The Future is Now: Experience with Remediating and Managing Groundwater Contamination at Uranium Mill Tailing Sites – 14587

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ABSTRACT

A primary mission of the US DOE Office of Legacy Management (LM) is protecting human health and the environment through effective and efficient long-term surveillance and maintenance (LTS&M) at Cold War sites that have been remediated, but where residual contamination remains. LM currently manages 91 sites in the United States, including 26 Uranium Mill Tailings Radiation Control Act (UMTRCA) Title I and Title II sites. At an additional uranium mill site in Monticello, Utah, remediation has taken place under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA). At many of the uranium mill tailing sites, uranium and other metal contaminants are present in groundwater. LM faces many of the challenges of remediating and managing groundwater contaminants at these sites, as identified in the 2012 National Academy of Science report, *Alternatives for Managing the Nation's Complex Contaminated Groundwater Sites*. Restoring groundwater at these sites may take longer than originally estimated because of biogeochemical processes not originally understood when remediation goals were set. In addition, remediation to drinking water standards may not be feasible because many of the same contaminants associated with uranium milling are also present as naturally occurring constituents in the groundwater.

Characterization and groundwater remediation strategies at UMTRCA sites are described in US DOE's 1996 *Final Programmatic Environmental Impact Statement for the Uranium Mill Tailings Remedial Action Ground Water Project* (PEIS). The PEIS identifies three basic compliance strategies for attaining groundwater standards: no remediation, natural flushing, and active remediation. The Nuclear Regulatory Commission concurred with the compliance strategy selection framework, and the groundwater cleanup standards were set by US EPA in Title 40 *US Code of Federal Regulations* Part 192, Subpart B. These regulations allow the use of background levels, numerical maximum concentration limits, alternate concentration limits (ACLs), or supplemental "narrative" standards (including technical impracticability) as groundwater cleanup standards. Additionally, the regulations specify a period of 100 years for achieving compliance with these standards if natural flushing is relied on in full or in part.

Groundwater monitoring data to date indicate natural flushing at some UMTRCA sites is proceeding slower than predicted by models. Two examples are provided at the Riverton, Wyoming, Processing Site and the Old Rifle, Colorado, Processing Site. As reported at the Waste Management 2012 Symposium, a flood at the Riverton UMTRCA Title I site in 2010 produced increases in groundwater concentrations of uranium and other contaminants, resulting in the need for new investigations of rock-water interactions to determine additional contaminant sources that could influence compliance strategy decisions. At the Old Rifle UMTRCA Title I site, ACLs are being proposed to replace the previous natural flushing strategy. Research by the US DOE Office

of Science at this site indicates that even in the shallow alluvial aquifer systems, small-scale variations in redox conditions affect the mobility of metals and indicate that natural flushing is far more complex than the simple advective-dispersion transport of contaminants envisioned when remediation goals were first set.

Beyond the monitoring of groundwater that LM conducts as part of LTS&M of these sites, an important part of ensuring that site conditions remain protective of public health and the environment while remediation is ongoing is the use of institutional controls (ICs). Examples of ICs that are in place at different UMTRCA sites to help prevent exposure to contaminated groundwater include providing potable water supplies, restricting well permits, and controlling site use through various environmental covenants, use restrictions, or zone overlays. Because of the importance of ICs as at least an interim step for managing pathways of exposure to contamination, evaluating their effectiveness and whether they are being properly enforced is becoming an increasingly important and challenging part of LTS&M, particularly since most administrative ICs depend on US DOE working with states, tribal governments, and local governments that usually have the authority to establish and enforce them.

