

# New Mexico GEOLOGY

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## Cover Image

Jackpile mine, looking north, around 1980. The Jackpile-Paquate open pit mine was discovered by aerial reconnaissance in 1951 and closed in 1983. It was the largest uranium mine at that time, consisting of 4 coalescing pits. The mine complex produced more than 80 million pounds of  $U_3O_8$  from the Jackpile Sandstone of the Morrison Formation. The Jackpile-Paquate mine is now an EPA Superfund site. W.L. Chenoweth photo.

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# Uranium in New Mexico: A Special Issue of *New Mexico Geology*

Virginia T. McLemore and Bonnie A. Frey

In January 2016, a team of experts from the uranium industry and researchers from two New Mexico universities assembled at the Sevilleta National Wildlife Refuge for a three-day workshop to discuss topics associated with in situ recovery of uranium. Part of the motivation for this workshop was the recognition that there has been little new research completed by the uranium industry and that much of the current uranium knowledge base consists of research, mapping and technology from the 1970s and 1980s. Researchers and industry workers today have many new technologies available to help re-evaluate exploration, mining, processing, reclamation and restoration. Furthermore, today's industry has become much more open to sharing data, in part because today's strenuous permitting process has turned previously proprietary information into public record.

During a period of nearly three decades (1951–1980), the Grants district in northwestern New Mexico yielded more uranium than any other district in the United States, thereby making New Mexico a major producer of uranium. Today, uranium is used primarily in nuclear reactors to produce electricity via nuclear fission. Although no producing operations exist in New Mexico today, numerous companies have acquired uranium properties within the Grants, Hooks Ranch-Riley, and Red Basin-Pietown districts and plan to explore and develop deposits in the future (shown on McLemore and Chenoweth, 1989; listed in <http://nmgs.nmt.edu/repository/index.cfm?rid=2013002>). New Mexico has world-class uranium deposits in the Grants district and ranks 2nd in uranium reserves in the United States, after Wyoming. The New Mexico reserves amount to 64 million short tons of ore at 0.14%  $U_3O_8$  (179 million pounds  $U_3O_8$ ) at \$50/pound. The most important deposits in the state are within the sandstones of the Jurassic Morrison Formation in the Grants district (McLemore and Chenoweth, 2003). More than 340 million pounds of  $U_3O_8$  have been produced from Morrison Formation deposits from 1948–2002, accounting for 97% of the total production in New Mexico and more than 30% of the total production in the country.

Companies face several challenges to begin producing uranium in the Grants district and elsewhere in New Mexico again. These challenges include the following:

- No conventional mills remain in New Mexico to process the ore, adding to the cost of producing uranium in the state. Currently, all conventional ore must be processed by the White Mesa Mill near Blanding, Utah, or heap-leached on site. New infrastructure will need to be built before conventional mining can resume.
- Permitting for new in situ recovery and conventional mines and mills will take years to complete.
- Closure plans, including reclamation, must be developed before mining or in situ

recovery begins. Modern regulatory costs will add to the cost of producing uranium in the United States.

- Some communities, especially the Navajo Nation communities, do not view development of uranium properties as favorable. The Navajo Nation has declared that no uranium production will occur on tribal lands. Most of Mount Taylor and adjacent mesas have been designated as the Mount Taylor Traditional Cultural Property; the effect of this designation on uranium exploration and mining is uncertain.
- High-grade, low-cost uranium deposits in Canada and Australia and the large low-grade deposits in Kazakhstan are sufficient to meet current international demands; additional resources will be required to meet long-term future requirements.

With some of these limitations in mind, the group that met in January compiled a list of research activities that could support renewed activity in the New Mexico uranium industry. Topics include workforce training, resource characterization, hydrogeological and geochemical modeling, updated environmental and regulatory protections, improved understanding of depositional mineralogy, microbiology and geochemistry, and the development of new recovery and restoration technology.

The group also settled on several outreach activities that could foster discussion within the community. It was determined that a conference held in New Mexico would be the best venue for showcasing the current research and collected knowledge associated with today's uranium industry. In response, Virginia McLemore and Bonnie Frey volunteered to co-chair a key-note session on uranium for the 2017 spring meeting of the New Mexico Geological Society (NMGS). Another outcome are two special editions of *New Mexico Geology*, including this fall issue and the upcoming spring issue. We hope that these editions of *New Mexico Geology* will provide information about current uranium-mining topics. We also hope that today's mining environment can encourage collaboration among researchers, industry, government agencies, and community representatives. Finally, we hope that interested readers of these articles will join us during the April 2017 NMGS spring meeting in Socorro.

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# In situ recovery of sandstone-hosted uranium deposits in New Mexico: past, present, and future issues and potential

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## Abstract

In situ recovery (ISR) operations have been proposed to recover uranium from sandstone-hosted uranium deposits in New Mexico. ISR (also known as in situ leaching, solution mining, solution-leach mining, leach mining) is conducted by wells that circulate native groundwater, amended with oxygen (or other forms of oxidant) to dissolve the uranium and gaseous carbon dioxide (or some form of sodium bicarbonate) to complex the uranium in order to keep it in solution through the ore zone. This amended groundwater is commonly referred to as lixiviant. The lixiviant dissolves uranium as it is drawn from injection wells through the uranium-bearing host rock by pumps in nearby extraction wells, and is subsequently piped to a processing plant where the uranium is extracted from the solution. The groundwater is then refortified and sent back to the ore zone through the injection wells to recover additional uranium. The cycle continues until the desired uranium extraction is complete. Thereafter groundwater restoration is conducted. Several technical and regulatory criteria must be met in order for ISR to be successful. To comply with post-mining restoration criteria dictated by state and federal regulations, the groundwater in the mined areas is restored to baseline or other agreed upon water quality standards. This is usually accomplished by circulating clean groundwater through the mined zones to remove the lixiviant. Because groundwater is the fundamental leaching agent, the uranium deposit must be hosted within permeable sandstone below the water table and generally confined by less permeable strata for proper hydrodynamic wellfield control. The mineralized portion of the aquifer must qualify for an "Exemption" from the EPA (U.S. Environmental Protection Administration) from being an underground source of drinking water. A number of ISR test and pilot operations have been conducted in New Mexico in the past (Mobil, Crownpoint; UNC-Teton, Section 23; Grace Nuclear, Hook's Ranch, Seboyeta, Church Rock; Anaconda, Windwhip). Also, analogous to the ISR process, United Nuclear and Kerr-McGee (later Quivira Mining Co., Rio Algom) successfully produced uranium from mine-water recovery (recirculated mine water) from underground mines in the Ambrosia Lake area during the mid-1960s to 2002. Potential ISR site locations in the U.S., including in New Mexico, require careful aquifer characterization and project operational design and monitoring. With such proper site characterization and design, ISR is a viable alternative mining technology to provide future uranium recovery from many of New Mexico's known uranium deposits. This initial investigation suggests that a significant portion of deposits in the Grants uranium district may be amenable to ISR production.

## Introduction

Uranium in New Mexico is found in rocks of all ages and lithologies, ranging from Precambrian granites to recent travertine deposits (Fig. 1; McLemore, 1983, 2007; McLemore and Chenoweth, 1989, 2017). Uranium is found in sandstones, coals, limestones, shales, igneous and metamorphic rocks, pegmatites, veins, volcanic rocks, and breccia pipe deposits. However, most of the economic uranium deposits are hosted by sandstones and most of the uranium production in New Mexico has come from the Westwater Canyon Member of the Jurassic Morrison Formation in the Grants uranium district, in McKinley and Cibola (formerly Valencia) Counties (Table 1; McLemore, 1983). The Grants uranium district represents one large area in the southern part of the San Juan Basin, extending from east of Laguna to west of Gallup and consists of eight sub-districts (Fig. 2; McLemore and Chenoweth, 1989, 2016; McLemore, 2007). During a period of nearly three decades (1951–1980), the Grants uranium district yielded nearly 347 million lbs of  $U_3O_8$ , almost all of New Mexico's production, and more uranium than any other district in the United States (Table 1). The Grants district is probably 7th in total world production behind East Germany, the Athabasca Basin in Canada, Kazakhstan and South Africa (Tom Pool, International Nuclear, Golden, Colorado, written communication, December 3, 2002). Although there are no operating mines in the Grants district today, numerous companies have acquired uranium properties and plan to explore and develop deposits in the district in the future.

Several ISR operations have been proposed to recover uranium from sandstone-uranium deposits in New Mexico, mostly in the Grants uranium district, although one operation was tested in the Hooks Ranch area in Socorro County (Hook Ranch—Riley district). In situ recovery (also known as in situ leaching, solution mining, solution-leach mining, leach mining) uses a series of injection and extraction wells, which circulate amended native groundwater through the ore zone. This groundwater solution, commonly referred to as lixiviant, dissolves and complexes uranium as it is drawn from injection wells through the uranium-bearing host rock by pumps in nearby production wells, which then sends the uranium-rich water to the processing plant where the uranium is removed. The water is then refortified and sent back to the ore zone through the injection wells to recover more uranium. The cycle continues until the desired uranium extraction is complete. Thereafter, groundwater restoration is conducted.

The purpose of this paper is to briefly describe the production histories, geology, resources, environmental issues, and future potential of uranium deposits in New Mexico that are possibly amenable to ISR. Although

