

**Monitored Natural Attenuation of Groundwater:
Is Past Performance an Indication of Future Results? – 11222**

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ABSTRACT

Monitored natural attenuation, or MNA, is a groundwater remediation strategy at 14 sites managed by the U.S. Department of Energy Office of Legacy Management (LM). Monitoring data for the MNA remedies are being interpreted on contaminant-specific and site-specific bases to assess the progress of groundwater cleanup. Results from the monitoring conducted thus far are mixed; some sites show notable decreases in contaminant concentration and others indicate little to no decreases. Temporal plots of concentration at wells designed for evaluating MNA progress occasionally match trends predicted by previously developed mathematical models, but water quality trends observed at most sites managed by LM tend to diverge from model projections. Departures between predicted and observed contaminant concentrations may pose concerns about the long-term effectiveness of MNA but do not raise immediate issues regarding human health impacts or environmental risk. Ongoing evaluations of the natural attenuation remedies, in which particular focus is given to identifying the key physical and chemical phenomena that influence contaminant levels on a sitewide scale, will largely determine whether modified groundwater remediation approaches are needed.

INTRODUCTION

Interest in monitored natural attenuation, or MNA, as a form of groundwater remediation heightened about 10 to 15 years ago based on Superfund remedy selection data [1]. Much of this interest was generated by the realization that many engineered remediation systems were showing limited effectiveness and that natural processes occurring in some groundwater media could mitigate the impacts of subsurface contamination without the expense associated with aggressive water treatment. As a consequence, numerous papers and books were published [2,3] that described the relative benefits of natural attenuation and the conditions under which its various forms might be applicable to contaminated sites. About the same time, regulatory agencies such as the U.S. Environmental Protection Agency (EPA) were developing directives and protocols [4] designed to provide guidance on the acceptable use of natural subsurface processes as groundwater remedies. The guidance documents universally emphasized the need for thorough characterization and regular monitoring of the impacts of natural processes on groundwater chemistry. Accordingly, MNA was adopted as a viable means of cleaning up contaminated subsurface media.

The U.S. Department of Energy (DOE) Office of Legacy Management (LM) currently manages 14 sites at which MNA has been adopted as a primary remedy for reducing the effects of groundwater contamination left as a legacy of historical site activities. Although organic chemicals such as chlorinated solvents or nitroaromatics are found in groundwater at some of the LM facilities, inorganic constituents in the form of dissolved anions, metals, metalloids, and

radionuclides comprise the vast majority of the contaminants. Using flow and transport modeling as a guide, DOE chose MNA as the groundwater remedy at these sites in the late 1990s or early 2000s. Thus, in some cases as many as 10 years of concentration data have been collected since implementation of the remedy, making it possible to preliminarily assess the effectiveness of MNA.

Many of the LM locations at which MNA is an operative remedy are uranium-ore processing sites that are regulated under Title I of the 1978 Uranium Mill Tailings Radiation Control Act (UMTRCA). Title I of UMTRCA required DOE to complete remediation of more than 20 former mill facilities that were inactive as of the late 1970s (known as “Title I sites”). DOE established the Uranium Mill Tailings Remedial Action (UMTRA) Project and began surface cleanup in 1983. EPA established criteria, and surface cleanup work progressed into the early 1990s, but EPA’s standards for cleanup of groundwater at the Title I sites were not finalized until January 1995. The groundwater cleanup regulations explicitly provide for the use of MNA (referred to in the regulation as “natural flushing”) for situations in which passive remediation can attain appropriate groundwater standards within an extended 100-year remediation period. It was assumed that natural flushing could achieve remedial objectives using a combination of natural groundwater discharge to surface waterways and ambient hydrologic and geochemical processes. When the MNA remedy is selected, institutional controls, sometimes involving deed restrictions, are required to help ensure the protection of human health and the environment.

For the Title I processing sites, the natural flushing compliance strategy has been DOE’s default remediation option for cases in which contaminant fate and transport modeling indicates compliance can be achieved within the allotted 100-year time frame. Modeling was completed for the majority of the sites by the early 2000s. DOE has selected remedies for all of the Title I sites over the last decade or so, though some are still pending approval by the U.S. Nuclear Regulatory Commission, DOE’s regulatory agency. DOE has been acquiring monitoring data to assess the performance of MNA over this period. LM assumed management responsibility for the Title I sites in 2003.

The Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) is the regulatory driver for additional LM sites at which MNA is either a primary groundwater remedy or a polishing mechanism. Under CERCLA, no specific remedial time frames are prescribed for MNA; however, EPA guidance indicates that cleanup should be accomplished within a time frame that is “reasonable,” given site-specific circumstances.

Now that MNA is underway, performance monitoring at the LM sites is geared to understanding why contaminant concentrations are increasing, decreasing, or remaining relatively constant. Each new monitoring event is viewed as an opportunity to assess whether environmental conditions are changing and whether observed data support the modeling predictions. In some cases, the short-term (<10 years) trends suggest MNA is proceeding rapidly, while in other cases it appears to be having no effect whatsoever. Because of the long time frame allotted for the Title I sites and the fact that many do not yet have a formally approved remedy (i.e., the natural flushing “clock” has not yet started), most of these sites are in the early stages of cleanup. It may be premature to judge if the initial contaminant trends are either short-term aberrations or indications that contaminant concentrations will (or will not) attenuate naturally within a

reasonable time. However, certain patterns are emerging that bring into question the effectiveness of MNA, at least for certain portions of these sites or for certain contaminants. At all sites, the MNA strategy is administered in a manner that ensures protection of human health and the environment.

Monitoring of the MNA remedies has revealed much new information about the flow and fate and transport processes affecting contaminant concentrations in groundwater. As discussed later in this paper, these processes reflect subsurface system complexities, the effects of which could not be fully taken into account when modeling of natural attenuation was initially used to select a groundwater remedy. Subsequent sections describe observations made regarding the progress of MNA at four LM sites. Groundwater contamination at all of the facilities is the result of uranium milling activities; three of the locations are former UMTRA sites and one is a CERCLA site.

SUBSURFACE SYSTEM COMPLEXITY

Previous Groundwater Modeling

Groundwater modeling was used to simulate MNA processes at the LM sites where cleanup standards were deemed achievable within a reasonable time frame (e.g., 100 years). Given the general practice of subsurface modeling during the late 1990s, numerical models incorporating simplified flow and transport conditions were employed. Frequently, spatially uniform aquifer properties, such as hydraulic conductivity, were applied to single hydrogeologic units within a model even if evidence of heterogeneity in aquifer materials was manifest. The logic at the time was that the use of a single, average value for an aquifer property would generally capture the most important behavior of an aquifer under natural ambient conditions. Typically, this approach resulted in steady-state groundwater flow models that performed well in matching the average value of periodically measured water levels at monitoring wells. Yet attempts to develop transient versions of the flow models as calibrated to hydrographs at individual wells were often thwarted by a lack of continuous hydraulic head information over periods of multiple years.

At some UMTRA sites with a natural flushing remedy, the flow modeling was conducted using probabilistic methods based on a Monte Carlo simulation approach. That is, several hundred simulations, each based on a randomly generated hydraulic conductivity field, were conducted with the intent of developing a suite of flushing projections. To some extent, this allowed the observed heterogeneity of an aquifer to be taken into account. Although this approach typically produced steady-state flow fields that generally comported with an “average” representation of an aquifer’s potentiometric surface, it could not fully account for actual heterogeneity, in which distinct, low-permeability zones significantly reduced the rate at which water advected into more permeable, preferential flow zones. Such model limitations had the potential to produce overly optimistic projections of aquifer cleanup.

As with flow simulation at MNA sites, the transport portions of the models used to project the effectiveness of natural flushing also contained numerous simplifications. Modelers were generally aware that some contaminants were potentially affected by chemical reactions in the subsurface, but the nature of those reactions was typically not identified, let alone quantified, during earlier site characterization. Consequently, contaminant sorption onto the solid media of an aquifer was usually the only reaction that was simulated in a transport model. Moreover,

contaminant sorption was modeled simply as a linear, equilibrium partitioning process, and a single soil-aquifer distribution coefficient (K_d) was uniformly applied to the entire extent of a given hydrogeologic unit. This approach simulated the retarded transport of a sorbing contaminant in a general manner but did not account for more complex sorption (and desorption) phenomena as affected by spatially and temporally variable geochemical conditions, such as surface area limitations on sorption sites or the presence of additional constituents that can either enhance or inhibit sorptive processes. Calibration of transport models was limited by the fact that the concentration data collected during site characterization were limited to a relatively short time span that occurred many years after contaminant plumes first started developing due to site activities.