In addition, even in the relatively short time that LM has managed these sites, due to changes in resource use nearby, LM is evaluating potential new exposure pathways. For example, around UMTRCA sites in Wyoming and South Dakota, in situ leaching (ISL) systems for uranium are common. Because ISL on a large scale is a relatively new technology, LM is evaluating whether ISL operations could affect groundwater flow and contaminant levels at the UMTRCA sites. Oil and gas drilling around many of these same sites and others that LM manages in the Rocky Mountain region could change potential human health pathways. At the Rifle UMTRCA site, several gas drilling activities, which LM has no authority to control, have resulted in surface contamination at the LM site. Lastly, US DOE is still learning what realistic cleanup levels are for many of these sites. Many of the same contaminants that were associated with uranium ores and mill tailings (e.g., uranium, vanadium, molybdenum, and arsenic) occur naturally as well, particularly at UMTRCA sites on the Colorado Plateau. LM may be successful in remediating groundwater to background levels within 100 years, but the water may still not be suitable for unrestricted use.

INTRODUCTION

The Uranium Mill Tailings Radiation Control Act (UMTRCA) of 1978 established a program for the cleanup and stabilization of uranium (U) mill tailing sites. Title I of UMTRCA designated 22 inactive uranium-ore-processing sites for remediation. The Title I sites had been privately operated but had shut down by the time the law was passed. Remediation of these sites by US DOE resulted in the creation of 19 disposal cells that contain encapsulated uranium mill tailings and associated contaminated material [1]. When it was established in 2003, the US DOE Office of Legacy Management (LM) became responsible for long-term surveillance and maintenance (LTS&M) of these sites (Fig. 1).

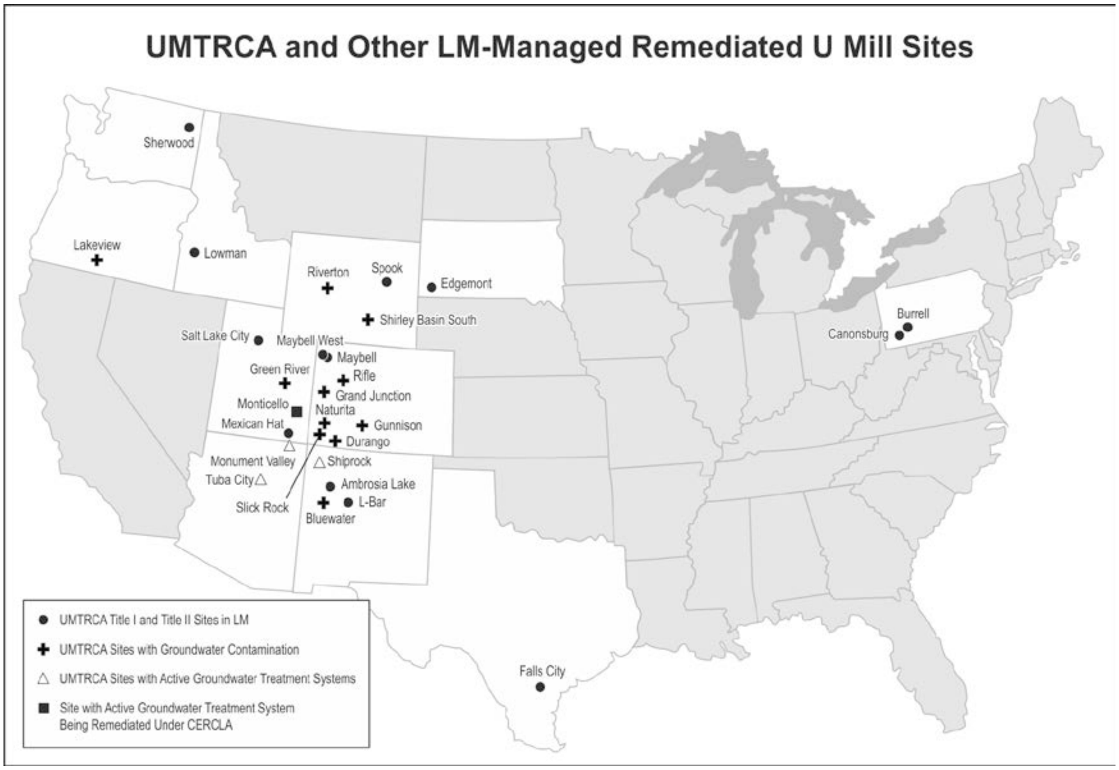

Fig. 1. UMTRCA and other former uranium mill sites in the United States at which LM performs long-term surveillance and maintenance. At 12 of the sites, LM is managing contaminated groundwater.