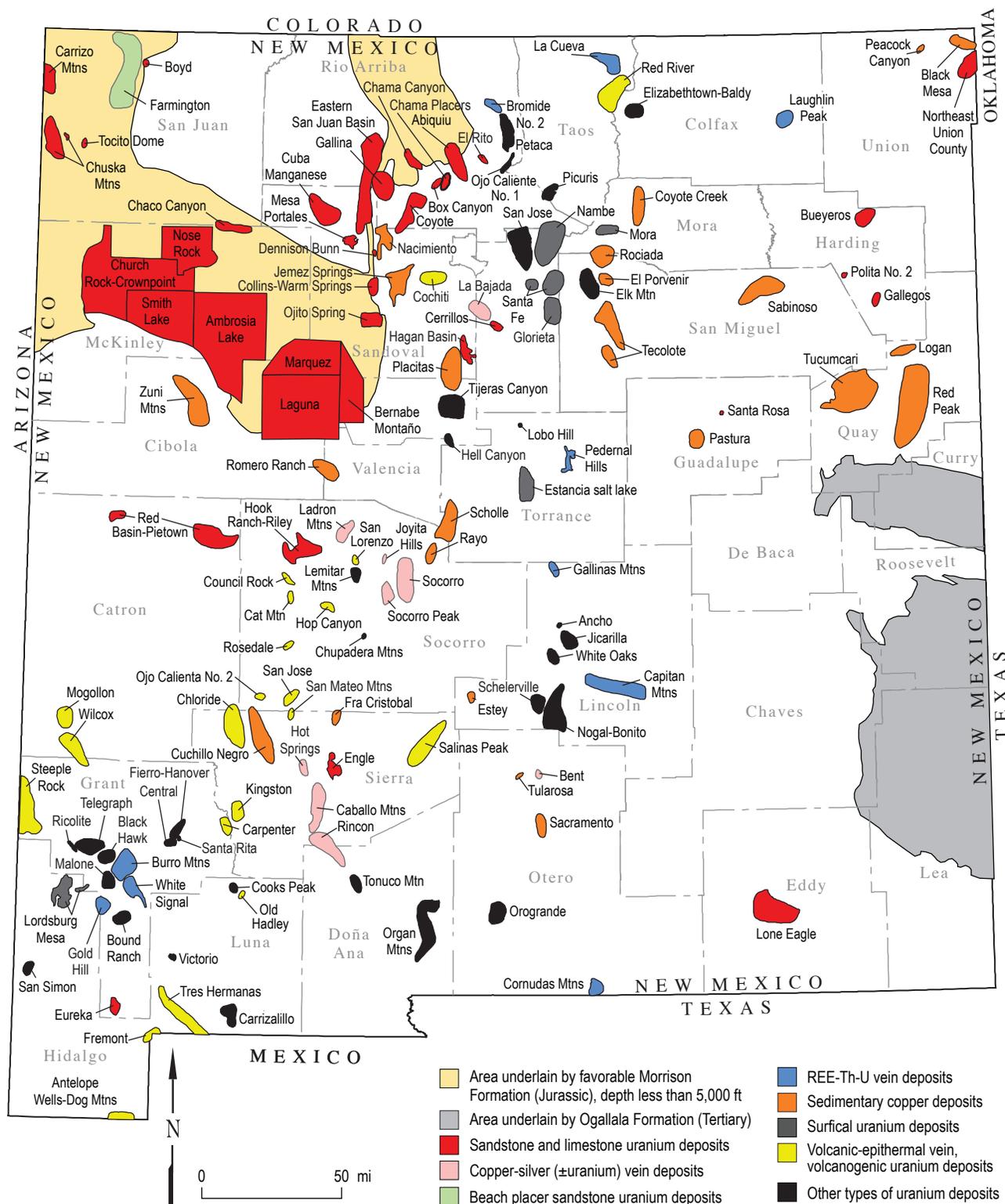


Figure 1. Mining districts that have uranium deposits and other areas favorable for uranium in New Mexico (modified from McLemore and Chenoweth, 1989). Each district is color-coded according to the predominant type of deposit; other types of uranium deposits are found in most districts.

there has been no large-scale commercial ISR production in New Mexico, several small-scale pilot projects have been conducted in the past. This paper will include a summary of these past ISR operations, mostly summarized and updated from Holen and Hatchell (1986). Much of this paper is summarized from McLemore (1983), Holen and Hatchell (1986),

McLemore and Chenoweth (1989, 2017), McLemore et al. (2002), McLemore (2007) and other reports as cited. Information on specific mines and deposits in New Mexico can be found in cited references, McLemore (1983, 2007), McLemore and Chenoweth (1989, 2016), and McLemore et al. (2002).

## In situ recovery

### What is in situ recovery?

ISR is the extraction of uranium by recirculating, through wells, groundwater fortified with relatively benign chemical solutions; oxidizing, complexing, and mobilizing the uranium; and recovery of the uraniumiferous solutions through production wells and pumping the solution to the surface for further processing. ISR is commonly called in situ leaching or ISL. The term ISR is appropriately used when the entire uranium recovery cycle is described from subsurface dissolution, through processing, drying and packaging. The injection solution is fundamentally groundwater that has been pumped from the ore body aquifer, to which relatively small concentrations of an oxidant such as liquid oxygen or hydrogen peroxide and a complexing agent such as sodium bicarbonate have been added. Restoration of the aquifer is mandatory in the U.S. regardless of the type of lixiviant used. Sulfuric acid is an effective lixiviant reagent that is used elsewhere in the world, but in the U.S. using an acid lixiviant system is generally not done because acid lixivants may cause mineralogical changes to the host sandstones that could

adversely impact hydrological conditions and post-mining restoration efforts. As a result, commercial operations in the U.S. have used the sodium bicarbonate-type alkaline lixiviant chemistry that has been described previously.

From 2004–2009, ISR production was the source of 20–34% of the total world production, mostly from mines in Australia, China, Kazakhstan, United States, and Uzbekistan, whereas in 2014, ISR production increased to 51% of the total world production of uranium (IAEA, 2004; *The Changing World of Uranium Mining, A Monday Morning Musing* from Mickey the Mercenary Geologist, [http://www.goldgeologist.com/mercenary\\_musings/musing-160125-The-Changing-World-of-Uranium-Mining.pdf](http://www.goldgeologist.com/mercenary_musings/musing-160125-The-Changing-World-of-Uranium-Mining.pdf), accessed 1/28/16). Uranium ISR production costs are generally lower than those associated with conventional (open pit or underground) mining and milling, with considerably lower capital and operating costs. Certain environmental considerations associated with ISR operations are more favorable than traditional conventional mining and milling operations because there are no surface waste rock dumps, mill tailings, or dewatering of aquifers. Labor costs for ISR operations are appreciably lower, as fewer workers are required in ISR operations as compared to conventional mining.

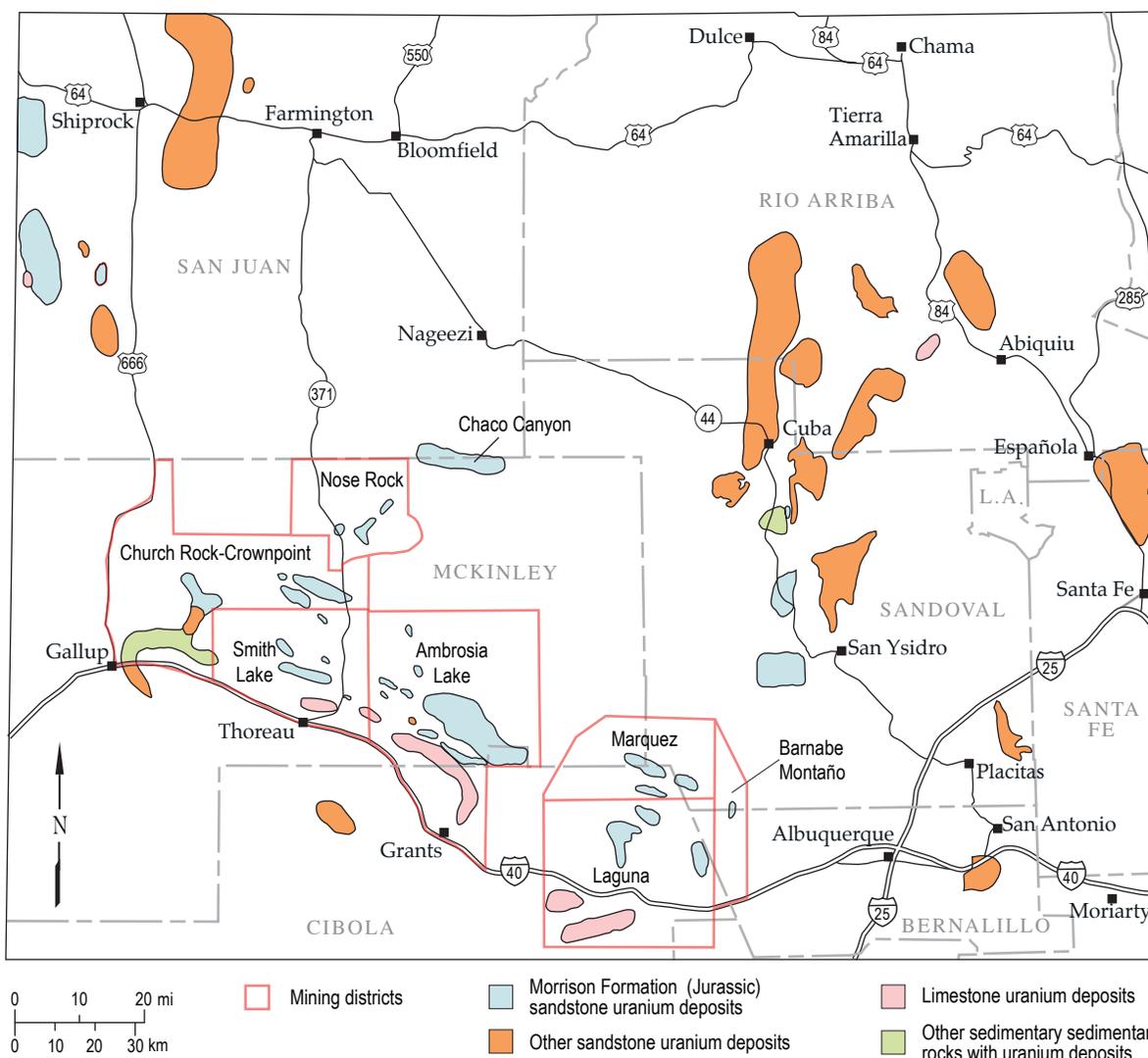


FIGURE 2. Northwestern part of New Mexico showing uranium deposits in the San Juan Basin and subdistricts of the Grants uranium district. Polygons outline approximate areas of known uranium deposits (McLemore and Chenoweth, 1989).

Although ISR recoveries can be lower than those realized from conventional mining, this disadvantage is offset by lower operational, labor, and environmental costs and exploitation of lower-grade and smaller deposits than are economical by conventional mining. For these reasons, it is expected that ISR production will increase in the future. However, there are some other potential issues, which are summarized in this paper.

### Criteria required for successful in situ recovery

Several technical criteria are required for ISR to operate effectively. The uranium deposit must lie beneath the water table in a saturated zone at all times during ISR operations. The uranium host rock should be horizontal or nearly horizontal and confined by less permeable strata, above and below the uranium-bearing unit. The deposit must be permeable and remain permeable throughout the life of the ISR operation. The uranium minerals must be amenable to dissolution by the mild lixiviant chemistry. Other challenges can exist. Some uranium minerals are insoluble or in host rock that lacks permeability and/or, depending on uranium market prices, some may be too deep for commercially viable ISR operations because of well construction costs. Finally, the surface topography must be suitable for placing of multiple injection and recovery wells (well fields). Many of the deposits in the Grants uranium district fulfill many of the above criteria.

## Uranium deposits in New Mexico

The types of uranium deposits in New Mexico are summarized in Table 2, many of which are found in the Grants district. The most important type of deposit in terms of production (Table 2) and resources are sandstone-hosted uranium deposits in the Morrison Formation (Jurassic), which are also the most important type amenable to ISR. More than 340,565,370 lbs of  $U_3O_8$  were produced from the Morrison from 1948 to 2002 (Table 1). The largest ore deposits in the Grants uranium district contain more than 30 million lbs of  $U_3O_8$  each. Other sandstone deposits throughout New Mexico also could have potential for ISR production and are described below. Other types of uranium deposits are described by McLemore (1983, 2007) and McLemore and Chenoweth (1989, 2017).

### Sandstone uranium deposits in the Morrison Formation (Jurassic)

Sandstone uranium deposits account for the vast majority of the historical uranium production from New Mexico (McLemore and Chenoweth, 1989, 2003, 2017). The most significant deposits are those in the Morrison Formation, specifically the Westwater Canyon Member (see McLemore and Chenoweth, 2017, table 3 for production statistics and table 6 for a summary of major uranium deposits and reserves in New Mexico). There

TABLE 1. Uranium production by type of deposit from the San Juan Basin, New Mexico 1947–2002 (McLemore and Chenoweth, 1989, 2003, 2017; production from 1988–2002 estimated by the senior author based upon company annual reports and total yearly production values). Type of deposit refers to Table 3. Total U.S. production from McLemore and Chenoweth (1989) and Energy Information Administration (2010).