Although site characterization of geologic media and constituent concentrations at the MNA sites was extensive, challenges frequently arose when attempting to identify the nature and extent of contaminant sources within individual hydrogeologic units. As a result, the quantitative representations of those sources in site models were often simple in concept and highly uncertain. The possibility that contaminants residing in vadose zone materials might be periodically added to the underlying saturated zone was generally not considered. Similarly, continuing saturated zone sources in the form of immobile contamination residues (e.g., chemical precipitates, contaminant sequestration in low-permeability material) were ignored if there were no site characterization data to indicate their presence. Such modeling simplifications add to the possibility that predictions of natural flushing are overly optimistic.

Over the past 10 years, considerable progress has been made with respect to understanding the transport processes occurring in contaminated subsurface media. On the whole, the advances made illustrate that groundwater systems are affected by numerous complexities that can have profound effects on the effectiveness of existing groundwater remedies. Because the simplified models used to select MNA as a remedy were unable to account for such complexities, it stands to reason that divergences between model projections and measured concentrations will be observed. The following sections summarize findings made during the last decade regarding subsurface flow and transport and how these might influence MNA at LM sites.

Subsurface System Heterogeneity

It has long been recognized that aquifers are naturally heterogeneous and that spatial variability of properties affecting groundwater flow can also significantly influence advective-dispersive transport in the subsurface. Relatively recent studies have demonstrated more clearly how heterogeneity can slow the removal of contaminants from an aquifer. For example, tracer studies at heavily monitored field sites have been able to further illustrate how rate-limited mass transfer of dissolved constituents from decimeter-scale zones of low hydraulic conductivity materials (immobile domain) into high-permeability preferential flow channels (mobile domain) causes long-lasting persistence, or tailing, of constituent concentrations on a sitewide scale [5,6]. In many cases, researchers have found that the tailing effects are more adequately simulated using models based on dual-domain formulations [7,8], as opposed to conventional models based on the classical advection-dispersion equation [9].

To a large extent, the rate-limited mass transfer in highly heterogeneous units from low-permeability zones to more permeable preferential flow paths is governed by the transport

processes of advection, mechanical dispersion, and molecular diffusion. However, logic suggests that the slow release of contaminants from low-permeability parts of a porous medium to high-permeability parts is also strongly influenced by geochemical processes that are different in fine-grained sediments than in coarse-grained materials. Such logic is supported by numerous investigations [10] that have documented how geochemical (or mineralogical) heterogeneity can lead to chemical reactions within subsurface microzones that vary greatly from those in bulk aquifer sediments. Correspondingly, because both the chemical makeup of pore waters and sediment grain size and lithology affect the capacity for dissolved constituents to sorb onto sediments, orders-of-magnitude differences in K_d values can be expected between fine- and coarse-grained aquifer materials.

Research during the past 15 years has shown that chemical processes occurring in groundwater domains are often biotic in nature, and microbial respiration is the main driver of significant reactions in the subsurface. Often, these enzymatically mediated processes involve oxidation-reduction (redox) reactions that can result in the immobilization of metals, metalloids, or additional inorganic species. As with abiotic subsurface chemistry, biologically driven reactions have a tendency to differ significantly between sediment types.

The effects of sediment, geochemical, and microbial heterogeneities on ambient processes in a groundwater system are complex, which in turn signifies that the long-term success of an MNA remedy is inherently uncertain. Continued monitoring at an MNA site provides a mechanism for identifying if subsurface system heterogeneities are potentially impacting the natural rate of contaminant attenuation.

System Transients

Temporal variability of subsurface flow and transport processes adds to the complexity of groundwater systems. Despite observations that transient hydrologic stresses can strongly impact subsurface flow and chemistry, it is common to design groundwater remedies under the assumption that observed flow patterns and contaminant plume concentrations do not change rapidly. As a consequence, evaluation of an MNA remedy on the basis of “average” groundwater conditions may fail to capture significant changes within a contaminant plume in response to extreme hydrologic events. For example, anomalously high groundwater levels induced by flooding of a nearby river connected to an aquifer could enhance leaching of vadose zone contaminants that normally have minimal impact on the saturated zone. Similarly, vadose zone contaminants entrained by downward-seeping moisture in response to infiltrating rainfall could cause local contaminant levels in the plume to periodically spike above typically observed concentrations. Because MNA can require long-term monitoring over periods of decades, transient effects on plume concentrations due to occasional hydrologic stresses are expected.