Title II of UMTRCA addressed uranium mills that were still active in 1978. These sites were commercially owned and regulated under a US Nuclear Regulatory Commission (NRC) license. For license termination, the owner conducts an NRC-approved reclamation of any onsite radioactive waste remaining from uranium-ore-processing operations [1]. When site remediation is complete, US DOE accepts title and is responsible for "long-term custody and care," which it implements through its LTS&M program. LM currently manages 6 UMTRCA Title II sites, although it may eventually manage as many as 27. In addition to the UMTRCA sites, the US Atomic Energy Commission operated a U mill in Monticello, Utah between 1948 and 1960 (Fig.1). Because of federal ownership of the site, it has undergone remediation under CERCLA. At the Monticello, Utah, Disposal and Processing Sites, the last remaining operable unit being addressed is for surface and groundwater contaminants [2, 3].

Because of the large amount of water needed to process U ore in the arid west, many UMTRCA sites are located along major rivers in the region (e.g., the Colorado, San Juan, Gunnison, and Green Rivers). Conventional practice during production, prior to the promulgation of UMTRCA, was to use open, unlined ponds to receive waste chemicals (such as sulfuric or nitric acid) used for U ore processing. These were allowed to infiltrate into the ground, resulting in U as a groundwater contaminant along with a suite of other metals. With the exception of three UMTRCA sites and the Monticello site, US DOE selected natural flushing as the groundwater remediation strategy, which was one of the remedies evaluated in US DOE's 1996 *Final Programmatic Environmental Impact*

Statement for the Uranium Mill Tailings Remedial Action Groundwater Project (PEIS) [4]. The NRC concurred with this strategy, and the groundwater cleanup strategies were codified by US EPA in Title 40 *Code of Federal Regulations* Part 192 Subpart B.

This remedy of natural flushing, largely simulated by advective transport of contaminants, could be selected if groundwater remediation objectives could be shown, through modeling, to be met within 100 years. To date, groundwater monitoring data from many UMTRCA sites suggests that natural flushing is occurring at a slower rate than predicted by these models. However, during this same period, a greater understanding of subsurface processes and properties is developing that is helping LM to understand groundwater remediation processes at these sites. In the Methods section that follows, three sites are reviewed that have provided insights on groundwater remediation at former U mill sites in general. These insights include our understanding of the source term of contaminants, the presence of naturally occurring constituents similar to those that occur as contaminants from U ore processing, and biogeochemical processes that affect the mobility of U and other metals.

LM shares the challenges involved in remediating metals, groundwater, and other recalcitrant contaminants with other agencies. In addition, now and in the future, LM will not rely solely on remediation of groundwater to protect public health and the environment. A variety of administrative institutional controls (ICs) are already in place, and additional ones may be required because of changing land and natural resource uses at these sites.

METHODS

Riverton, Wyoming, Processing Site—Contaminant Source Term Larger Than Previously Known

The Riverton UMTRCA Title I site is an example of a location at which US DOE probably had insufficient knowledge of the U source term to accurately predict when natural flushing will remediate groundwater at the site. A U and vanadium ore processing mill operated from 1958 to 1963 at this site on the Wind River Indian Reservation. Although about 1.4 million cubic meters of mill tailings were removed from the site in 1989, U contamination remained in the shallow groundwater along with manganese, molybdenum, and sulfate (SO_4) [5]. The site is located on an alluvial terrace between the Wind River and the Little Wind River, where groundwater discharges. In the central areas of the groundwater plume, U has been measured at greater than 2 mg/L, which exceeds the U standard for UMTRCA (codified in 40 CFR 192) of 0.044 mg/L (Fig. 2).