Type of Deposit	Production (lbs $U_3O_8$ )	Period of Production (Years)	Production Total in New Mexico (Percent)
Primary, redistributed, remnant sandstone uranium deposits (Morrison Formation, Grants district)	330,453,000 <sup>1</sup>	1951–1988	95.4
Mine water recovery (Morrison Formation, Grants district)	9,635,869	1963–2002	2.4
Tabular sandstone uranium deposits (Morrison Formation, Shiprock district)	493,510	1948–1982	0.1
Other Morrison Formation Sandstone uranium deposits (San Juan Basin)	991	1955–1959	—
Other sandstone uranium deposits (San Juan Basin)	503,279	1952–1970	0.1
Limestone uranium deposits (Todilto Formation <sup>2</sup> predominantly Grants district)	6,671,798	1950–1985	1.9
Other sedimentary rocks with uranium deposits (total NM)	34,889	1952–1970	—
Vein-type uranium deposits (total NM)	226,162	1953–1966	—
Igneous and metamorphic rocks with uranium deposits (total NM)	69	1954–1956	—
Total in New Mexico	348,019,000 <sup>1</sup>	1948–2002	100
Total in United States	927,917,000 <sup>1</sup>	1947–2002	NM is 37.5 of total US

<sup>1</sup>Approximate production figures rounded to the nearest 1,000 pounds. There has been no uranium production from New Mexico since 2002.

<sup>2</sup>Todilto Formation (Cather et al., 2013).

are three types of deposits in the Westwater Canyon Member of the Morrison Formation: primary (trend or tabular), redistributed, and remnant-primary sandstone uranium deposits.

Primary sandstone-hosted uranium deposits, also known as pre-fault, trend, blanket, and black-band ores, are found as blanket-like, roughly parallel ore bodies along trends, mostly in sandstones of the Westwater

Canyon Member. These deposits are characteristically less than 8 ft thick, average more than 0.20%  $U_3O_8$ , and have sharp ore-to-waste boundaries. Primary deposits are high in organic carbon and are known to be difficult to recover by conventional milling techniques and will provide challenges to ISR operations (Holen and Hatchell, 1986).

Redistributed sandstone-hosted uranium deposits, also known as post-fault, stack, secondary, roll-type and

TABLE 2. Classification of uranium deposits in New Mexico (modified from McLemore and Chenoweth, 1989; McLemore, 2001, 2007). Deposit types in **bold** are possibly amenable for ISR operations. <sup>1</sup>Mine identification numbers, prefixed by NM, and district identification numbers, prefixed by DIS are from the New Mexico Mines Database (McLemore et al., 2002, 2005a, 2005b).

Type of Deposit	Example <sup>1</sup>
I. Peneconcordant uranium deposits in sedimentary host rocks	
<b>Morrison Formation (Jurassic) sandstone uranium deposits</b>	
• <b>Primary, tabular sandstone uranium-humate deposits in the Morrison Formation</b>	Roca Honda (NMMK0142)
• <b>Redistributed sandstone uranium deposits in the Morrison Formation</b>	Church Rock (Section 17) (NMMK0034)
• <b>Remnant sandstone uranium deposits in the Morrison Formation</b>	Ruby 3 (NMMK0147)
• Tabular sandstone uranium-vanadium deposits in the Salt Wash and Recapture Members of the Morrison Formation	Enos Johnson 1–4 (NMSJ0047)
<b>Other sandstone uranium deposits</b>	
• <b>Redistributed uranium deposits in the Dakota Sandstone (Cretaceous)</b>	Church Rock (NMMK0034)
• <b>Roll-front sandstone uranium deposits in Cretaceous and Tertiary sandstones</b>	C de Baca (NMSO0515)
• <b>Sedimentary uranium deposits</b>	Boyd (NMSJ0028)
• <b>Sedimentary-copper deposits</b>	Nacimiento (NMSA0064)
• Beach placer sandstone uranium deposits	Sanostee (NMSJ0088)
Limestone uranium deposits	
• Limestone uranium deposits in the Todilto Formation (Jurassic)	Barbara J 2 (NMMK0008)
• Other limestone deposits	Rocky Arroyo (NMED0018)
Other sedimentary rocks with uranium deposits	
• Carbonaceous shale and lignite uranium deposits	Butler Brothers (NMSA0031)
• Surficial uranium deposits	
Calcrete	Lordsburg Mesa (DIS266)
Playa lake deposits	Estancia Salt (DIS243)
II. Fracture-controlled uranium deposits	
Vein-type uranium deposits	
• Rio Grande Rift (RGR) Copper-silver ( $\pm$ uranium) veins (formerly called Jeter-type, low-temperature vein-type uranium deposits and La Bajada-type, low-temperature uranium-base metal vein-type uranium deposits)	Jeter (NMSO0023)
• Collapse-breccia pipes (including clastic plugs)	Woodrow (NMCI0106)
• Volcanic epithermal veins	Union Hill (NMGR0112)
• Polymetallic veins (formerly Laramide veins)	Merry Widow (NMGR0054)
III. Disseminated uranium deposits in igneous and metamorphic rocks	
Igneous and metamorphic rocks with disseminated uranium deposits	
• Metasomatic or metasomatite deposits	Red Rock 1 (NMSI0072)
• Pegmatites	Harding (NMTA0015)
• Alkaline rocks	Pajarito (NMOt0095)
• Granitic rocks	La Cueva prospect (NMTA0559)
• Carbonatites	Lemitar (NMSO0115)
• Caldera-related volcanogenic uranium deposits	San Juan Peak (NMSO0080)
IV. Other potential types of uranium deposits in New Mexico	
• Iron Oxide-Cu-Au (IOCG) (Olympic Dam deposits, hematite breccia deposits)	Possibly Chupadera Mesa (DIS241)
• By-product copper processing	None in NM

roll-front ores, are younger than the primary sandstone-hosted uranium deposits. By definition the uranium has been “redistributed” into the present deposit by a natural process in the past. They are discordant, asymmetrical, irregularly shaped, characteristically more than 8 ft thick, have diffuse ore-to-waste contacts, and cut across sedimentary structures. The average redistributed deposit contains approximately 18.8 million lbs  $U_3O_8$  with an average grade of 0.16%. Some redistributed uranium deposits are vertically stacked along faults. These deposits are not associated with significant amounts of organic material and are generally amenable for ISR.

Remnant sandstone-hosted uranium deposits were preserved entirely within oxidized sandstones after the oxidizing waters that formed redistributed uranium deposits had migrated down dip. Some remnant sandstone-hosted uranium deposits were preserved, because they were surrounded by or found in less permeable sandstone and could not be oxidized as oxidizing groundwaters moved through the aquifer system. These deposits are similar to primary sandstone-hosted uranium deposits, but are more difficult to locate because they occur sporadically within the oxidized sandstone. The average size is approximately 2.7 million lbs  $U_3O_8$  at a grade of 0.20%. These deposits are not associated with significant amounts of organic material, are relatively insoluble and/or hosted by less-permeable sandstones and are generally not amenable to anthropogenic ISR for the same reason they were not subject to natural redistribution processes.

Tabular sandstone uranium-vanadium deposits in the Salt Wash and Recapture Members of the Morrison Formation are restricted to the east Carrizo (including the King Tutt Mesa area) and Chuska Mountains subdistricts of the Shiprock district, western San Juan Basin, where historical production totals 493,510 lbs of  $U_3O_8$  (Table 2). The Salt Wash Member is the basal member of the Morrison Formation in this part of the Colorado Plateau and is overlain by the Brushy Basin Member (Anderson and Lucas, 1992, 1995). It unconformably overlies the Bluff-Summerville Formation, using older stratigraphic nomenclature (Anderson and Lucas, 1992), or the Wanakah Formation as proposed by Condon and Peterson (1986). The Salt Wash Member consists of 190–220 ft of interbedded fluvial sandstones and floodplain mudstones, shales, and siltstones. The mudstone and siltstone make up approximately 5–45% of the total thickness of the unit (Masters et al., 1955; Chenoweth, 1993).

The tabular uranium deposits of the Salt Wash Member are generally elongated parallel to paleostream channels and are associated with carbonized fossil plant material. A cluster of small uranium deposits along a channel trend could contain as much as 4,000 short tons of ore averaging 0.23%  $U_3O_8$  (Hilpert, 1969; McLemore and Chenoweth, 1989, 2017). The deposits tend to form subhorizontal clusters that are elongated and blanket-like. Ore bodies in the King Tutt Mesa area are small and irregular and only a few ore bodies have yielded more than 1,000 lbs of  $U_3O_8$ . A typical ore body in the King Tutt Mesa area is 150–200 ft long, 50–75 ft wide, and approximately 5 ft thick (McLemore and Chenoweth, 1989). The deposits are typically concordant to bedding, although discordant lenses of uranium-vanadium minerals cross-cut bedding planes locally. The ore bodies typically

float in the sandstone; locally, they occur at the interface between sandstone and less permeable shale or siltstone. However, unlike uranium deposits in the Grants district, the deposits at King Tutt Mesa are high in vanadium. The U:V ratio averages 1:10 and ranges 1:1 to 1:16.

It is unlikely that the Salt Wash deposits in New Mexico are amenable for ISR, because most of them are situated above the water table, have low permeability (calcite and gypsum cement and abundant clay minerals), and contain abundant organic material and vanadium that makes recovery difficult. These high carbon ores, which have complex mineralogy (including appreciable vanadium content), are known to be difficult to recover by conventional milling techniques and will provide challenges to ISR operations (Holen and Hatchell, 1986). Furthermore, the lenticular nature of the mineralized sandstone channels will make ISR challenging. However, no pilot studies have been released to determine the solubility or recovery of these deposits.

## Other Sandstone Uranium Deposits

### Redistributed uranium deposits in the Dakota Sandstone (Cretaceous)

A total of 501,169 lbs of  $U_3O_8$  has been produced from redistributed uranium deposits in the Dakota Sandstone in the southern part of the San Juan Basin (Table 2; Chenoweth, 1989). These deposits are similar to redistributed uranium deposits in the Morrison Formation and are found near primary and redistributed deposits in the Morrison Formation. Deposits in the Dakota Sandstone are typically tabular masses that range in size from thin pods a few feet long and wide to masses as much as 2,500 ft long and 1,000 ft wide. The largest known deposits in the Dakota Sandstone are found in the Church Rock mine (NMMK0034, Old Church Rock) in the Church Rock subdistrict of the Grants district, where uranium is associated with a major northeast-trending fault. More than 188,000 lbs of  $U_3O_8$  have been produced from the Dakota Sandstone in the Church Rock mine (Chenoweth, 1989). These deposits are amenable for ISR.