RIVERTON SITE

The Riverton, Wyoming, Processing Site is an UMTRCA site in Fremont County, Wyoming, about 3 km southwest of the town of Riverton and within the boundaries of the Wind River Indian Reservation. A mill processed uranium-vanadium ores at the location from 1958 to 1963, generating contaminated tailings containing both radioactive (radium, thorium, and uranium) and nonradioactive (manganese, molybdenum, sulfate) constituents. At one time, a 1.2-m-deep

tailings pile covered about 29 ha of the 57-ha site. About 51,000 m³ of contaminated tailings and other materials were removed from the site in the late 1980s and relocated to a disposal facility about 72 km away.

In addition to producing surface contamination, mill operations and leachates from the tailings resulted in contamination of nearby surface water and groundwater in a shallow, 4.5- to 6-m-thick alluvial aquifer underlying the site. Local groundwater in the alluvium flows to the southeast and discharges to the Little Wind River about 0.8 km downgradient of the former mill and tailings areas. Using the results of probabilistic modeling of subsurface processes that accounted for advection, dispersion, equilibrium sorption of dissolved constituents on alluvial sediments, and groundwater seepage to the river, DOE selected MNA in the form of natural flushing as the compliance strategy for the alluvial aquifer. Groundwater monitoring at several wells completed in the alluvium (Fig. 1) has been used to track progress of this strategy.