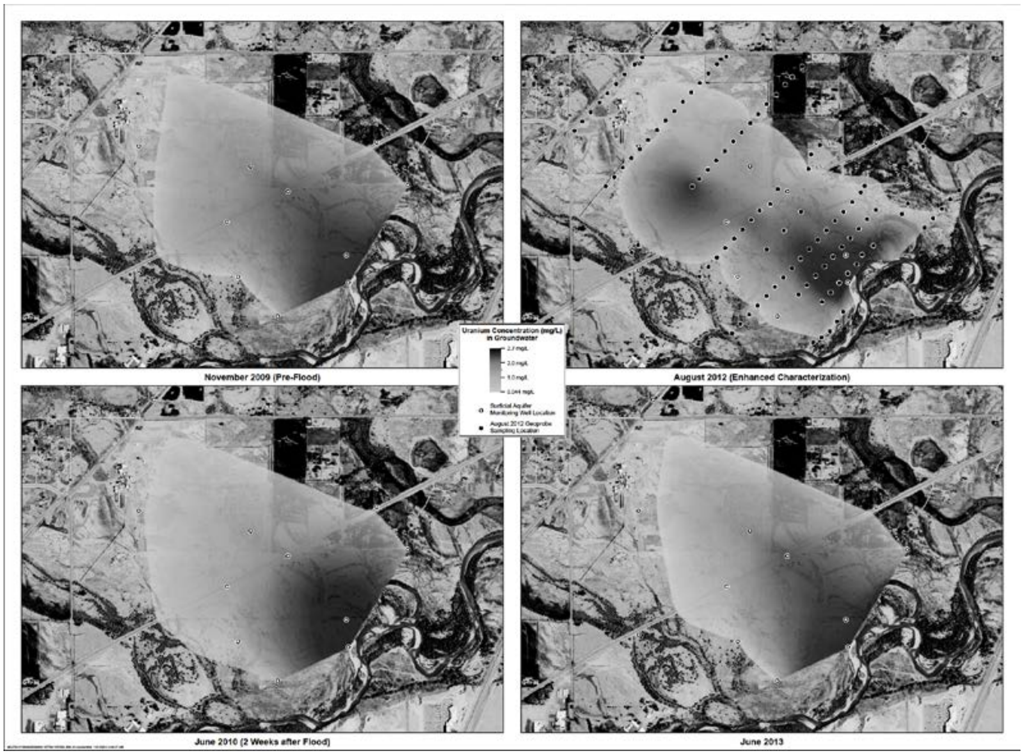

Fig. 2. Changes in the plume dimensions and U concentration in groundwater at the Riverton UMTRCA Title I site before and after the June 2010 flooding of the Little Wind River. Additional characterization conducted in August 2012 showed two areas where U concentrations were as high as 2.7 mg/L as opposed to the understanding of the plume before the flood.

Flooding of the Little Wind River in June 2010 resulted in a threefold increase in U and SO_4 concentrations in groundwater in a well close to the river [6]. While natural flushing certainly occurred during the flood, the level of contaminants during the short-lived event suggested that additional contaminants in what is normally the unsaturated zone at the site were mobilized as well. Model-derived estimates of the time required for contaminants to flush to acceptable levels had not accounted for the mass of U in the unsaturated zone. Additional characterization in June 2012 showed that post-flood contaminant concentrations still remained above the UMTRCA standard for U in groundwater, and the elevated concentrations were spread over a slightly larger area than before the flood [3]. Sampling of monitoring wells in June 2013 indicates that contaminant concentrations in the plume have returned to pre-flood conditions.