### Roll-front sandstone uranium deposits

Roll-front sandstone uranium deposits are found in the Crevasse Canyon-Baca Formations (Hook Ranch-Riley district), Tesuque Formation (San Jose district) and Ojo Alamo Sandstone (Farmington, Mesa Portales districts) areas in northern New Mexico, where production totals 60 lbs of  $U_3O_8$  (Table 2; McLemore and Chenoweth, 1989). Roll-front uranium deposits typically are found in permeable fluvial channel sandstones and are associated with carbonaceous material, clay galls, sandstone-shale interfaces, and pyrite at an oxidation-reduction interface. Although only a few minor and unverified uranium occurrences have been reported at Mesa Portales (McLemore, 1983); radiometric anomalies are detected by water, stream-sediment, drill logs, and aerial-radiometric studies. Past drilling at Mesa Portales (Fig. 1) indicated that low-grade uranium is found in blanket-like bodies in several horizons. The known mineralization pattern from drill logs suggests that these deposits are modified roll-type ore bodies. These deposits are low grade, but the deeper deposits that are below the water table could be amenable for ISR.

## Sedimentary sandstone uranium deposits

Sedimentary sandstone uranium deposits are stratabound deposits associated with syngenetic organic material or iron oxides, or both, such as at the Boyd deposit (Fruitland Formation) near Farmington and deposits in the Chinle Formation throughout northern New Mexico. Uranium contents vary, but average grades of shipments from these deposits rarely exceeded 0.1% U<sub>3</sub>O<sub>8</sub>. These deposits tend to be small, containing only a few tons of ore, and the potential for future production is low. More information is needed to determine if these deposits have ISR potential.

## Sedimentary-copper deposits

Stratabound, sedimentary-copper deposits containing Cu, Ag, and locally Au, Pb, Zn, U, V, and Mo are found throughout New Mexico. These deposits also have been called “red-bed” or “sandstone” copper deposits by previous workers (Soulé, 1956; Phillips, 1960; Cox and Singer, 1986). They typically occur in bleached gray, pink, green, or tan sandstones, siltstones, shales, and limestones within or marginal to typical thick red-bed sequences of red, brown, purple, or yellow sedimentary rocks deposited in fluvial, deltaic or marginal-marine environments of Pennsylvanian, Permian, or Triassic age (Coyote, Gallina). The majority of sedimentary-copper deposits in New Mexico are found at or near the base of these sediments; some deposits such as those in the Zuni Mountains and Nacimiento districts, are in sedimentary rocks that unconformably overlie mineralized Proterozoic granitic rocks. The mineralized bodies typically form as lenses or blankets of disseminated and/or fracture coatings of copper minerals, predominantly chalcopyrite, chalcocite, malachite, and azurite with minor to trace uranium minerals. Copper and uranium minerals in these sedimentary-copper deposits are commonly associated with organic debris and other carbonaceous material. More information is needed to determine if these deposits have ISR potential.

## History of in situ recovery operations in New Mexico

Several ISR tests and pilot studies have been conducted in New Mexico (Table 3). In addition, more than 9.6 million pounds of U<sub>3</sub>O<sub>8</sub> was produced by mine water recovery from 1963 through 2002 (Table 1), mostly from the Ambrosia Lake, Church Rock, and Smith Lake areas (Table 4). Native groundwater, without any additional additives, was circulated throughout the surface or underground workings and collected for processing.

## Environmental issues

ISR exploitation of uranium deposits provides decided advantages to the environment, in comparison to conventional open pit and underground mining and conventional milling. These advantages include much smaller surface disturbances and shorter duration (allowing the timely return of the surface to traditional land uses), significant reductions of the introduction of radionuclides into the surface environment, and other reduced impacts to local ecosystems. In evaluating the possibilities of developing an ISR mine, it is important to recognize that in the portion of the aquifer in which the uranium deposit is situated, the groundwater prior to ISR operations is not potable because the concentrations of uranium and/or uranium progeny such as <sup>226</sup>Ra and <sup>222</sup>Rn exceed acceptable drinking water standards by a large margin (Pelizza 2014); in other words it is naturally contaminated.

The aforementioned notwithstanding, it is essential that all proposed ISR operations consider certain geological processes as well as undergo rigorous and detailed pre-mining aquifer characterization studies, careful and detailed mineralogical and geochemical studies of the uranium mineralized zones, and comprehensive modeling of the entire hydrologic regime that is based on physical testing and subsequent modeling. Of particular importance are:

TABLE 3. Past pilot and small-scale ISR operations in New Mexico (updated from Holen and Hatchell, 1986). Mine identification number is from the New Mexico Mines Database (McLemore et al., 2005a, b).

Mine Identification Number	Project	Company	Location (latitude, longitude in degrees) <sup>1</sup>	Approximate depth to deposit (feet)	Comments
NMMK0040	Crownpoint ISR—South Trend	Mobil/TVA	35.706678 108.22052	2,000	commercial wellfield that was drilled but never commissioned
NMMK0038	Crownpoint—Section 9	Mobil/TVA	35.71751 108.226809	2,000	1979, successful recovery and ground water restoration
NMMK0109	Monument	Mobil/TVA	35.676444 108.118645	2,000	no information available, may have been a lab test
NMMK0124	Section 13 (Push-pull)	UNC-Teton	35.61626 108.589759	1,300	1980
NMMK0209	Leach Site No. 1 (Section 23)	Grace Nuclear	35.608667 108.602083	500	1975
NMSA0076	Leach Site No. 2 (Section 13)	Grace Nuclear	35.273278 107.205778	380	1975
NMSO0098	Hook Ranch	Grace Nuclear	34.306972 107.424611	50?	Unsuccessful test
NMCI0105	Windwhip	Anaconda	35.142548 107.339932	200–240	1970

<sup>1</sup>Latitudinal, longitudinal in NAD27.

- Mobilization of uranium is part of a broader geochemical process that also mobilizes other elements such as molybdenum and radium, and operational procedures are required to control these constituents during ISR mining and to mitigate their presence after completion of operations (post-closure).
- Study of clay species in the mineralized zones, and their impacts, not only upon porosity and permeability characteristics during uranium extraction, but their geochemical interactions with various elements and compounds during and after groundwater restoration (post-closure).
- Development of detailed hydrological models of the aquifer, relying not only on the results of rigorous aquifer tests, but also thorough incorporation of a detailed geologic model that incorporates all data relating to faults, fractures and joints that could otherwise impact the management of lixivants during mining.
- During ISR operations, slightly higher production rates are maintained within the wellfields to help ensure none of the lixiviant migrates from the mining area. Proper disposal of this “bleed water” during mining and the larger quantity of water that is produced during the restoration process.
- Thorough and honest communication with the public and regulators.

While the environmental, technological and operational applications of ISR mining of uranium have advanced appreciably since the time of ISR pilot test programs in New Mexico, these important environmental issues continue to require the attention of mine operators and regulators alike.

The mineralogical, chemical, hydrological, and physical parameters of the aquifer are characterized well before ISR, but these parameters are also important to characterize during and even after ISR is completed. The process dissolves not only uranium, but other minerals, which could alter the geochemistry of the aquifer, and

affect recovery of the lixiviant as well as the composition of groundwater. Precipitation of minerals such as gypsum and calcite can occur during the process that could seal the well bore, affect recoveries and potentially impact future use of the aquifer. Complex reactions with clay minerals also can lead to changes in aquifer conditions. Changes in mineralogy of the host sandstone during and after ISR should be monitored using the most advanced geochemical techniques that are available.

In the Grants district, structural discontinuities (faults, folds, pinch-outs of beds) are visible during open-pit and underground mining that are not observed on the surface or by exploration drilling. These structural discontinuities are encountered in ISR, but the effects of these discontinuities may be difficult to plan for in during ISR wellfield development because in the U.S., most exploration drilling is conducted at 100-ft centers, which typically detects and characterizes many, but not all of these features. For this reason, during ISR wellfield development, delineation drilling is conducted in advance of well installation where drill spacing is generally reduced to 50 ft or less. It is important that the project wellfield development geological staff pay particularly close attention to structural and stratigraphic changes that may not have become apparent during the wider spaced exploration drilling program to assure the best wellfield design and optimal ISR performance.

Most ISR operations are relatively shallow, less than 500 ft deep, but many of the Grants uranium deposits are deeper than 1,000 ft, with some deposits as much as 4,000 ft deep. Depending on uranium market prices, some of the remaining Grants deposits may be too deep for commercially viable ISR operations because of well construction costs and irregular surface topography. Despite these difficulties, ISR could be a viable alternative to the conventional mining of uranium and can be used in the Grants district with proper advanced geological and aquifer characterization and monitoring by the ISL companies, all which are evaluated during the permitting process by state and federal regulatory agencies. Every location should be evaluated, tested and reviewed by regulators on a case by case basis.

## Summary

Sandstone-hosted uranium deposits in New Mexico have played a major role in global historical uranium

TABLE 4. Mine water recovery from stope leaching (modified from Holen and Hatchell, 1986). Mine identification number is from the New Mexico Mines Database (McLemore et al., 2005a, b).

Project	Company	Mine (Mine Id)
Ambrosia Lake	United Nuclear	Section 27 (NMMK0226), Ann Lee (NMMK0003), Sandstone (NMMK0149)
Church Rock	United Nuclear	Northeast Church Rock (NMMK0117), Old Church Rock (NMMK0034)
Ambrosia Lake	Kerr-McGee (Quivira)	Sections 17, 19, 22, 24, 30, 30 West, 33, 35 (NMMK0191, NMMK0199, NMMK0206, NMMK0213, NMMK0237, NMMK0236, NMMK0250)
Church Rock	Kerr-McGee (Quivira)	NE Church Rock 1 (NMMK0112)
Ambrosia Lake	United Nuclear-Homestake Partners (later Homestake, now Quivira)	Sections 15, 23, 25, 32 (NMMK0183, NMMK0208, NMMK0216, NMMK0244)
Smith Lake	Gulf	Mariano Lake (NMMK0102)
San Mateo	Gulf (Chevron)	Mount Taylor (NMC0027)
Seboyeta	Sohio-Western	L Bar (NMC0019)

production. Although some other types of uranium deposits in the world are higher in grade and larger in tonnage, the Grants uranium district could become a significant source of uranium because as ISR technologies improve, production costs decrease. However, several challenges need to be addressed by companies before uranium could be produced once again from the Grants uranium district, especially by ISR, and these can be planned for during the permitting process:

- Permitting for new ISR and especially for conventional mines and mills will take years to complete at significant up-front costs.
- Geological and technical issues need to be resolved on a deposit by deposit basis.
- In the United States, closure plans, including reclamation, are developed during the permitting process, prior to the commencement of mining or ISR activities. Modern regulatory costs will add to the cost of producing uranium in the U.S.

- Some communities, especially the Navajo Nation communities, do not view development of uranium properties as favorable. The Navajo Nation has declared that no uranium production will occur on Navajo lands.
- High-grade, low-cost uranium deposits in Canada and Australia are sufficient to meet a portion of today's demands; however, additional resources, possibly including those in New Mexico, will be required to meet long-term future requirements.

