former mill tailings area. Most of the tailings were moved to the New Rifle site after 1958 for reprocessing.

Surface remediation at the Old Rifle site started in spring 1992 and was completed in October 1996 (DOE 2017h). This resulted in the removal and offsite disposal of mill tailings and contaminated soils from most of the site to meet the 40 CFR 192 activity standard of 5 picocuries per gram (pCi/g) for ^{226}Ra averaged over the first 15 centimeters of soil below the surface (DOE 2017h). Removal of soils and alluvial sediments generally stopped at the water table due to difficulties in excavating below this level; therefore, some soils exceeding the ^{226}Ra standard were left in place (DOE 2017i). Fine-grained soils from offsite were imported and used as fill to replace contaminated sediments that had been relocated. The fill was applied across most of the site and comprises the uppermost 5–10 feet of local surficial material.

In addition, an estimated 24,000 cubic yards of tailings qualified for supplemental standards because of concerns about worker safety and were left in place on adjoining vicinity properties north of the site, beneath U.S. Highway 6 & 24, and beneath the embankment on the highway's south side. Other supplemental standards areas exist under the railroad right-of-way and along the riverbank south of the site. Residual solid-phase activity concentrations for ^{226}Ra in the supplemental standards areas ranged as high as 1320 pCi/g and averaged about 150 pCi/g (DOE 2017h).

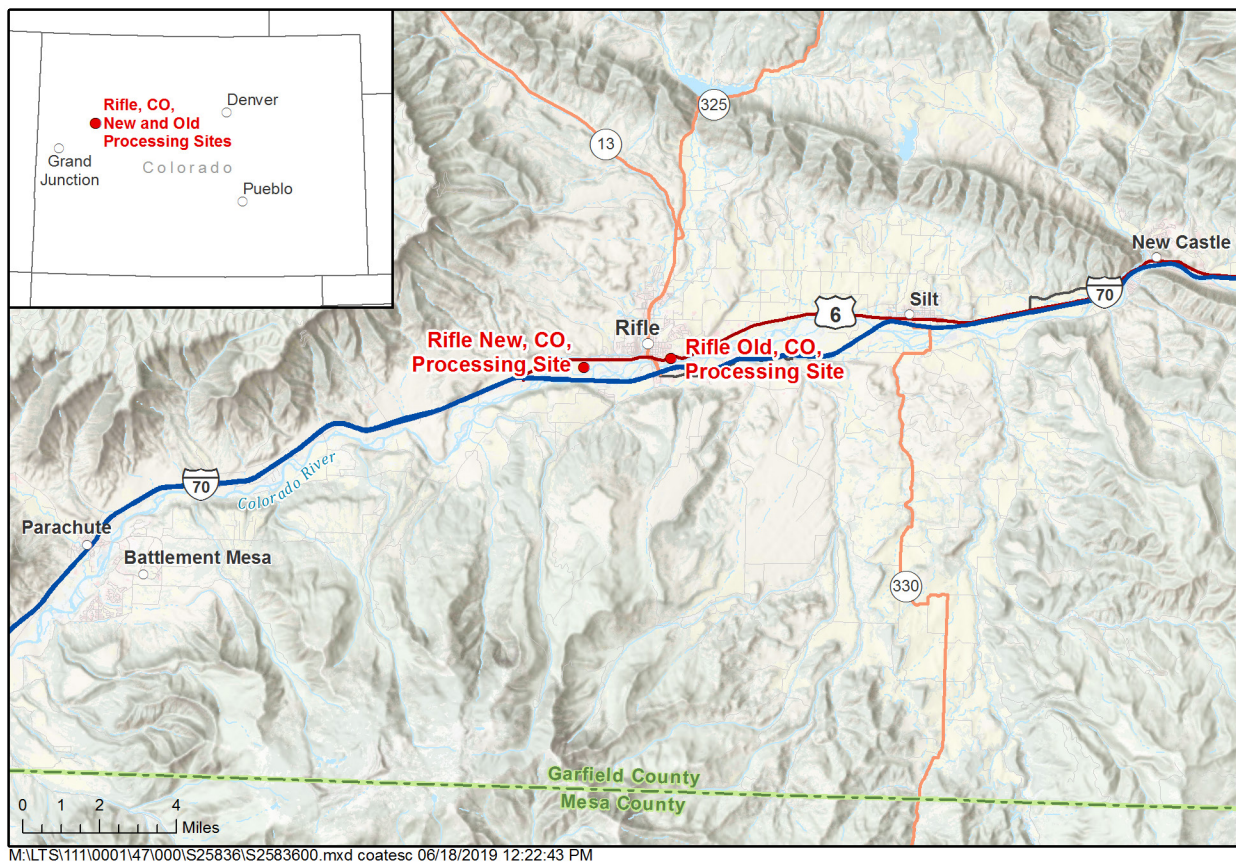


Figure 10. Site Location Map of the Rifle, Colorado, Old Processing Site

The Rifle, Colorado, Old Processing Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/rifle-colorado-disposal-site-and-processing-sites>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=RFO>.