Many Devils Wash—Naturally-Occurring Contaminants

LM has also learned that groundwater remediation goals for some UMTRCA sites were unrealistic because of a lack of understanding of naturally occurring constituents. The most dramatic example of this is the result of recent work on naturally occurring contaminants in the Mancos Shale [7]. The Mancos Shale is a widespread, Cretaceous marine deposit of fine-grained sediments in the Colorado Plateau and western slope of the Rocky Mountains [8] that has been used as a natural, low-permeability "liner" below UMTRCA disposal cells at the Shiprock, New Mexico, Disposal Site; Tuba City, Arizona, Disposal Site; Green River, Utah, Disposal Site; and the Grand Junction, Colorado, Site, as well as for the cell under construction for the US DOE Environmental Restoration Project in Moab, Utah [9]. Earlier work at the time of the establishment of the UMTRCA Groundwater Program [10, 11] indicated that constituents such as nitrate, selenium, SO_4, and U could be leached from the Mancos Shale. However, these same constituents are common as contaminants tailings produced from the processing of U ores at UMTRCA mills, including at the Shiprock site [12, 13].

The similarity of constituents occurring in seeps in Many Devils Wash (MDW), an arroyo 0.8 kilometer north of the former Shiprock mill site, to groundwater contaminants from U ore processing led to the assumption that the MDW constituents were from mill operations. However, subsequent investigations have shown that these constituents were leaching from MDW far upstream (hydraulically upgradient) of the mill site, and that similar suites of constituents were being derived from the Mancos Shale in many other areas [12, 7]. Several lines of evidence have further helped to distinguish naturally occurring constituents from contaminants derived from U ore processing at sites such as MDW. One of the most compelling pieces of evidence consists of differences in the ratios of U-234 to U-238. U in groundwater from ore processing tends to have ratios of these two isotopes near 1. However, work by Fleischer [14] has shown that U-234 is more easily leached from bedrock and sediments, leading to U-234: U-238 ratios greater than 2 when the U in groundwater and surface water is naturally occurring, as is the case with MDW [13]. Work by the US Geological Survey in conjunction with the Navajo Nation EPA is confirming LM's conclusions about MDW [15].

Rifle, Colorado, Disposal/Processing Site—The Complexity Of U Mobility And Sequestration

The Rifle UMTRCA Title I site is another site where U concentrations in groundwater have stayed relatively constant since the U mill tailings were removed from the site and regular monitoring of the site began in 1996 [16]. Rifle was the site of two U mills constructed along alluvial floodplains of the Colorado River.

An alternative strategy to relying solely on natural flushing to meet groundwater quality standards for U and other metals is to immobilize them in sediments. Understanding naturally occurring processes for this and how it can be augmented has been a major objective of the US DOE Office of Science Integrated Field Research Challenge (IFRC) at the Rifle UMTRCA Title I site. The Rifle IFRC is a multinational laboratory and university consortium led by the Lawrence Berkeley National Laboratory (http://esd.lbl.gov/research/projects/rifrc).

A key property in the fate and transport of U is its valence state, which is usually either hexavalent [U(VI)] or tetravalent [U(IV)]. In its hexavalent state, U is very soluble and travels with water, but U in the tetravalent state is insoluble and essentially immobile. In addition, in slightly basic to alkaline waters (e.g., pH 6–8), U tends to occur in the more soluble hexavalent state. The majority of UMTRCA sites, as well as the Monticello site, are in arid and semiarid environments where U is more mobile. However, certain microorganisms can serve as electron donors, facilitating its reduction from U(VI) to U(IV) and potentially immobilizing it in the soil [17]. In addition, some microbes have been discovered that can sorb U(VI), the form of the element that is normally soluble (Fig.3).