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# Investigation of in situ leach (ISL) mining of uranium in New Mexico and post-mining reclamation

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## Abstract

The purpose of this study was to consider the effectiveness of two methods of restoring groundwater quality in a subsurface uranium-bearing formation following in situ leach (ISL) mining. To accomplish this it was necessary to develop an understanding of the geochemical characteristics of an aqueous solution that might be produced by an ISL mine. Samples of material from three different uranium (U) mines were collected and their acid leachable elemental concentration determined. Additional samples were then leached with aerated sodium bicarbonate ( $\text{NaHCO}_3$ ) solutions at concentrations ranging from 1 mM to 500 mM. The fraction of the acid-leachable U and other trace elements released by this leaching process depended on  $\text{NaHCO}_3$  concentration, U mineralogy, and the amount of solid organic matter in the samples. Less than 5% U was leached from samples with high organic matter using the  $\text{NaHCO}_3$  leach solution. Groundwater restoration methods were then investigated using column experiments. Two methods were evaluated, a chemical stabilization method based on addition of phosphate ( $\text{PO}_4$ ) and a microbial method in which lactate was added to stimulate growth of dissimilatory sulfate and metal reducing organisms. Neither method was effective. This was believed to be due to sweeping of the leach solution from the columns by the phosphate- or lactate-amended solutions. This hypothesis is consistent with limited mixing in an aquifer as a result of plug flow through the formation.

## Introduction

New Mexico has played a major role in the production of uranium (U) for the nuclear power industry and the nation's weapons programs, producing more than 340 M lbs of yellowcake ( $\text{U}_3\text{O}_8$ ) (McLemore et al., 2013). Though the U mining and milling industry in New Mexico is inactive at present, increased interest in nuclear energy as a  $\text{CO}_2$ -free power source has led to renewed interest in development of U resources. Recently, projects have been proposed using both conventional underground mining and in situ leach (ISL) mining.

Currently, nearly all United States U production is from ISL mines. In 2015, total U.S. production was 3.3 M lbs of  $\text{U}_3\text{O}_8$  from one underground mine and six ISL mines (U.S. Energy Information Administration 2016). The ISL mines are located in Nebraska, Texas, and Wyoming. Note that the domestic U industry continues to struggle as reflected by the fact that total production in 2015 was 32% less than in 2014 (U.S. Energy Information Administration, 2016).

ISL mining, also referred to as in situ recovery (ISR) mining, is accomplished by using a system of injection and extraction wells to circulate a solution or "lixiviant," through the subsurface ore formation (U.S. Nuclear Regulatory Commission, 2009). The lixiviant typically consists of an oxidant (oxygen or hydrogen

peroxide) and a carbonate-complexing agent. It is injected into the aquifer to solubilize U by oxidizing it to more soluble phases, which then dissolve through a series of complexation reactions. The soluble U complexes are transported by groundwater flow to the extraction wells where they are pumped to the surface and are recovered in a mill. The barren lixiviant is then pumped back into the aquifer through injection wells and the process is repeated.

The major advantages of ISL mining are that (U.S. Nuclear Regulatory Commission, 2009): 1) it produces little disturbance of the land surface, 2) construction costs are much less than conventional underground mining, 3) it is much safer for workers than underground mining, 4) very little solid waste is generated because there is virtually no excavation, and 5) there is little impact on groundwater resources due to mine dewatering. Often these advantages result in significant cost savings.

However, there are also notable limitations of ISL mining. The ore deposit must be below the water table; the hydrogeology of the host formation must be amenable to circulation of lixiviant solutions through it; and the U minerals must be reactive with the leach solutions. Though ISL mining has been extensively practiced in other states, there is little experience with it in New Mexico.

A major challenge facing the ISL mining industry is that groundwater must be returned to acceptable quality at the conclusion of mining. ISL mining alters the geochemistry of the ore body resulting in release of other constituents at concentrations that may exceed state groundwater standards and federal drinking water standards. In New Mexico, aquifer restoration is especially important because groundwater may be the sole water supply for drinking and other uses, such as in the Grants Mining District.

The principal objective of this research was to evaluate two methods of stabilizing aqueous geochemistry and of restoring groundwater quality following ISL mining. A two-phase laboratory investigation was conducted in which different leach solutions were used to evaluate extraction of U from several low-grade ore samples. In contrast to other studies, which were done in batch systems, this investigation was performed using columns filled with U ore to better simulate ISL conditions. The purpose was to generate information regarding the chemistry of fluids that would be produced by ISL mining of NM U ores. The second phase of the investigation consisted of a series of column studies to evaluate two strategies for restoring groundwater quality. One method was based on chemical addition to stabilize minerals that release U and other constituents. This involved circulation of a phosphate ( $\text{PO}_4$ ) solution. The second method was based on re-establishing reducing conditions in the aquifer by stimulating growth of anaerobic microorganisms.

## Background and theoretical considerations

This section provides a brief summary of the theoretical considerations associated with ISL U mining in order to establish a fundamental basis for the research described in this paper. This summary also includes a brief overview of ISL mining and previous groundwater restoration studies.

### Geochemistry of uranium and co-constituents

In the natural environment, U most commonly occurs in two oxidation states, U(IV) and U(VI). Uranium geochemistry strongly depends on its oxidation state and the presence of inorganic complexants, especially carbonate ( $\text{CO}_3$ ). Its geochemistry can be conveniently summarized in a pe-pH diagram (also referred to as an Eh-pH diagram) in which the oxidation-reduction (redox) conditions of the solution are plotted on the vertical axis and acid-base conditions are plotted on the horizontal axis (Langmuir, 1997). A pe-pH diagram for U is presented in Figure 1. This diagram represents U-equilibrium chemistry in a solution containing  $10^{-6}$  M U and  $10^{-3}$  M dissolved  $\text{CO}_2$  at  $25^\circ\text{C}$ . The diagram shows that under oxidizing conditions, U(VI) is thermodynamically stable, and its speciation depends on the pH of the solution. Below pH 5, the cationic uranyl ion ( $\text{UO}_2^{2+}$ ) is present, while at higher pH the uranyl ion forms soluble mono-, di-, and tri-carbonate complexes. These complexes are soluble. Ternary complexes of uranyl with calcium ( $\text{Ca}^{2+}$ )

and carbonate ( $\text{CO}_3^{2-}$ ) near neutral pH have also been reported (Dong and Brooks 2006), but for the sake of simplicity are not shown on this diagram. Because most soil minerals have net negative surface charges above pH  $\sim 5$ , uranyl-carbonate complexes do not interact with soil mineral surfaces and therefore are readily transported by groundwater through aquifer materials. In contrast, U(IV) forms insoluble precipitates; uraninite ( $\text{UO}_2(\text{s})$ ) and coffinite ( $\text{USiO}_4(\text{s})$ ) are shown in this diagram. Deep ore deposits often are dominated by reduced U phases of which nearly 100 different minerals are known (De Voto, 1978; Burns and Finch, 1999).

Extracting insoluble U from ore generally involves oxidation followed by dissolution whether in a conventional mill or in an ISL mine (Thomson and Heggen, 1983). In a conventional mill, ore is pulverized then leached with an oxygenated strong acid such as sulfuric acid or a strong basic carbonate solution. The dissolved U is then recovered from solution by ion exchange (IX) or solvent extraction. In an ISL mine an oxygenated carbonate solution is circulated through the formation to oxidize, complex and dissolve the U, which is then recovered at the surface by IX. The geochemical reactions in the ISL process have been presented elsewhere (Davis and Curtis, 2007).

Several other metals and metalloids have redox chemistry similar to U, including arsenic (As), molybdenum (Mo), selenium (Se), and vanadium (V). Accordingly, it is common to find one or more of these elements present

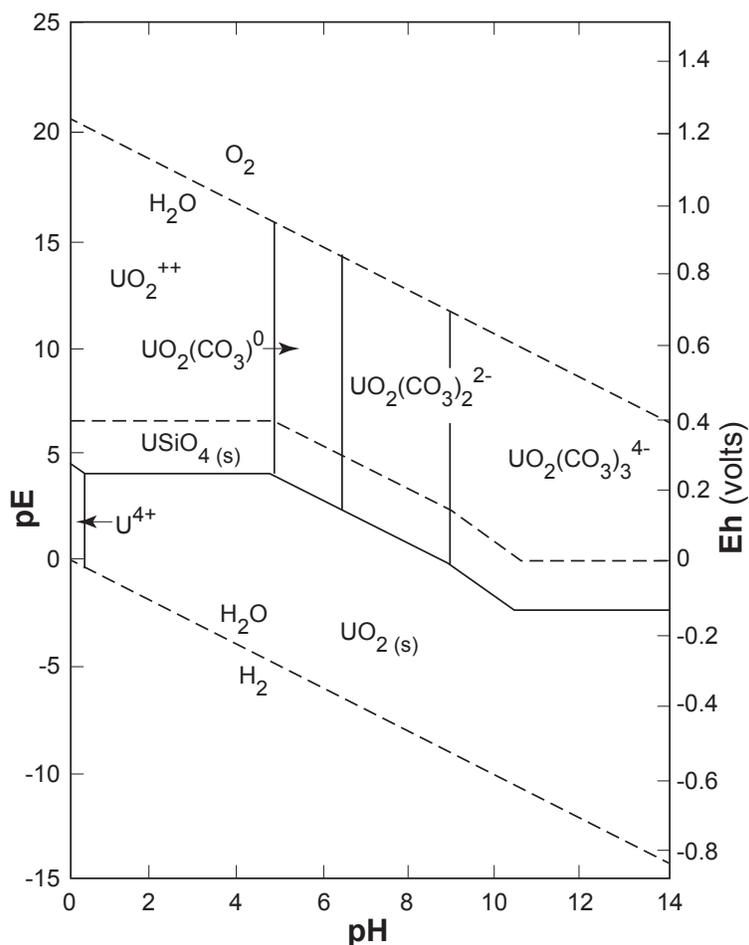


Figure 1. pe-pH diagram for U for a closed system where the total U concentration is  $10^{-6}$  M and the total dissolved  $\text{CO}_2$  and  $\text{H}_4\text{SiO}_4$  concentrations are  $10^{-3}$  M (diagram prepared by Thomson after Langmuir, 1997).

in U-bearing formations or as mixed U minerals such as carnotite ( $K_2(UO_2)_2(VO_4)_2 \cdot 1-3H_2O$ ) or tyuyamunite ( $Ca(UO_2)_2(VO_4)_2 \cdot 5-8H_2O$ ). This is why very high concentrations of these constituents are frequently found in U mill-tailing solutions (Thomson and Heggen, 1983).

### ISL mining of uranium

It is necessary to understand the ISL process and aquifer geochemistry prior to considering groundwater restoration methods. Conditions that are required for successful ISL mining include (U.S. Nuclear Regulatory Commission, 2009; Maerten, 2013): 1) the ore body must be located below the water table, 2) the deposit geometry must be of sufficient size and shape to allow circulation of lixiviant through injection and extraction wells, 3) the host rock must be sufficiently permeable to allow the lixiviant to pass through the formation, and 4) the formation must be confined so that lixiviant flow can be contained and directed by injection and extraction wells, and, perhaps most importantly, 5) the mineralogy must be such that the U can be extracted from the ore material by the relatively benign oxidants and complexants used in ISL mining.

A conceptual diagram of the ISL process is presented in Figure 2. Barren lixiviant (i.e., a solution containing no dissolved U) is injected, passes through the ore formation causing oxidation and dissolution of U minerals, and soluble U is recovered in an extraction well. Uranium is removed from the lixiviant by IX, and the U-depleted solution is recirculated back through the formation.