2.10.1 Hydrogeology

Groundwater at the Old Rifle site occurs primarily within a surficial aquifer composed of Holocene-age alluvium that was deposited by the ancestral Colorado River (DOE 2011). The alluvium consists mostly of sandy gravels and gravelly sands interspersed with silts and clays. Sediments described as clayey gravels, silty gravels, and sandy silts are also reported in logs for boreholes drilled into the aquifer. The fine-grained fill that was used to replace contaminated alluvium that was excavated is typically described as silty sand and silt that also contains fine-grained sands, subrounded gravels, clays, and roots. Also present within the alluvium are NRZs near the Colorado River. An outcrop of the Wasatch Formation south of Highway 6 forms the north boundary of the aquifer. The Colorado River forms the south boundary of the aquifer, and the aquifer's east and west ends are located in areas where the outcropping Wasatch Formation abuts the river, effectively pinching the alluvium out. The areal extent of the alluvial aquifer takes up most of the area contained within the site's property boundary.

The alluvial aquifer is underlain by the Tertiary Wasatch Formation consisting mostly of erosion-resistant variegated claystone, siltstone, and sandstone. Depth to the top of the Wasatch

from land surface (i.e., from the top of the fill where present) typically varies between 20 and 30 feet. The alluvium directly overlies an 8–13 feet thick zone of weathered Wasatch Formation claystone that appears to be hydraulically connected to the unconsolidated alluvium. The alluvial aquifer maintains a relatively uniform thickness of approximately 20–25 feet (DOE 1999c). Depth to groundwater ranges from 5 to 15 feet bgs.

2.10.2 Evidence of Persistent Secondary Uranium Sources

Evidence of persistent secondary uranium sources at the Old Rifle site discussed in DOE 2011 includes (1) slow diffusion of uranium from low-permeability zones (category 3), and (2) mobilization of uranium from the unsaturated zone below the former tailings and ore storage areas (category 1). Diffusion of uranium from low-permeability zones was discussed as a process in DOE 2011 based on observed heterogeneity in lithologic logs during well drilling, but additional field-scale data for the evaluation of heterogeneity are not provided. DOE 2011 refers to DOE 1999c for former tailings and ore storage area subpile soil analyses. As stated in DOE 1999c, “All radioactive tailings and material were removed from the Old Rifle site in 1996 as part of the surface remediation program... However, the potential exists for nonradionuclide contaminants to have seeped into the soils below the limit of remediation. Contaminated soils could contaminate infiltrating water as it passes through them and prolong the ground water cleanup effort.” Results in DOE 1999c indicate background solid-phase uranium values of 0.47 mg/kg with a 5% nitric acid leach for two samples compared to subpile soil analyses that averaged 2.8 mg/kg uranium from four samples (the highest subpile soil uranium concentration was 8.4 mg/kg). In addition, limited subpile soil sampling conducted as part of the Site Observational Work Plan identified residual uranium concentrations in sediments beneath the former ore stockpile area as high as 12 mg/kg with an average residual uranium concentration less than 2 mg/kg (DOE 2017i).

DOE 2011 mentions that alluvial groundwater levels increase up to 6 feet during high-river-runoff months, which implies that a rising water table could periodically leach uranium from the unsaturated zone. Similarly, it is also possible that downward seeping moisture from episodic precipitation events could leach uranium from the unsaturated zone (DOE 2011).

NRZs are present at several locations and have been mapped by Yabusaki et al. (2017). The NRZs contain an abundance of organic carbon, relatively large amounts of solid-phase constituents in the form of U(IV), adsorbed U(VI), iron oxides, and ferrous sulfides. The U(IV) present in the NRZs (category 2) has been found to consist of both crystalline uraninite and noncrystalline, monomeric U(IV), with uraninite being more resistant to mobilization in groundwater and, therefore, a more enduring contributor to uranium plume persistence (Qafoku et al. 2014). The NRZs are composed of more fine-grained sediments (silts and clays) than are associated with the alluvium (DOE 2017h). Thus, the NRZs are expected to have a low hydraulic conductivity (DOE 2017h). In the NRZs, molecular diffusion of the dissolved uranium derived from the oxidation of U(IV) is thought to slow the release of U(VI) to the preferential flow paths (category 3) that convey most of the contaminant mass discharging to the Colorado River. Seasonal changes in hydraulic and chemical processes in the unsaturated zone also play an important role in the release of uranium from the NRZs (Campbell et al. 2011; Qafoku et al. 2014). Additional data on the NRZs at the Old Rifle site are provided in Janot et al. (2016) and Noël et al. (2017). Of all the LM sites discussed in this report, the Old Rifle site has the most data and published papers related to NRZs.