The biogeochemical reduction or sorption of U is occurring in thin zones of finer-grained sediments of what otherwise ranges from relatively sandy to gravel sediments of the approximately 3.5 m thick alluvial aquifer at the Rifle site. The hydraulic conductivities of the coarser-grained material may be 3 to 4 times those of the finer-grained sediments. The Rifle site may be a situation where contaminant transport occurs relatively rapidly through preferential pathways in the coarser-grained sediments, carrying U and other metals in these zones to the Colorado River but largely bypassing the finer-grained units where biogeochemical sequestering of U is occurring [18]. Natural flushing may be effective with time if the biochemical sequestering of U is permanent or if diffusion of U from the finer-grained sediments to water in the coarser sediments is slow enough to keep U concentrations below regulatory limits. However, these are questions still to be answered about the site, and whether processing is affecting contaminant transport.

DISCUSSION

The Challenges Of Managing Contaminated Groundwater—LM Is Not Alone

LM is not alone in the challenges it faces in remediating and managing groundwater contamination at UMTRCA sites. A 2012 National Academy of Science (NAS) report resonates with many of LM’s issues. In the report, *Alternatives for Managing the Nation’s Complex Contaminated Groundwater Sites*, it was concluded that at about 10 percent of the more than 126,000 sites in the United States with groundwater contamination, restoration was unlikely to be achieved in the next 50 to 100 years because of limitations in technologies [19]. In addition, the committee that prepared the report noted that at 50 percent of the sites deleted from US EPA’s Superfund list, water treatment systems are still being operated or there is a need to manage access to groundwater that does not yet meet standards for unrestricted use. Among the committee’s recommendations was a more formal process for transitioning sites from active remediation to passive remediation or passive long-term management, as well as more research on risk assessment and other management tools to ensure that sites with residual contamination protect human health.

The US EPA Office of Superfund Remediation and Technology Innovation has expressed a willingness to consider recommendations from the NAS [19] report, citing monitored natural attenuation as a passive groundwater treatment strategy that could be implemented when active treatment, such as pump-and-treat, for recalcitrant contaminants (e.g., metals and dense nonaqueous phase liquids) show diminishing returns in terms of contaminant reduction [20].

Monitoring and Institutional Controls

At all UMTRCA Title I and Title II sites managed by LM, groundwater monitoring is an important part of its LTS&M program. Objectives of the monitoring have progressed far beyond simply measuring contaminant concentrations compared to regulatory limits. Other objectives include assessing the rates and levels of success of remediation processes and gaining a better understanding of subsurface properties. This is particularly important given that LM is largely relying on passive remediation to meet groundwater remediation objectives at the majority of the UMTRCA sites.

However, now and in the future, LM does not rely solely on monitoring and remediation to ensure that the public and the environment remain protected. At each of these sites, different forms of administrative ICs are in place to prevent contamination or limit the use of water that may be contaminated, such as by restricting the drilling of new water wells that create new potential pathways for exposure. In addition, at three UMTRCA Title I sites it was determined early in the assessment of groundwater that people lived sufficiently close to the former uranium-ore-processing site that there was potential for exposure, particularly for people using water from private wells. In these situations, alternate water systems were installed. The sites where alternative water supplies are still part of the remedy include the New Rifle site; the Gunnison, Colorado, Processing Site; and the Riverton site.

New Natural Resource Use Around UMTRCA Sites—The Future Is Now

LM has responsibility in perpetuity to ensure that the public and the environment remain protected at the sites it manages. However, while land use and population have changed very little around some of the sites discussed herein, at others LM already needs to address issues associated with groundwater, particularly because of other natural resource industries.

Although the current price of U in large part makes it unprofitable to do conventional mining in the United States, in situ leaching (ISL) of U has become common, especially in the central Rocky Mountain region. For ISL, U ores are leached underground by injecting a solvent solution, or lixiviant, which adds oxygen, carbon dioxide, or sodium bicarbonate. The lixiviant is essentially acting as an oxygen donor, converting U(IV) to U(VI) and making it more soluble (Fig. 3). The U-bearing solution is pumped to the surface where solvent extraction or ion exchange is used to precipitate the U [21]. Around several UMTRCA sites in Wyoming as well as the Edgemont, South Dakota, Disposal Site, ISL fields have been or are being developed. A concern of LM is ensuring that the ISL operations do not inadvertently change groundwater flow directions or the solubility of U in groundwater at the UMTRCA sites and create compliance issues.