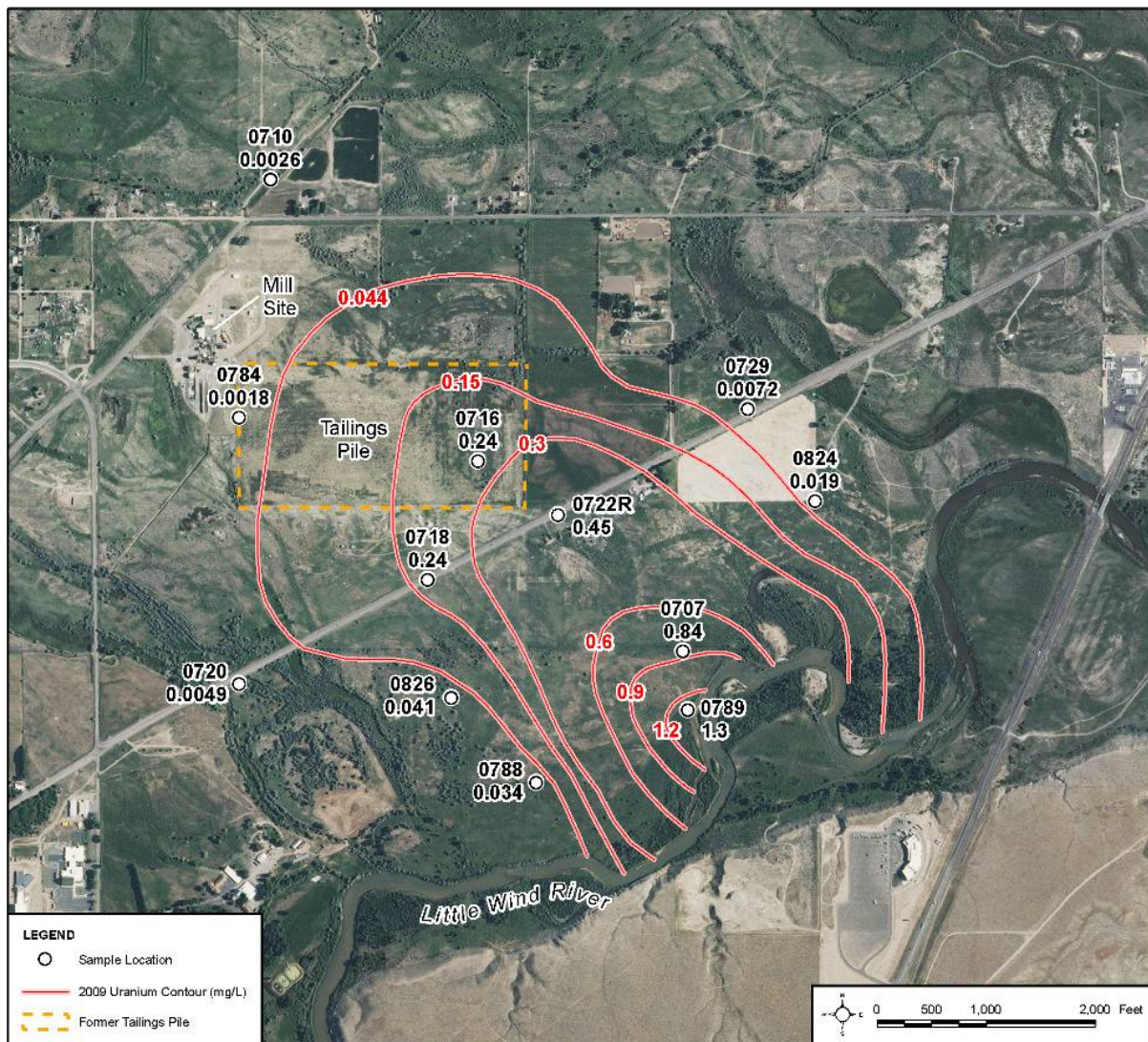


Fig. 1. Monitoring wells at the Riverton site and uranium plume concentrations in 2009.

Monitoring conducted through 2009 has generally indicated that the alluvial aquifer is flushing, as demonstrated by declining concentrations measured at several wells located within the uranium plume at the site. A temporal plot of uranium concentrations (Fig. 2) at a well located about midway between the former mill area and the river (well 0722/0722R) provides an example of the general decline in contaminant concentrations. The graph also suggests that the trend in uranium concentrations at this location follows an exponential decline.

Observed decreases in uranium concentration for multiple alluvial wells at the Riverton site has led to a recent uranium plume (Fig. 1) that looks considerably different from the plume that existed in the late 1990s. In effect, the concentration decreases suggest that much of the contaminant mass that previously existed within the plume has discharged to the Little Wind River, which is encouraging with respect to the progress of natural flushing. This finding is somewhat tempered by the observation that uranium concentrations in alluvium directly underlying the former tailings pile tend to remain above the regulatory standard (0.044 mg/L) for this constituent.

The decreasing concentration trends at Riverton wells were recently interrupted during a flood of the Little Wind River in June 2010. The hydraulic effects of the flood were manifested as short-term groundwater level increases of 1 m or more at several of the wells located closest to the river, whereas constituent concentrations typically increased by factors of two or more. During the several months since flooding, the concentrations of key contaminants, including uranium, have shown signs of decreasing to values that are much closer to those that were observed in mid- to late 2009. These observations suggest that sorbed or precipitated contaminant phases in materials not normally influenced by average groundwater flow conditions, possibly vadose zone sediments, were temporarily mobilized by the water level increases associated with the flood.