2.11 Riverton, Wyoming, Processing Site

The Riverton, Wyoming, Processing Site is the location of a former uranium- and vanadium-ore processing mill that operated from 1958 to 1963 and processed 816,470 tons of uranium ore mined in the Gas Hills mining district in Wyoming (Dam et al. 2015). The site is located on an alluvial terrace between the Wind River and the Little Wind River in Fremont County, approximately 2.3 miles southwest of the town of Riverton, Wyoming, within the boundaries of the Wind River Indian Reservation on land now owned by Chemtrade Refinery Services (Figure 11). Milling operations created radioactive mill tailings and uranium, radium, and thorium contamination in soils and construction debris (DOE 2016g). A tailings pile covered about 72 acres of the 140-acre site to an average depth of 4 feet. In 1988, about 1.8 million cubic yards of the contaminated materials were removed from the site and relocated to the Gas Hills east disposal site 45 miles away. DOE completed surface remediation of the Riverton site in 1989 (DOE 2016g).

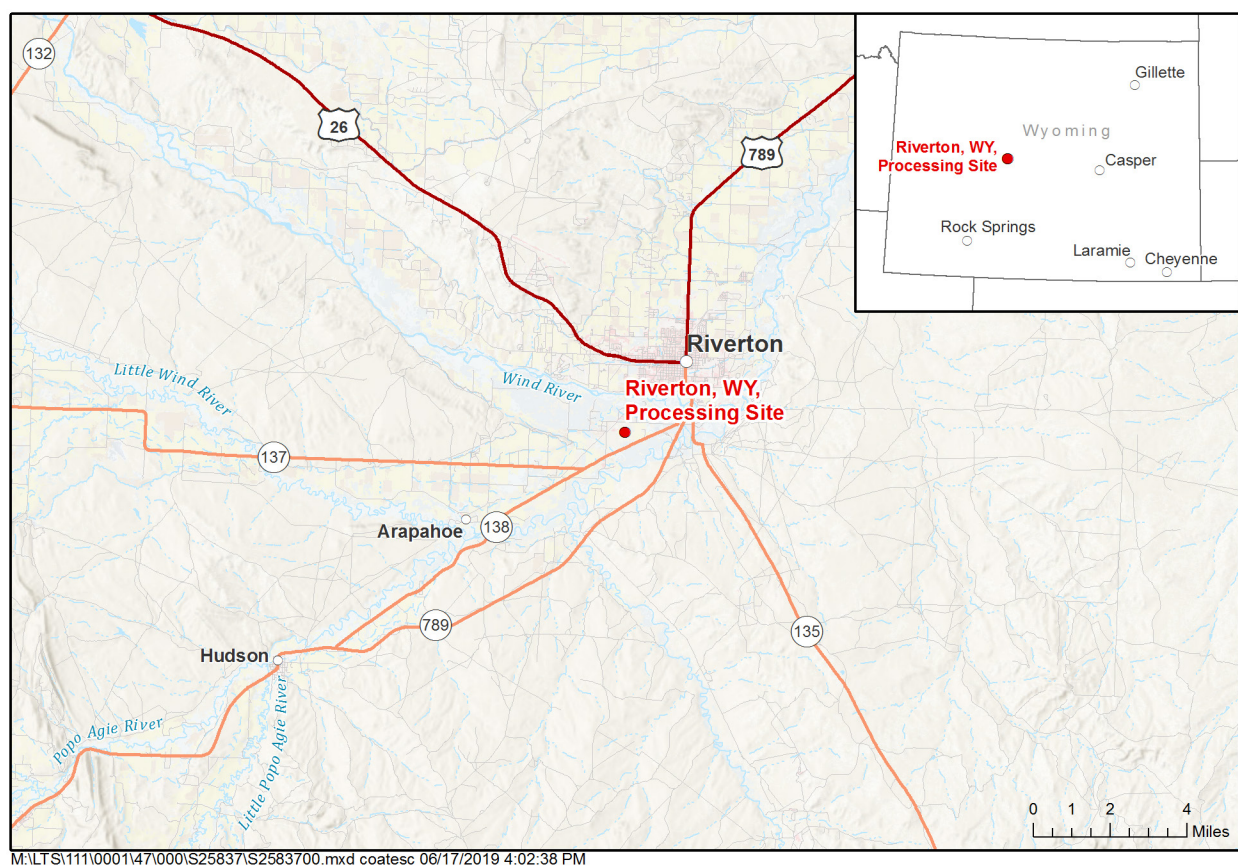


Figure 11. Site Location Map of the Riverton, Wyoming, Processing Site

The Riverton, Wyoming, Processing Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/riverton-wyoming-processing-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=RVT>.

2.11.1 Hydrogeology

Groundwater occurs in three aquifers beneath the site: (1) an upper unconfined alluvial aquifer (alluvial aquifer), (2) a middle semiconfined aquifer, and (3) a deeper confined aquifer (DOE 1998b). The alluvial aquifer is approximately 15–20 feet thick and consists primarily of sand and gravel, with lesser amounts of clay-sized to cobble-sized particles (Dam et al. 2015). The middle semiconfined aquifer and the deeper confined aquifer are composed of the upper units of the Eocene Wind River Formation, which is over 500 feet thick in the vicinity of the site (Dam et al. 2015). The middle semiconfined aquifer is composed of 15–30 feet of sandstone separated from the unconsolidated alluvial material by a discontinuous layer of shale 5–10 feet thick (Dam et al. 2015). The deeper confined aquifer is composed of shales and sandstones. The confining unit between the semiconfined aquifer and the confined aquifer is a 10- to 25-foot-thick layer of shale (DOE 2016h). Depth to groundwater in the surficial aquifer is generally less than 10 feet bgs (DOE 2016h). Groundwater in the alluvial aquifer flows from the former mill site southeast to the Little Wind River.