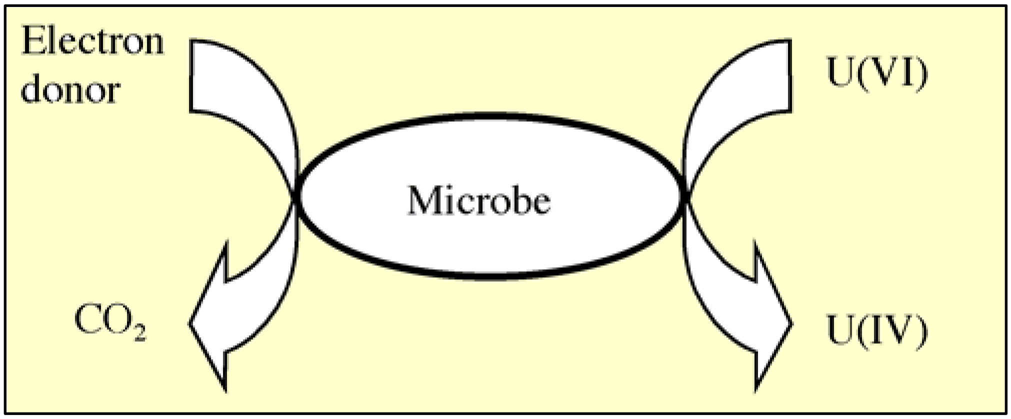

Fig. 3. A schematic of microbial-enhanced U reduction in groundwater (http://www.stanford.edu/group/evpilot/uranium.htm).

Another natural resource extraction use that is occurring in the vicinity of some UMTRCA sites, particularly in Colorado and Wyoming, is oil and gas drilling, including the use of hydraulic fracturing. In both states, subsurface rights for oil and gas are separate from those for water. An example of why this is significant is the Rifle processing site. Although US DOE has established ICs to limit use of groundwater (including drilling new water wells), the Colorado Oil and Gas Conservation Commission has permitted natural gas drilling within the site boundary since the zone of resource extraction is below the alluvial aquifer where the UMTRCA groundwater contaminants occur. In Wyoming, several of the UMTRCA Title II sites for which LM will eventually have responsibility occur at least in part on land managed by the US Bureau of Land Management. For these sites, LM will need to do land withdrawals when it takes responsibility for them, but the program is also evaluating the need to secure subsurface rights for oil and natural gas too. Clearly, future use and potential receptor scenarios for these sites are not abstract concepts that may need to be addressed in the future. It is an issue that LM is addressing today.

CONCLUSIONS

Similar to other agencies and programs dealing with remediated sites, LM's management of groundwater at its UMTRCA and other sites is one of the most significant challenges for LM. It is too soon to know if natural flushing will meet compliance objectives in a 100-year time frame at the UMTRCA sites, but it is now understood that natural flushing is much more complex than a process of advective-dispersion. However, as illustrated above, work by LM, the US DOE Office of Science, and others is providing a better understanding of the processes that affect the mobility of contaminants at these sites, as well as illustrating what is realistic in terms of remediation goals given the incidence of naturally occurring constituents similar to groundwater contaminants that result from U ore processing. One fact that is clearly emerging with improved understanding of naturally occurring constituents is that LM may be successful in returning groundwater to background levels for contaminants of concern, but the water may still not be suitable for unrestricted use.

While issues associated with UMTRCA groundwater contamination are important to resolve, equally important is the fact that at no LM sites, UMTRCA or otherwise, do groundwater contaminants pose a risk to people or the environment. However, because most of the UMTRCA sites are in the western United States, where water resources are coveted, and natural resource industries are creating potential new pathways for contaminant mobility and even receptor scenarios, management of groundwater at these sites will continue to remain challenging and requirements will continue to evolve.

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