The U.S. Energy Information Agency (2016) lists seven ISL U mines that were operating at the end of 2015, one U mine in standby status, eight in the permitting and licensing process, and three in the restoration/reclamation process. These mines are located in Nebraska (1), New Mexico (2), South Dakota (1), Texas (7), and Wyoming (8). The two proposed New Mexico mines are the Church

Rock and Crownpoint mines proposed by HydroResources Inc., both with a planned capacity of 1 M lbs/yr of  $U_3O_8$ .

Though commercial U production by ISL mining has not been done in NM, a field-scale pilot study was performed beginning in 1979 near Crownpoint, NM. The project was conducted by Mobil Oil and consisted of four, five-spot well clusters drilled to approximately 2,000 ft with 100 ft spacing (Uranium Producers of America, 1995). The purpose of the pilot test was to evaluate the ability to extract U by ISL mining and then to evaluate groundwater restoration methods following mining. The latter was especially important because of the excellent background groundwater quality that met drinking water standards. Further, the aquifer is the sole source of water supply for the community of Crownpoint located approximately 5 miles from the proposed mine site.

The Crownpoint pilot test consisted of leaching for 10 months followed by restoration activities over a period of 12 months (Uranium Producers of America, 1995). Leaching was performed with a solution of hydrogen peroxide ( $H_2O_2$ ) and sodium bicarbonate ( $NaHCO_3$ ) at a total dissolved  $CO_2$  concentration of 1,500 to 2,000 mg/L and pH 8.3. The pilot test produced maximum U concentrations of approximately 1,000 mg/L but also Mo concentrations of 100 to 200 mg/L. Due to the short duration of the pilot test, Vogt et al. (1982) estimated that only 15% of the U in the ore body was recovered; however, this was not confirmed. Aquifer restoration methods were investigated after completing testing of ISL mining. The restoration method involved flushing the ore body with formation water that had been treated by softening, IX and then reverse osmosis (RO) to remove contaminants. An estimated 11.3 pore volumes of water were passed through the ore body. By the end of the restoration test all of the regulated constituents were below New Mexico groundwater standards and most were near background levels except for pH, Mo, U, and  $^{226}Ra/^{228}Ra$ .

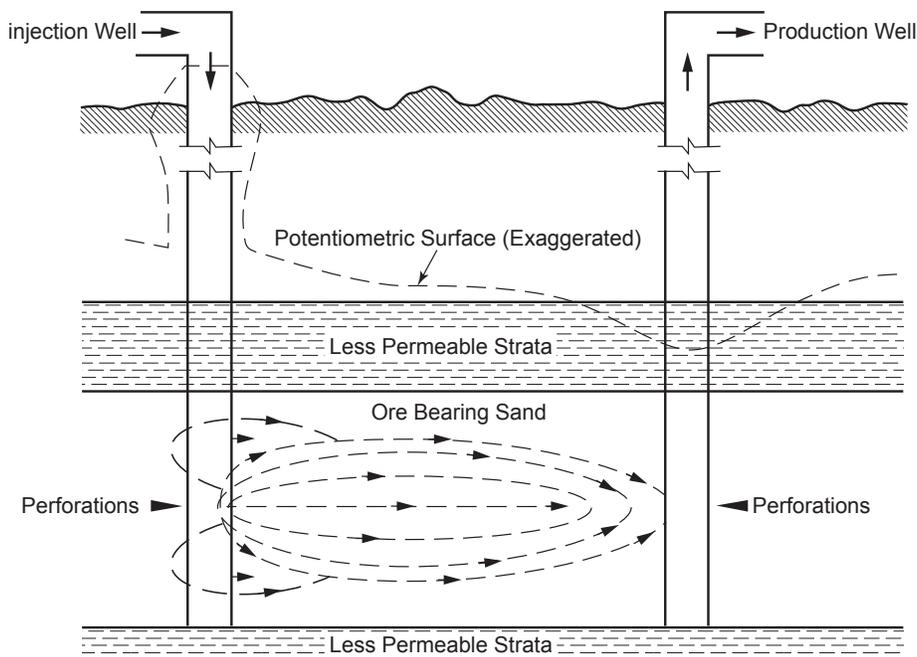


Figure 2. Conceptual diagram of the in situ leach (ISL) mining process (U.S. Nuclear Regulatory Commission, 2009).

## Groundwater restoration following ISL mining

Four methods of groundwater restoration following ISL mining have been identified by the U.S. Nuclear Regulatory Commission (2009): groundwater transfer, groundwater sweep, RO permeate recirculation, and in situ stabilization. The International Atomic Energy Agency (IAEA) also includes natural attenuation as an alternative; however, this approach is unlikely to be acceptable in New Mexico where U-bearing aquifers are the sole source of water supply, so this strategy is not discussed here (IAEA, 2001). Groundwater transfer involves moving groundwater from a depleted ore body to a nearby formation undergoing mining so that uncontaminated water dilutes and transports contaminants through the depleted formation. Groundwater sweep consists of pumping water from all ISL wells, both injection and extraction, without injection. Uncontaminated native groundwater then sweeps through the depleted formation. Recirculation of RO treated groundwater involves pumping contaminated water from the mine, treating by RO at the surface to remove contaminants, then reinjecting and recirculating the water back through the mine. The recirculation pattern can be in either a forward or a reverse circulation pattern (Charbeneau, 1984). In situ stabilization consists of altering subsurface geochemical conditions to achieve immobilization of constituents released by the ISL process. Methods involving both chemical addition and stimulation of microbial growth to achieve biological reduction have been proposed (Davis and Curtis, 2007; Gallegos et al., 2015).

Investigations of chemical methods to restore groundwater quality have focused on addition of phosphate ( $\text{PO}_4$ ) (Arey et al., 1999) and sulfide ( $\text{S}^{2-}$ ) (Borch et al., 2012). Phosphate addition as a restoration method is based on the formation of insoluble U(VI)- $\text{PO}_4$  phases. Arey et al. (1999) investigated the addition of hydroxylapatite ( $\text{Ca}_5(\text{PO}_4)_3\text{OH}$ ) as a restoration method by performing batch tests followed by sequential extraction. Apatite addition reduced U solubility, presumably by precipitation of secondary  $\text{PO}_4$  phases. The presence of aluminum (Al) and iron (Fe) in the phosphate phases was noted. Residual U concentrations were higher in groundwater samples with higher dissolved organic carbon, presumably due to complexation reactions. Sulfide was considered for aquifer stabilization because it is both a strong reducing agent and will form insoluble precipitates with many transition metals. However, addition of a sulfide to a depleted ISL mine following groundwater sweep resulted in little attenuation of U, Fe, or Mn (Borch et al., 2012). It was thought that formation of strong U-carbonate complexes was responsible for poor U stabilization.

Use of microbial reduction to stabilize a depleted U formation is based on the premise that U and its frequent co-constituents of Mo, Se, and V are insoluble under reducing conditions. Microbial reduction can be achieved through stimulation of growth of dissimilatory sulfate and metal reducing organisms under anaerobic conditions. Two recent reviews of the microbiology and geochemistry have been provided by Williams et al. (2013) and Gallegos et al. (2015). The study by Gallegos et al. (2015) was of particular relevance as it involved collection of core samples from a mined U sandstone deposit in Wyoming and analysis of the mineralogy and microbial population. The

study found residual U(IV) associated with organic-rich deposits that was resistant to oxidation and dissolution by ISL lixiviants. Much of the residual U(VI) was associated with Fe(III) minerals. This finding suggests that microbial reduction may present a short-term restoration challenge because adsorbed U(VI) species would become soluble if Fe(III) phases were reduced to more soluble Fe(II) species. In the long term residual reductants such as pyrite and organic carbon may provide long-term immobilization of contaminants from ISL mining.

## Methods

The objective of this study was to investigate chemical and microbial stabilization of U and co-constituents following ISL mining. The investigation consisted of two phases. The first phase involved sample collection and leaching experiments to determine leachability of U and to develop an estimate of ISL lixiviant chemistry. The second phase consisted of scoping experiments to simulate in situ restoration by chemical and microbial methods. In contrast to other studies, which were done in batch systems, this investigation was performed using columns filled with U ore to better simulate ISL conditions.

### Sample collection and characterization

Core samples of undisturbed underground ore weren't available, hence U-containing rock samples from three different sources were used: 1) stockpiled low-grade ore and waste rock from the Section 11/12 mine near Ambrosia Lake, 2) exposed beds from a mineralized zone at the Jackpile open pit mine on the Laguna Pueblo, and 3) stockpiled ore samples from the Mt. Taylor mine near San Mateo, NM. Several kilograms of samples were collected at each site. The Jackpile samples consisted of three samples from a mineralized zone with distinctive black, gray and yellow colors. The samples were broken using a stainless steel weight and then sieved. They were not pulverized in order to avoid fracturing the fine-grained minerals and unnecessarily exposing internal surfaces to leaching solutions. Particles between 0.425 mm and 0.075 mm were used in batch experiments. Particles between 2.00 mm and 0.425 mm were used in subsequent column experiments. The mineralogy of the samples was not determined.

The acid-extractable elemental composition was determined by leaching samples with aqua regia (three parts HCl and one part  $\text{HNO}_3$ ) for 12 hours, heating at 90°C for 3 hours, then analyzing the extract for major and trace elements. A PerkinElmer Optima 5300 DV inductively coupled plasma-optical emission spectrometer (ICP-OES) was used for general metal analyses and a PerkinElmer Optima NexION 300D inductively coupled plasma-mass spectrometer (ICP-MS) was used to measure U and other trace constituents. Anions were measured by ion chromatography using a Dionex ICS-1100 ion chromatograph equipped with a Dionex Ion Pac AS9-HC column. The organic fraction of the samples was determined by Loss on Ignition (LOI) analysis. This procedure involves drying the samples at 105°C, weighing to determine moisture content, then ashing the samples at 550°C and re-weighing. The difference in weight is assumed to be the loss of the organic solids, which are oxidized to  $\text{CO}_{2(g)}$ .

## Leaching experiments

Batch leaching experiments were conducted before running the column tests to select an appropriate bicarbonate lixiviant for subsequent column tests and to anticipate the concentrations of constituents in the leachate. The column tests were performed to simulate the ISL leach process and to collect information about the water quality produced by ISL mining. These columns were subsequently subjected to stabilization methods; one set of columns was treated with PO<sub>4</sub> solution, and a second set of columns received addition of lactic acid in order to simulate microbial reduction.

Batch leaching experiments were conducted using sodium bicarbonate (NaHCO<sub>3</sub>) solutions of varying concentrations to determine the U leachability of the different ore samples. Deionized water was used to prepare NaHCO<sub>3</sub> solutions of 1, 10, and 500 mM concentrations for these tests. Samples were leached in loosely capped plastic bottles to allow for oxygenation for 120 hours. Aliquots were collected periodically during the leach experiments to determine leaching kinetics. Samples were filtered through 0.45 μm membrane filters and preserved with HNO<sub>3</sub> to pH < 2 prior to elemental analysis.