2.11.2 Evidence of Persistent Secondary Uranium Sources

In June 2010, flooding of the Little Wind River, calculated to be a 75-year event, occurred at the site and resulted in overbank flow over a large area downgradient of the former mill site. The overbank flow possibly combined with a higher water table mobilized contaminants within the aquifer because DOE measured significant increases in contaminant concentrations in monitoring wells where the flooding occurred immediately after the flood receded (Dam et al. 2015).

The increase in concentration of the contaminants of potential concern (COPCs) that occurred as a result of the June 2010 flooding led DOE to conduct additional investigations focused on evaluating the solid-phase uranium inventory at the site (DOE 2016h). These additional investigations included backhoe trenching to obtain visual descriptions of the subsurface lithology and for identifying and sampling NRZs and evaporites, plus sonic drilling of 10 boreholes to allow the collection of representative soil samples to better characterize the distribution and concentration of COPCs in the subsurface.

The concentration of solid-phase uranium in the borehole and trench samples ranges from 0.26 to 22 mg/kg uranium (DOE 2016h). In the silt layer outside the uranium plume, solid-phase uranium concentrations are typically 1.4 mg/kg and are less than 1 mg/kg in the underlying sand and gravel throughout the site. The highest concentrations of solid-phase uranium occur (1) in the NRZs (up to 22 mg/kg, category 2), (2) in the silt layer over the uranium plume (up to 10 mg/kg, category 2), and (3) just above and below the water table beneath the former tailings impoundment area (up to 3.6 mg/kg above the water table and up to 5.5 mg/kg below the water table, category 1). The high concentrations of uranium in the silt are thought to result from the formation of evaporite minerals with the high ongoing source of uranium from the groundwater contaminant plume. Surficial evaporite samples collected along the bank of the Little Wind River in 2014 had a maximum uranium concentration of 66 mg/kg over the uranium plume compared to a background concentration of 1.4 mg/kg (DOE 2014b).

2.12 Shiprock, New Mexico, Disposal Site

The Shiprock, New Mexico, Disposal Site is a former uranium- and vanadium-ore processing facility that operated from 1954 to 1968 and is in the Navajo Nation near Shiprock, New Mexico, approximately 28 miles west of Farmington, New Mexico (Figure 12) (DOE 2017j). The disposal cell and adjacent former mill site occupy approximately 230 acres on a terrace south of the San Juan River (DOE 2017j). An escarpment about 50–60 feet high forms the boundary between the terrace and San Juan River floodplain, which extends 1500 feet north of the mill site (DOE 2000). The mill processed about 1.5 million tons of ore that initially contained carnotite and roscoelite ($K(V^{3+}, Al, Mg)_2AlSi_3O_{10}(OH)_2$) from the Salt Wash Member of the Morrison Formation in the Lukachukai Mountains of northeast Arizona, but beginning in 1963, more than half the ore was from mines in the Uraavan Mineral Belt (DOE 2000). The tailings were placed in two large piles that covered most of the area currently occupied by the disposal cell (DOE 2018a). Raffinate (liquid waste generated by the milling process) was stored in up to 10 unlined raffinate ponds south and southwest of the tailings piles (DOE 2000).