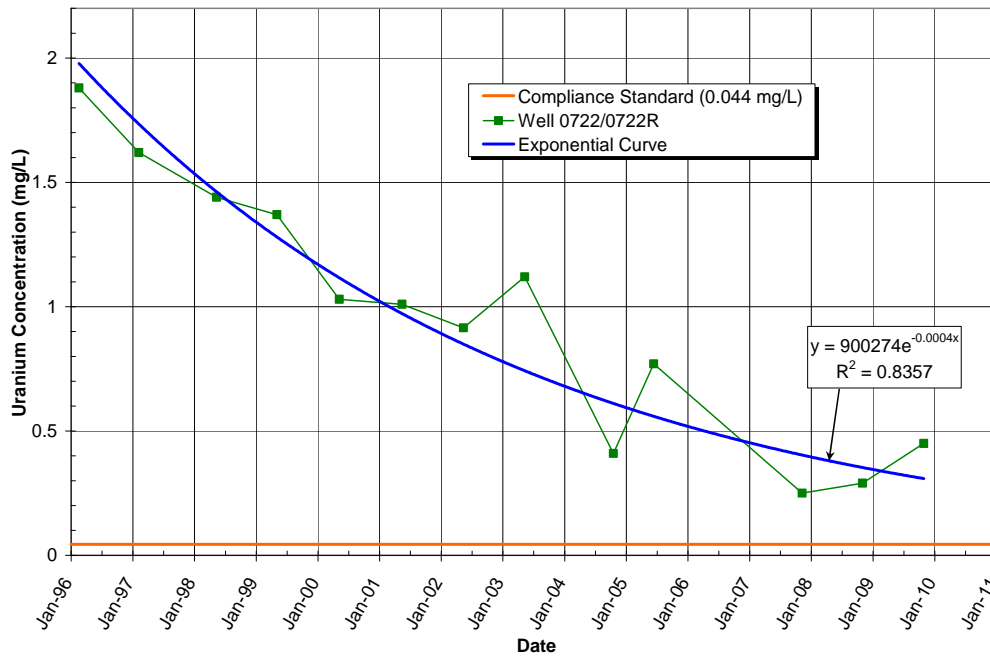


Fig. 2. Uranium concentrations at a Riverton site well located midway between the former mill site and the Little Wind River.

NEW RIFLE PROCESSING SITE

The New Rifle, Colorado, Processing Site, is an UMTRCA site in west-central Colorado near the town of Rifle. The uppermost aquifer at the site consists of shallow alluvium that is bounded on one side by the Colorado River (Fig. 3). Sediments range from clay- to cobble-sized; the aquifer is 6 to 9 m thick over most of the site, and depth to groundwater typically ranges from 1.5 to 3 m. Hydraulic conductivity averages about 35 m/day, and groundwater flow is predominantly to the west. The alluvial aquifer was used as a source of domestic water in the past, but currently institutional controls require connection to the city water supply system. Surface cleanup involved excavation of tailings and contaminated soils to the water table and replacement with clean fill. The primary sources of groundwater contamination were several evaporation or holding ponds and tailings piles.

A number of different contaminants of concern were identified at the New Rifle site, including arsenic, selenium, ammonia, uranium, molybdenum, and vanadium. The different behaviors of these constituents offer insight into some of the attenuation processes at several LM facilities. Arsenic and selenium levels were elevated at the New Rifle site in the past but were not detected above background levels in wells downgradient of the site. In the more than 10 years since surface remediation, concentrations of these constituents have declined to levels below standards. This decline is believed to be largely due to effective source removal and adsorption of these constituents to fine-grained particles in the subsurface. Because these constituents were not present in extremely high concentrations, the subsurface materials were an effective attenuation medium. Ammonia concentrations, which were once fairly high in site groundwater, have also declined below levels of concern in the aquifer. Increases in nitrate accompanying decreases in ammonia indicate that ammonia attenuation is accomplished largely through the biologically driven process of nitrification.

Uranium and molybdenum were thought to be two of the most mobile constituents at the site based on studies to estimate soil-water distribution coefficients. Concentrations in on-site wells appear to have leveled off at concentrations above their respective standards, although they do show significant fluctuation. Fig. 3 presents a map view of recent monitoring results for molybdenum; Fig. 4 shows molybdenum concentration trends over time for selected wells. Concentrations in at least one downgradient off-site well (well 0195) show a classic increasing-then-decreasing trend that could be interpreted as the passage of the peak concentration portion of an off-site plume. While this is an indication that MNA can be effective, it is not clear if this represents a stable condition. Two ponds—an upgradient extraction pond and a downgradient holding pond—were once in operation between wells 0195 and 0201 (Fig. 3). The operation of these ponds during gravel mining caused a temporary groundwater divide in this area and cut off the downgradient portion of the plume from the original source of contamination. Now that pond operations have ceased, it is unclear if additional contamination from the site will recontaminate downgradient areas. As can be seen in Fig. 4, high concentrations of molybdenum remain in on-site wells. This also holds true for uranium. Generally, uranium trends at monitoring wells and spatial distributions of uranium are similar to those for molybdenum; however, interpretation of uranium distribution is further complicated by the likelihood that the area downgradient of the site is affected by naturally elevated concentrations derived from areas serving as a source of recharge.