Column leach experiments were performed in 5 cm x 25 cm Plexiglas® columns packed with ore samples from the Mt. Taylor mine with particle sizes between #10 (2.0 mm) and #40 (0.425 mm) sieve sizes. The columns had 1 cm of inert glass wool at each end to contain the crushed ore and to distribute flow transversely. Five columns were used; one to serve as a control, and two each to provide duplicates for chemical and microbial stabilization experiments. All columns were operated in an upflow direction and were sealed to prevent introduction of air. The columns were first fed a simulated groundwater solution (see Table 1) with major ion chemistry similar to that reported at the Crownpoint, NM, ISL pilot test (Uranium Producers of America, 1995). Other major ions commonly found in water were present at low concentrations, hence were not included in this recipe. The pH of the synthetic groundwater was 8.3. It was aerated by shaking each time solution was added to the columns to assure a well-oxidized solution.

Groundwater-flow velocities in an ISL mine are very low such that one pore volume of fluid might be replaced every few weeks or longer. It was not feasible to pump water continuously through the columns at such a slow velocity so instead they were fed discontinuously by feeding one pore volume of leach solution every two days for the chemical restoration experiments, and one pore volume every seven days for the microbial restoration experiments. The column studies began by passing one pore volume of the leach solution through each column and then allowing them to equilibrate for 11 days. Restoration experiments began after this equilibration period.

TABLE 1. Chemistry of synthetic groundwater used in column experiments.

Constituent	Concentration (mM)
Ca <sup>2+</sup>	0.3
Na <sup>+</sup>	4.8
HCO <sub>3</sub> <sup>-</sup>	2.6
SO <sub>4</sub> <sup>2-</sup>	0.4
Cl <sup>-</sup>	0.7

Two types of restoration methods were simulated: chemical restoration using a PO<sub>4</sub> solution and biological stabilization achieved by stimulation of reducing microorganisms. The feed water solution consisted of synthetic groundwater (Table 1) with addition of PO<sub>4</sub> or lactate. Phosphate was added in the form of mono-basic phosphate (NaH<sub>2</sub>PO<sub>4</sub>) at a concentration of 100 μM (10 mg/L) of PO<sub>4</sub>, a concentration similar to that used in previous studies (Arey et al., 1999; Mehta et al., 2016). Sodium lactate (NaC<sub>3</sub>H<sub>5</sub>O<sub>3</sub>) at a concentration of 3 mM (270 mg/L) was used to stimulate growth of naturally occurring, anaerobic, sulfate- and metal-reducing organisms.

During the ISL restoration experiments, synthetic groundwater containing PO<sub>4</sub> was fed discontinuously to the columns at the rate of one pore volume every two days, as stated above. Because microbial growth is much slower than the precipitation reactions expected with PO<sub>4</sub> addition, the feed rate for the synthetic groundwater amended with lactate was one pore volume every seven days. A control column containing U ore was leached with synthetic groundwater to serve as a reference for comparison of results.

Samples of effluent from each column were collected each time the columns were fed. The samples were filtered and split, with half of the sample preserved with HNO<sub>3</sub> for metals analysis and the other refrigerated with no preservation for analysis of non-metals.

## Results and Discussion

Concentrations of elements of interest extracted from the ore samples used in this study are summarized in Table 2. These elements were extracted from crushed ore by acid digestion. The concentration of U, other trace elements, and organic matter varied widely in the five samples. The highest U concentration was in the samples from the Mt. Taylor mine which is consistent with the fact that they were collected by mine staff from high U content ore exposed in the underground mine. Organic material in the samples was not characterized by any method other than LOI.

A simple aerobic batch leaching experiment was conducted to determine the leachability of the elements of interest using NaHCO<sub>3</sub> concentrations ranging from 1 mM

TABLE 2. Concentration of acid extractable elements of interest and organic matter as measured by loss on ignition in samples used in this study.

Sample	Mo (mg/kg)	Se (mg/kg)	U (mg/kg)	V (mg/kg)	LOI* (%)
Grants (Section 11/12 Mine)	2.54	12.3	1281	42.5	1.95
Mt. Taylor Mine	90.5	8.44	10767	512	3.99
Laguna Mine-Black	0.70	0.00	7602	24.0	21.8
Laguna Mine-Grey	0.00	0.00	1050	50.7	0.56
Laguna Mine-Yellow	1.20	0.00	38.4	0.00	1.74

\*LOI = Loss on ignition

to 500 mM, concentrations typical of those that might be used in an ISL mine. The percentage of the total acid-extractable U leached by each  $\text{NaHCO}_3$  solution depended in part on the U concentration and the presence of organic matter in the sample. This is illustrated in Figure 3 in which very little of the total U was leached from the Laguna Mine–Black sample which had very high organic carbon content. Because of this poor leachability, all subsequent testing was performed with the ore sample from the Mt. Taylor Mine.

The leachability of U and other constituents also depends on the  $\text{NaHCO}_3$  concentration of the lixiviant as shown in Figure 4 for the Section 11/12 sample. It is notable that less than 30% of the total acid extractable U was leached. Leach kinetic data not presented in this paper showed that leaching by the bicarbonate solutions

occurred rapidly and typically reached >90% of the final value within 24 hours (Ruiz Lopez, 2016). This suggests that the bicarbonate leaching only removed readily soluble forms of U and other elements and that little or no mineral oxidation occurred over the five-day batch leach process. It is also notable that a high fraction of Mo was released in the leaching experiments with comparatively small fractions of the other elements. This suggests that recovery of Mo may be feasible for some ISL U mines. It also indicates that Mo may present a challenge for groundwater restoration following ISL mining as was found during the Crownpoint pilot test (Uranium Producers of America, 1995).

The batch leach tests were performed to determine the appropriate  $\text{NaHCO}_3$  concentration for the subsequent column tests and to anticipate the solute concentrations in the column tests. Based on the batch tests, 50 mM  $\text{NaHCO}_3$

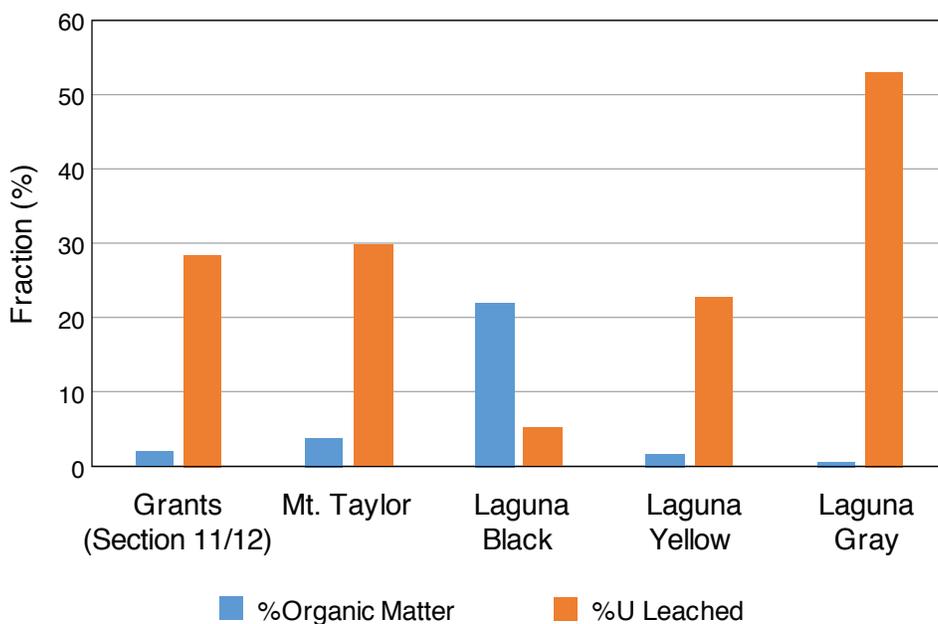


Figure 3. Fraction (%) of total U released by a 500 mM  $\text{NaHCO}_3$  solution and fraction (%) of organic matter measured as LOI in different samples

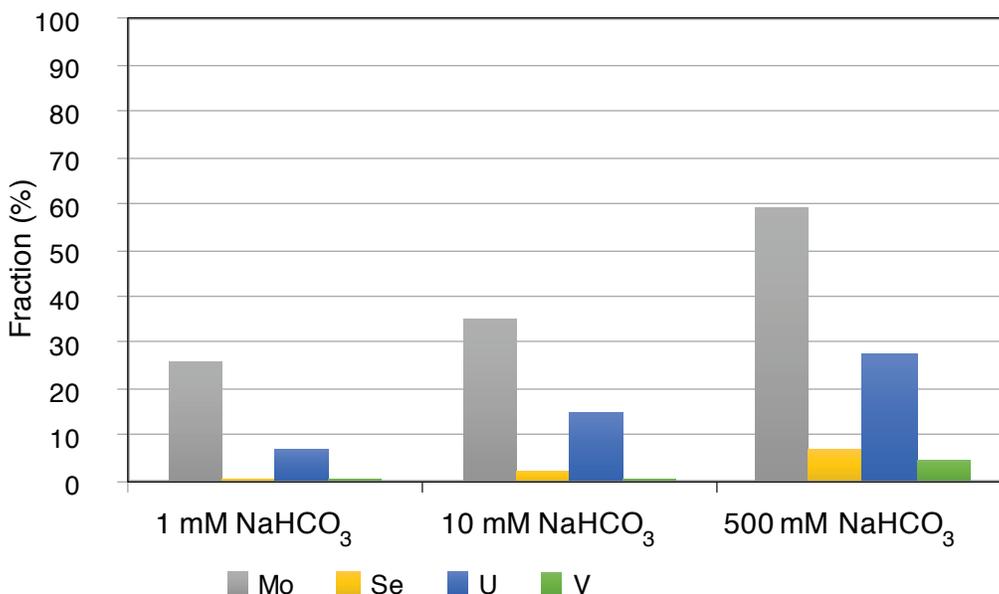


Figure 4. Fraction (%) of metals leached from the Grants (Section 11/12) mine sample after five days of leaching with different concentrations of  $\text{NaHCO}_3$

was added to the synthetic groundwater (Table 1) to serve as a leachate. This concentration was chosen based on the results of the batch tests and is similar to that used in practice. The columns all contained Mt. Taylor ore samples, which were allowed to equilibrate with the leach solution for 11 days. The average quality of the leachate at the end of the equilibration period is shown in Table 3. The extremely high U concentration is almost certainly due to the high concentration of U in the Mt. Taylor samples, approximately 1% (Table 2). Although 50 mM of NaHCO<sub>3</sub> was added to the leach solution corresponding to an alkalinity of 52.6 meq/L, the measured alkalinity of the leachate from the columns was roughly half that. The decrease in alkalinity and the relatively low initial pH are believed to be due to acid produced by partial oxidation of sulfide minerals, such as pyrite (FeS<sub>2</sub>), resulting from weathering reactions that occurred in the stockpiled ore.

Aquifer restoration experiments began in the sealed columns after the 11-day equilibration period. The results

of the restoration process using the PO<sub>4</sub> addition are summarized in Figure 5, which plots U concentration and pH in the column effluent versus pore volumes of PO<sub>4</sub>-amended synthetic groundwater fed to each column. The figure presents data for the two replicate columns (C1 and C2) and for the control column (CC) which was fed only with synthetic groundwater to simulate restoration by groundwater sweeping. The U concentration decayed asymptotically to below 20 mg/L while the pH climbed from an initial pH of 6.4 to about 7.5.