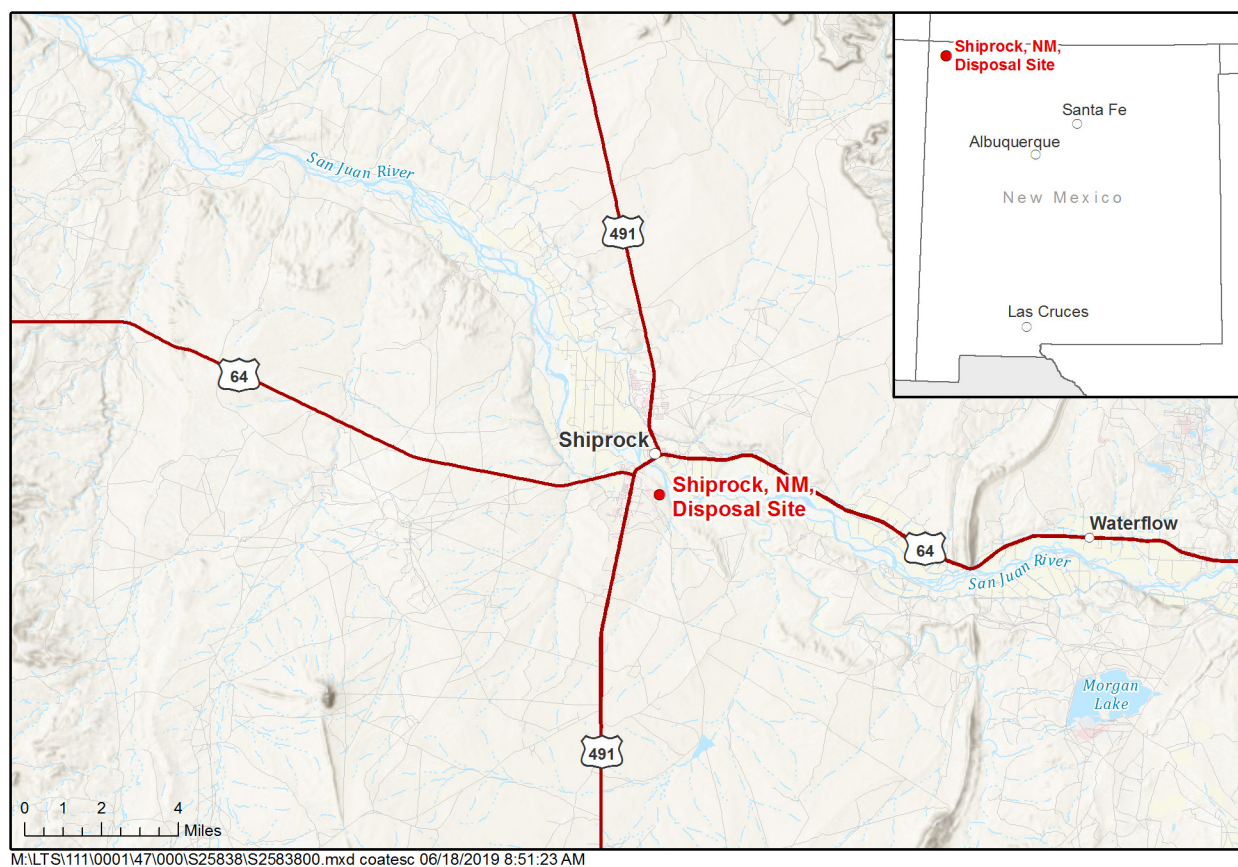


Figure 12. Site Location Map of the Shiprock, New Mexico, Disposal Site

The Shiprock, New Mexico, Disposal Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/shiprock-new-mexico-disposal-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=SHP>.

2.12.1 Hydrogeology

Groundwater on the terrace occurs within both weathered and unweathered portions of Mancos Shale bedrock and a few feet of the overlying alluvium (DOE 2018a). Prior to the mill beginning operations in 1954, the only potential source of groundwater recharge to the terrace was thought to be precipitation infiltration. The original mantle of eolian silt (loess) covering the terrace, however, is thought to have restricted recharge and favored runoff, especially just south of the disposal cell, where the silt attains a thickness of almost 30 feet (DOE 2000). Drilling data from 1998 to 2000 indicated that the loess is dry even in present conditions when anthropogenic water is present in the underlying gravel. Thus, the terrace gravel unit likely received little to no recharge under pristine conditions and is hypothesized to have been unsaturated (DOE 2000).

Groundwater on the floodplain occurs primarily within the alluvium (alluvial aquifer) deposited by the San Juan River on top of the Mancos Shale, the upper few feet of which are weathered (DOE 2018a). The alluvium is composed of unconsolidated medium- to coarse-grained sand, gravel, and cobbles (basal gravels) (DOE 2000). In most areas of the floodplain, the finer-grained alluvial sediments consisting of fine- to coarse-grained sand, silt, and some clay overlie the basal gravels. The thickness of the shallow, finer-grained alluvium ranges from about 2 to 6 feet in the south third of the floodplain to approximately 8 to 12 feet in the north half. The combined thickness of basal gravels and fine-grained surficial materials varies across the floodplain but can be as much as 25 feet in the north end of the floodplain.

The alluvial aquifer is considered unconfined over most of the floodplain and is hydraulically connected to the San Juan River, which borders the entire length of the alluvial aquifer (7300 feet) on its east and north sides. The San Juan River is a source of groundwater recharge and discharge to the alluvial aquifer, and prior to milling operations the alluvial aquifer is believed to have been recharged entirely by the San Juan River and by infiltration of precipitation and runoff (DOE 2000). Variable flows in the San Juan River and losses of subsurface water by evapotranspiration during summer and fall cause groundwater flow patterns in the alluvial aquifer to change noticeably between seasons and from year to year. Temporal changes in river stage cause the saturated thickness of the alluvial aquifer to vary as much as 3 to 4 feet (DOE 2018a). The depth to groundwater during low flow periods ranges from about 3 to 7 feet.

2.12.2 Evidence of Persistent Secondary Uranium Sources

At the Shiprock site, evaporite minerals (category 2) are present within Bob Lee Wash, on the face of the terrace, and on the floodplain adjacent to the San Juan River (DOE 2016d). Figure 7 in DOE 2016d shows the location of evaporite samples collected from these areas. The concentration of uranium in the samples collected from Bob Lee Wash ranged from 0.95 to 76 mg/kg. The concentration of uranium in samples collected from the face of the terrace and on the floodplain ranged from 0.51 to 53 mg/kg. Background uranium concentrations in evaporite samples were determined in Many Devils Wash, with uranium concentrations up to 1.8 mg/kg (DOE 2016d). The dissolution of the evaporites by rain and snowmelt can contribute high concentrations of uranium to groundwater, but no data (e.g., columns or seasonal groundwater quality variations) beyond solid-phase concentrations are provided.