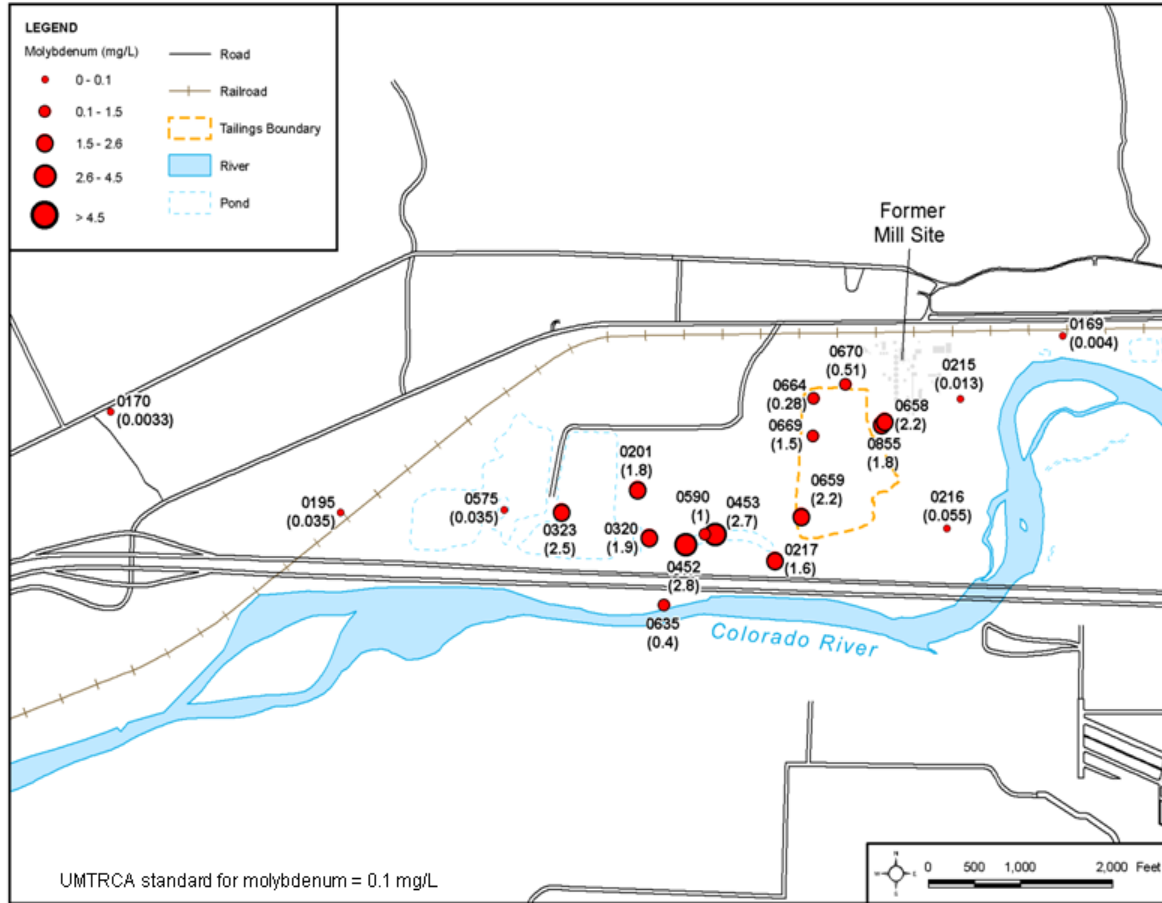


Fig. 3. Molybdenum concentrations at New Rifle site groundwater wells in June 2010.

Vanadium is not an EPA-regulated contaminant; however, because of high levels of vanadium detected in on-site wells, a pilot study was conducted to investigate the behavior of this constituent. Characterization of on-site soils indicated that high concentrations of vanadium (and likely other sorbed constituents) remain in site soils down to bedrock and may serve as a continuing source of groundwater contamination. Site sediments were only cleaned up to meet the UMTRCA standard for radium-226 in soils; no consideration was given to the effect of residual soils on groundwater during surface cleanup. While high levels of vanadium have been observed in site soils, the relative immobility of this constituent has severely limited the extent of its plume in groundwater. Elevated levels are confined to areas on and immediately downgradient of the mill site.