The results of the microbial restoration process in which lactate was used to stimulate growth of anaerobic microbial populations are summarized in Figure 6. The figure presents data for two replicate columns (C3 and C4) and a control column (CC) leached only with synthetic groundwater to simulate restoration by groundwater sweeping. The U concentration decayed asymptotically to below 20 mg/L while the pH climbed from an initial pH of 6.4 to about 7.5. For all of the columns it is suggested

TABLE 3. Average concentration of cations, anions, and metals in column leachate prior to starting stabilization tests. Solution pH = 6.4.

Constituent		Constituent		Constituent	
Major Cations	Conc (mg/L)	Major Anions	Conc (mg/L)	Trace Metals	Conc (mg/L)
Ca <sup>2+</sup>	198	Cl <sup>-</sup>	17.3	Mo	8.87
K <sup>+</sup>	9.9	SO <sub>4</sub> <sup>2-</sup>	257	Se	.03
Mg <sup>2+</sup>	12.8	Total Alkalinity <sup>1</sup>	1085	U	1627
Na <sup>+</sup>	376			V	0.69

<sup>1</sup>Units of mg/L CaCO<sub>3</sub>

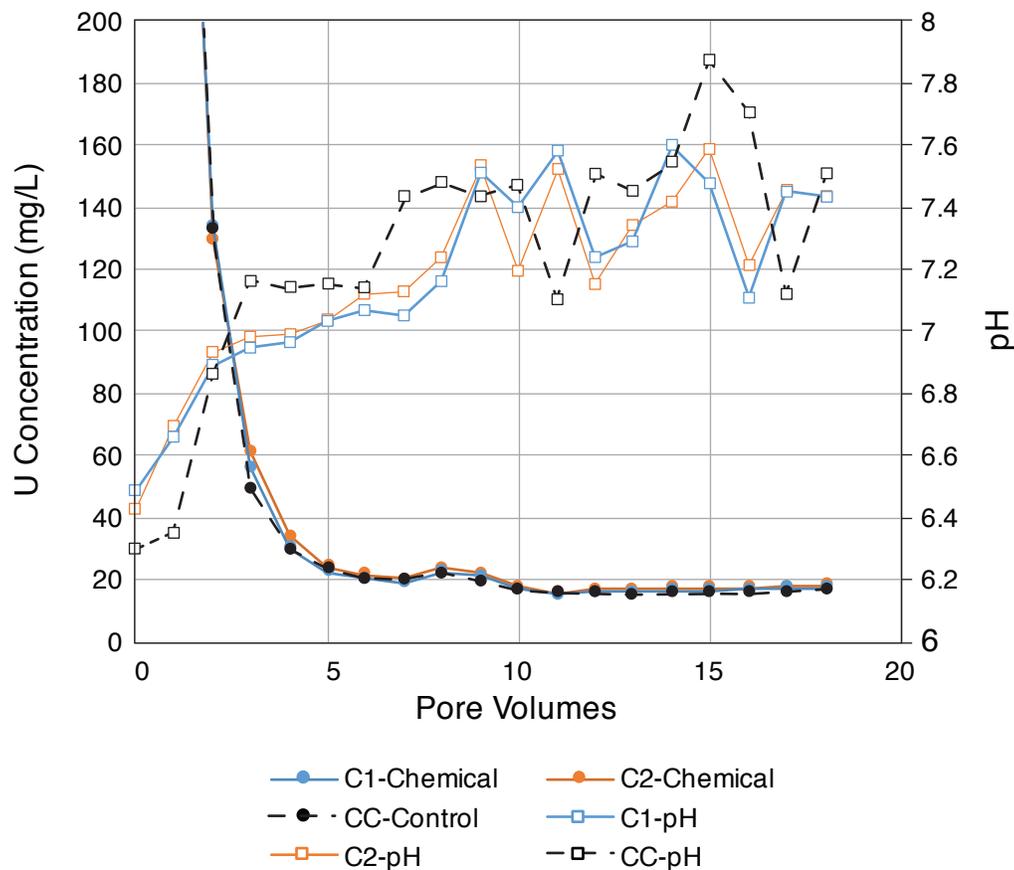


Figure 5. Uranium concentration (represented by solid dots) and pH (represented by open squares) plotted against pore volumes of feed solution for columns fed a 100 µM PO<sub>4</sub> solution. Columns C1 and C2 are replicates and column CC is a control column.

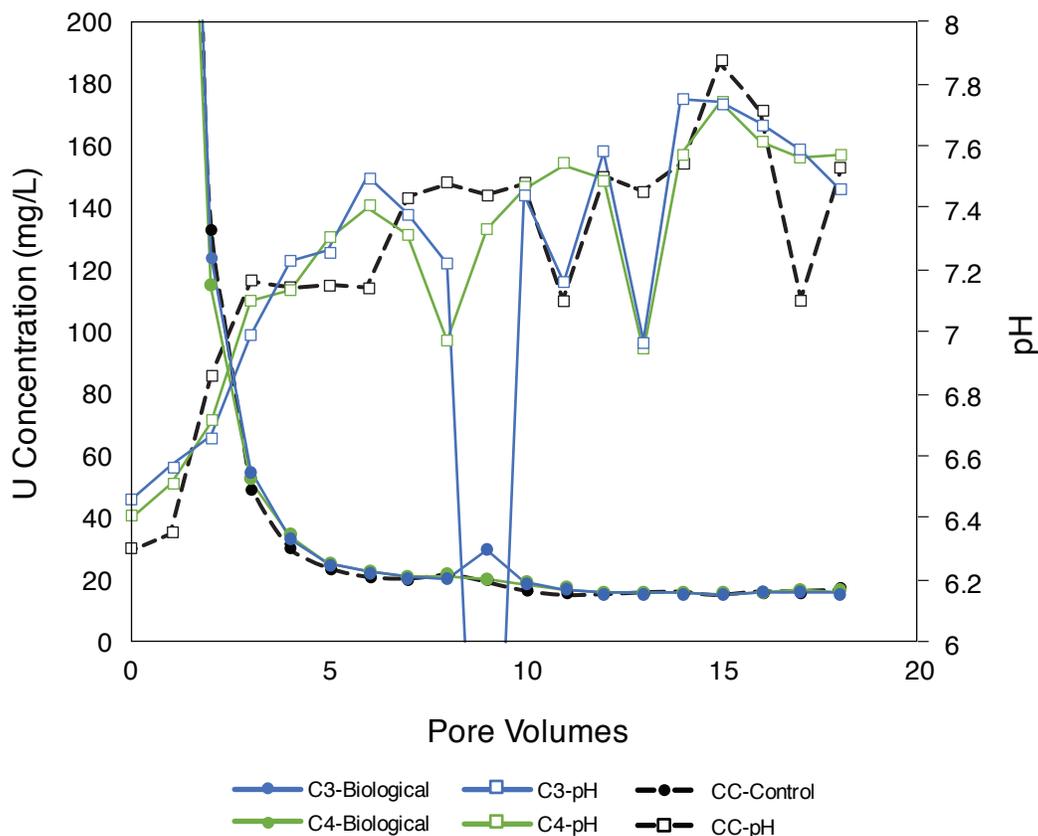


Figure 6. Uranium concentration (represented by solid dots) and pH (represented by open squares) plotted against pore volumes of feed solution for columns fed a 3 mM lactate solution to stimulate biological growth. Columns C3 and C4 are replicates while column CC is a control column.

that the residual U concentration was the result of continuing dissolution reactions from the ore material. Continued leaching of U from the Mt. Taylor ore samples in the columns may also be associated with slow diffusive transport of soluble U species from the interior of rock materials to the bulk fluid flow.

The most important conclusion drawn from these results is that neither chemical addition nor microbial reduction provided any beneficial effect in terms of decreasing U concentrations in synthetic groundwater passing through ore samples that had leached by high concentration of  $\text{NaHCO}_3$  as used in the ISL uranium mining process. The U elution curves in columns leached by groundwater amended with  $\text{PO}_4$  and lactate were essentially identical to a column leached by groundwater alone. It is not apparent that any chemical or biological reactions took place that would immobilize U in these experiments.

In comparing the results presented here with those reported by others it is important to recognize that this study used 1-dimensional packed columns whereas previous studies were done in batch systems. Chemical stabilization through  $\text{PO}_4$  addition in batch systems was investigated by Arey et al. (1999) and Mehta et al. (2016). In these studies the principal objective was to identify the chemical and geochemical reactions occurring, not simulate groundwater restoration methods. Similarly, microbial reduction studies such as those described in the review paper by Williams et al. (2013) were done in batch systems for the purpose of understanding the microbial and geochemical processes associated with microbial reduction. These studies all found good immobilization of U in contrast to the findings of the present study. In comparing the results reported here

with those reported by others it is important to recognize that this study utilized 1-dimensional packed columns to simulate ISL mining and subsequent aquifer stabilization methods, not batch studies in non-flowing systems.

The lack of U stabilization observed in this study is likely due to the hydrodynamics of the system. Specifically, under steady-state conditions, flow through a porous media occurs with little longitudinal or transverse mixing. To a large extent groundwater flow along a streamline can therefore be described as 1-dimensional plug flow. In the experimental system used in this study there was little interaction between the restoration fluids amended with  $\text{PO}_4$  or lactate and the ISL lixiviant in the column at the start of the test. In short, water amended with soluble reactants experiences little mixing with the contaminated solution. Thus, U and other solutes were swept from the column ahead of the restoration fluids. Calculations based on the Ogata and Banks analytical solution of the transport equation presented by Charbeneau (2000) were performed to show this limited mixing (Ruiz Lopez, 2016). This may in part explain the poor performance of a field test at an ISL mine in Wyoming in which  $\text{H}_2\text{S}$ , a strong precipitating and reducing agent, was added with little beneficial effect on groundwater quality (Borch et al., 2012).

The findings of this experimental study have important implications when considering groundwater restoration options following ISL U mining. While chemical or microbial stabilization may be possible restoration method, it is clear that there are hydrodynamic considerations that must be recognized that are every bit as important as the geochemistry, mineralogy, and microbiology.

## Conclusions

There are two notable conclusions that can be drawn from this study. The first is that bicarbonate leaching of U from an ore sample as used in ISL mining strongly depends on the mineralogy and geochemical environment of the U ore. Between 20% and 50% of the total acid extractable U could be leached by a 500 mM NaHCO<sub>3</sub> solution for most samples in five day leach tests. However, less than 5% of the total acid extractable U was leached from an ore sample containing 21.8% organic material. This result confirms that careful consideration of U ore characteristics and mineralogy are important to the success of an ISL-mining project.

Column studies were performed to simulate ISL mining followed by in situ groundwater restoration methods, also referred to as aquifer stabilization methods. Two approaches were considered: addition of PO<sub>4</sub> to achieve chemical stabilization through precipitation of U-PO<sub>4</sub> and related phases, and addition of lactate to stimulate growth of anaerobic organisms capable of reducing sulfate

and sulfide. Neither remediation method reduced the concentration of U or other constituents any more effectively than groundwater sweep conducted in a control column. This was believed to be due to limited mixing between the contaminated groundwater and the amended restoration fluid. Instead of achieving chemical or biological reactions to immobilize soluble U in the columns, the restoration fluid simply forced the contaminated leachate from the column. This finding emphasizes the importance of groundwater hydrodynamics in addition to geochemistry and mineralogy when developing in situ restoration strategies.

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