Three samples collected from Bob Lee Wash were analyzed for mineralogy using X-ray diffraction, and the results indicated the dominant mineral to be thenardite (Na_2SO_4) with minor amounts of blödite ($\text{Na}_2\text{Mg}(\text{SO}_4)_2 \cdot 4(\text{H}_2\text{O})$), and possibly sideronatriite ($\text{Na}_2\text{Fe}(\text{SO}_4)_2(\text{OH}) \cdot 3(\text{H}_2\text{O})$) (DOE 2016d). Other phases found in the samples were quartz and calcite, but no uranium minerals were identified. These minerals are consistent with a groundwater source that is high in sodium and sulfate concentrations. For uranium, background groundwater concentrations in this area can also be as high as 0.15 mg/L due to the occurrence of the Mancos Shale (Kamp and Morrison 2014). While naturally occurring uranium in the groundwater can provide some uranium to onsite evaporites, the much lower uranium concentrations in the evaporites at Many Devils Wash compared to those at the Shiprock site appear to indicate an onsite mill-derived source of uranium in the Shiprock site evaporites.

NRZs (category 2) have also been identified in the floodplain at the Shiprock site, with elevated solid-phase uranium concentrations (Noël et al. 2019). The Shiprock NRZ studied by Noël et al. (2019) was characterized as a fine-grained NRZ that experiences redox cycling due to seasonal fluctuations in moisture content. Prior research has indicated that the accumulation of uranium in NRZs can be reoxidized, solubilized, and transported away in groundwater, thus providing a sink/source relationship for these zones of secondary uranium. However, the work from Noël et al. (2019) indicates that the redox oscillations driven by annual water table fluctuations can be accompanied by strong evapotranspiration in low-permeability sediments. Data from Noël et al. (2019) indicate that this cycling can form relatively immobile uranium that is then resistant to future redox perturbations. Therefore, these NRZs may be more of a long-term sink and less of a long-term source for uranium than previously thought.

2.13 Tuba City, Arizona, Disposal Site

The Tuba City, Arizona, Disposal Site is in the Navajo Nation near the Hopi Reservation approximately 5 miles east of Tuba City and 85 miles northeast of Flagstaff, Arizona (Figure 13). This disposal site is on a gently sloping terrace above Moenkopi Wash, an intermittent stream about 4000 feet south of the site that drains into the Little Colorado River (DOE 2017k). A uranium mill was in operation at the site from 1956 to 1966 and processed approximately 800,000 tons of ore (DOE 2017k). The mill processed an average of 300 tons per day between 1956 and 1962 using a sulfuric acid leach and processed an average of 200 tons per day between 1963 and 1966 using sodium carbonate (DOE 1999d). The tailings were conveyed in a slurry from the mill to an unlined impoundment. Standing water in the impoundment and other mill process wastewater was conveyed to unlined evaporation ponds adjacent to the tailings impoundment. The surface area of the impoundment and ponds was about 55 acres (DOE 2017k). Windblown tailings covered an additional 250 acres northeast of the mill site (DOE 2017k).

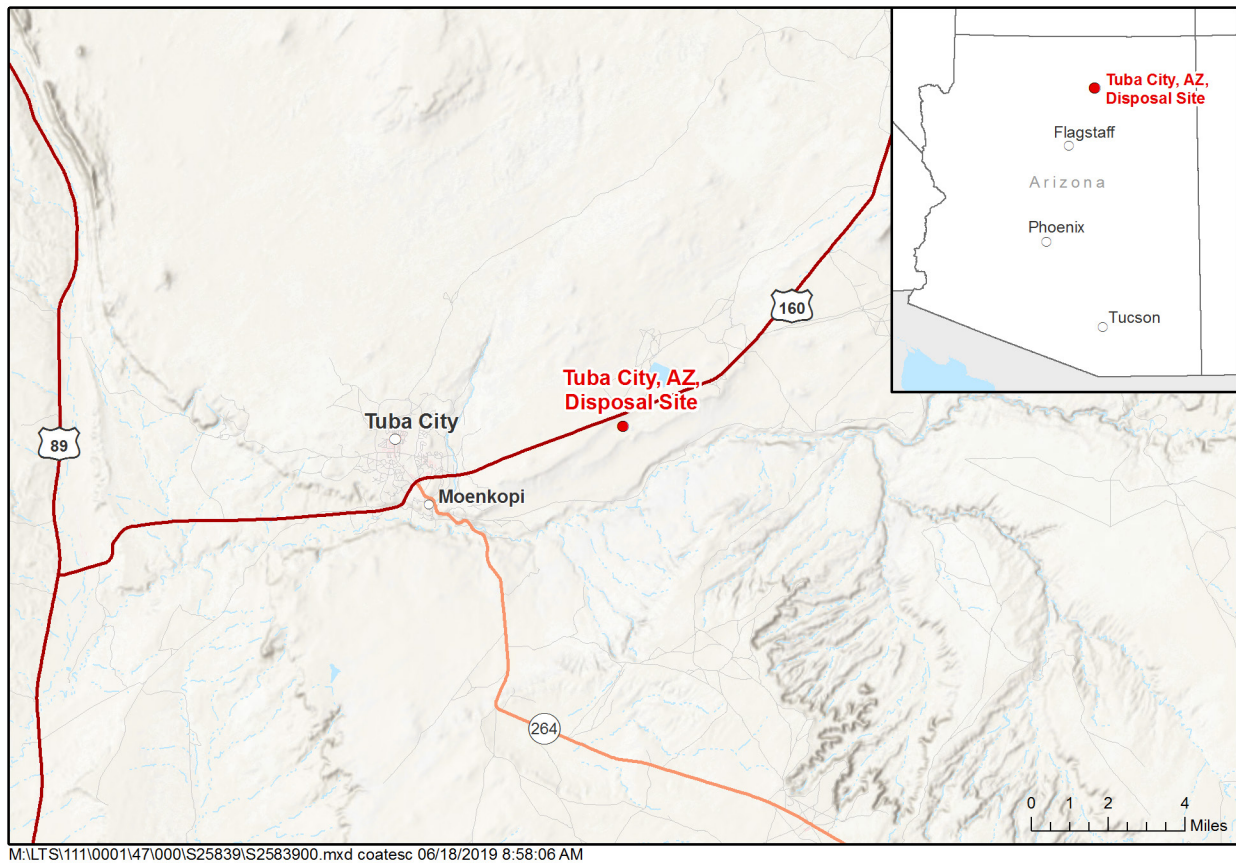


Figure 13. Site Location Map of the Tuba City, Arizona, Disposal Site

DOE began site cleanup in 1988 and placed the onsite tailings piles, debris from demolished buildings, and windblown tailings in an engineered disposal cell onsite, and the cleanup was completed in 1990. The disposal cell occupies an area of 50 acres on the 145-acre site (DOE 2017j). The disposal cell completely covers the mill tailings impoundment but does not fully cover the area where the mill's evaporation ponds were located.

The Tuba City, Arizona, Disposal Site fact sheet and related site documents can be found at <https://www.energy.gov/lm/tuba-city-arizona-disposal-site>, and a site map with associated data can be found at <https://gems.lm.doe.gov/#site=TUB>.

2.13.1 Hydrogeology

The aquifer underlying the site consists of, in descending order, the Navajo Sandstone, the Kayenta Formation, and the Moenave Formation and is referred to as the N-aquifer. Overlying the N-aquifer are thin surficial deposits of unconsolidated dune sand and alluvial gravels (DOE 2017k). The depth to the top of the saturated zone at the site in the Navajo Sandstone under natural conditions is approximately 35–50 feet (DOE 2004a). The Navajo Sandstone is fine- to medium-grained, is locally cemented with carbonate, and displays large-scale cross-beds (DOE 1996). The underlying Kayenta Formation consists of approximately 100 feet of less-resistant, thin-bedded, red silt and fine sand, with thin (<2 ft) limestone beds. Because of its fine-grained composition, the Kayenta Formation is not water bearing (DOE 2016i). The contact

between the Navajo Sandstone and the Kayenta is obscured by a thick (400 ft) “intertonguing interval” called the Kayenta/Navajo Transition Zone (KNTZ).

The N-aquifer is unconfined in the Tuba City area, and the saturated thickness of the N-aquifer near the disposal cell is about 350 feet but thins rapidly within 2000 feet south of the disposal cell (DOE 1996; DOE 2017k). The average hydraulic conductivity near the site is 0.5 foot per day (DOE 1999d), but measurements of conductivity within the Navajo Sandstone and KNTZ around the site ranged by 6 orders of magnitude (DOE 2016i). Approximately 4000 feet south of the site, seeps issue forth along cliff bands that border the incised valley of Moenkopi Wash and phreatic vegetation becomes more abundant south of the disposal cell (DOE 1999d). Downgradient of the disposal cell, groundwater elevations decrease toward the escarpment to the lower terrace, where depths-to-water range from 19 to 38 feet. The lower terrace extends about 4000 feet to sandstone cliffs that overlook the Moenkopi Wash floodplain.

2.13.2 Evidence of Persistent Secondary Uranium Sources

Elevated concentrations of uranium have been measured in the bedrock beneath former evaporation ponds (category 1). Five 3-inch-diameter cores were extracted at the Tuba City site in 1997 by MSE Technologies Inc. (DOE 2004b). Four cores were within the uranium plume area, with three of those being below former evaporation ponds. One core was to the northwest (upgradient) of the disposal cell and served as a source for background values. Uranium was determined using a 20% nitric acid leach solution (DOE 2004b). The highest uranium concentrations were found at the top of the core and decreased rapidly with depth in soil cores, suggesting that mill-derived uranium from the former evaporation ponds is in the shallow soil. The maximum uranium concentration was 3.5 mg/kg (in core MSE4) compared to background concentrations (in MSE6) that were all below 0.086 mg/kg uranium (DOE 2004b).

Column work was completed on the MSE4 core using deionized water as the influent. The resulting effluent had water quality that was similar to that in the current contaminant plume (DOE 2018b). This effluent had up to 740 µg/L uranium but was below uranium standards within 3 pore volumes.

Rate-limited mass transfer or nonequilibrium processes (category 3) may also be influential at this site because of the hydraulic conductivity variations in the Navajo Sandstone ranging over several orders of magnitude (DOE 2016i; DOE 2018b).

3.0 Summary of Persistent Secondary Uranium Sources Mechanisms

Uranium that is precipitated or sorbed in the unsaturated or saturated zone below former tailings, former evaporation ponds, former ore storage areas, or current disposal cells in concentrations and forms that are not typical of sorption to aquifer solids within the main groundwater uranium plume likely occurs at every LM site. This residual uranium is a persistent secondary source that continues to provide contamination to groundwater. Mechanisms discussed in this report were often not considered in original site conceptual models where tailings and evaporation ponds were removed. However, because site remediation standards did not include soil removal to background uranium concentrations, some residual uranium was left behind.

Of the sites discussed in this report, the only sites that did not have directly measured residual uranium were the Bluewater, Fernald Preserve, and Shiprock sites. For the Bluewater site, a mineralized zone with secondary uranium below the disposal cell is theorized but was not directly measured (DOE 2014a). At the Fernald Preserve site, residual uranium after source zones have been removed has not been directly measured, but remedial levels for solid-phase uranium were well above background values (DOE 1998a). For the Shiprock site, a mineralized zone below the disposal cell is also likely, but was not theorized in any documents and has not been directly measured.

Uranium retained in downgradient areas that is precipitated or sorbed in the unsaturated zone occurs in evaporite-rich areas. Evaporite-rich areas occur where the contaminated groundwater is close to the surface at sites with high evapotranspiration rates. Evaporites may occur either at the surface or in the subsurface and are summarized for LM sites in a prior report (DOE 2016d). Evaporite-rich areas provide a secondary uranium source that can be mobilized with large recharge events. Sites with documented excess uranium in the unsaturated zone or at the surface include the following:

- Green River, Utah, Disposal Site
- Monticello, Utah, Processing Site
- Riverton, Wyoming, Processing Site
- Shiprock, New Mexico, Disposal Site

The Monument Valley site is a special case where the surficial uranium may occur as a uranium/vanadium mineral below former evaporation ponds, and this mineral precipitation may have been enhanced by evaporation (DOE 2016d).

Uranium retained in downgradient areas that is precipitated or sorbed in the unsaturated or saturated zone in concentrations that are greater than typical of sorption within the main groundwater uranium plume also occurs in areas with high organic carbon (NRZs). Most of the documentation on high organic carbon retaining uranium refers to zones at and below the water table that create and maintain anoxic conditions. However, the cycling of uranium at NRZs with varying redox processes above and below the water table is a topic of current research (Noël et al. 2019). Although NRZs do appear to be uranium sinks that have higher solid-phase uranium concentrations than background (persistent secondary uranium sources), whether or not this uranium is released under normal conditions at LM sites is not clear. Conditions created in laboratory columns that subject NRZ material to oxidizing conditions can release high uranium concentrations to the column effluent (up to 1.6 mg/L, DOE 2017b). Thus, future oxidation of NRZs due to lower groundwater elevations could release uranium from the solid phase to the water phase. LM sites with documented uranium concentrations in NRZs include:

- Grand Junction, Colorado, Site
- Naturita, Colorado, Processing Site
- Rifle, Colorado, Old Processing Site
- Riverton, Wyoming, Processing Site
- Shiprock, New Mexico, Disposal Site

Rate-limited mass transfer or nonequilibrium processes, such as matrix diffusion due to large hydraulic conductivity variations, are suggested in documents for the following sites:

- Bluewater, New Mexico, Disposal Site
- Fernald Preserve, Ohio, Site
- Rifle, Colorado, Old Processing Site
- Tuba City, Arizona, Disposal Site

Matrix diffusion or dual-domain processes likely occur at all LM sites to varying degrees based on hydraulic conductivity variations and can create uranium concentrations that take much longer to decline than predicted. However, it is difficult to determine the exact processes that create slow declines in uranium concentrations (e.g., rate-limited mass transfer vs. residual uranium below former tailings impoundment). Thus, no specific documents with the quantification of rate-limited mass transfer are available. Determining processes that create such plume persistence issues requires thorough site conceptual models based on detailed site characterization data.

Non-DOE related scientific literature was reviewed to determine whether other uranium-contaminated sites outside of the United States have persistent secondary uranium sources. While there are many other uranium mill and uranium-related processing sites throughout the world, available literature on those sites is generally focused on uranium plume characterization related to known source areas (mill tailings, evaporation ponds, processing wastes, etc.). No publications that deal specifically with the secondary uranium sources discussed in this report were found.

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