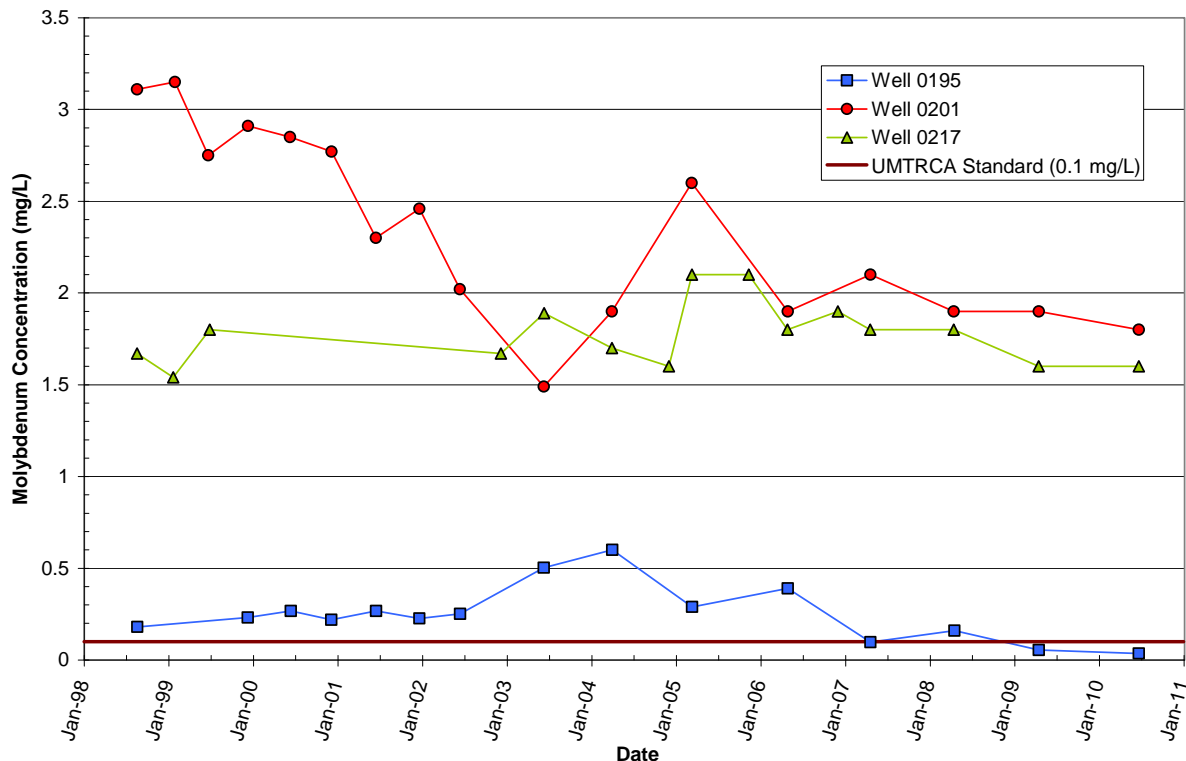


Fig. 4. Temporal plots of molybdenum concentration at selected New Rifle wells.

Ownership of the mill site has been transferred to the City of Rifle, which is using part of the area to expand its municipal sewage treatment plant. During construction activities, the City excavated and dewatered portions of the site. Monitoring data collected during these activities showed significant fluctuations in on-site groundwater concentrations, with higher concentrations appearing to be correlated with decreased saturated thickness of the aquifer. It is not clear if these activities will have any long-term effects on downgradient portions of the aquifer.

It does not appear that natural flushing will be an effective means of reducing concentrations of all constituents to levels below applicable standards at all monitoring locations at the site. Flushing of off-site areas appears feasible but does not seem to be effective in the source areas. However, less-mobile constituents will likely be confined to a relatively small area—on the mill site itself or locations immediately adjacent to the site. Only the most mobile constituents have migrated any distance from the mill site. It is possible that attenuation mechanisms may reduce these constituents to ambient levels in downgradient portions of the plume.

GUNNISON PROCESSING SITE

The Gunnison, Colorado, Processing Site is an UMTRCA site located near the town of Gunnison, in western Colorado between the Gunnison River and one of its tributaries, Tomichi Creek (Fig. 5). Contamination from the site is present in the alluvial aquifer, which consists mostly of sand and gravel with lesser amounts of cobbles, boulders, and clay. The thickness of the aquifer

ranges from 21 to 40 m, and the average depth to the water table is 1.5 m. Average linear velocity of the aquifer ranges from 0.6 to 1 m/day; groundwater flow is to the southwest. Domestic wells are in shallow portions of the aquifer.

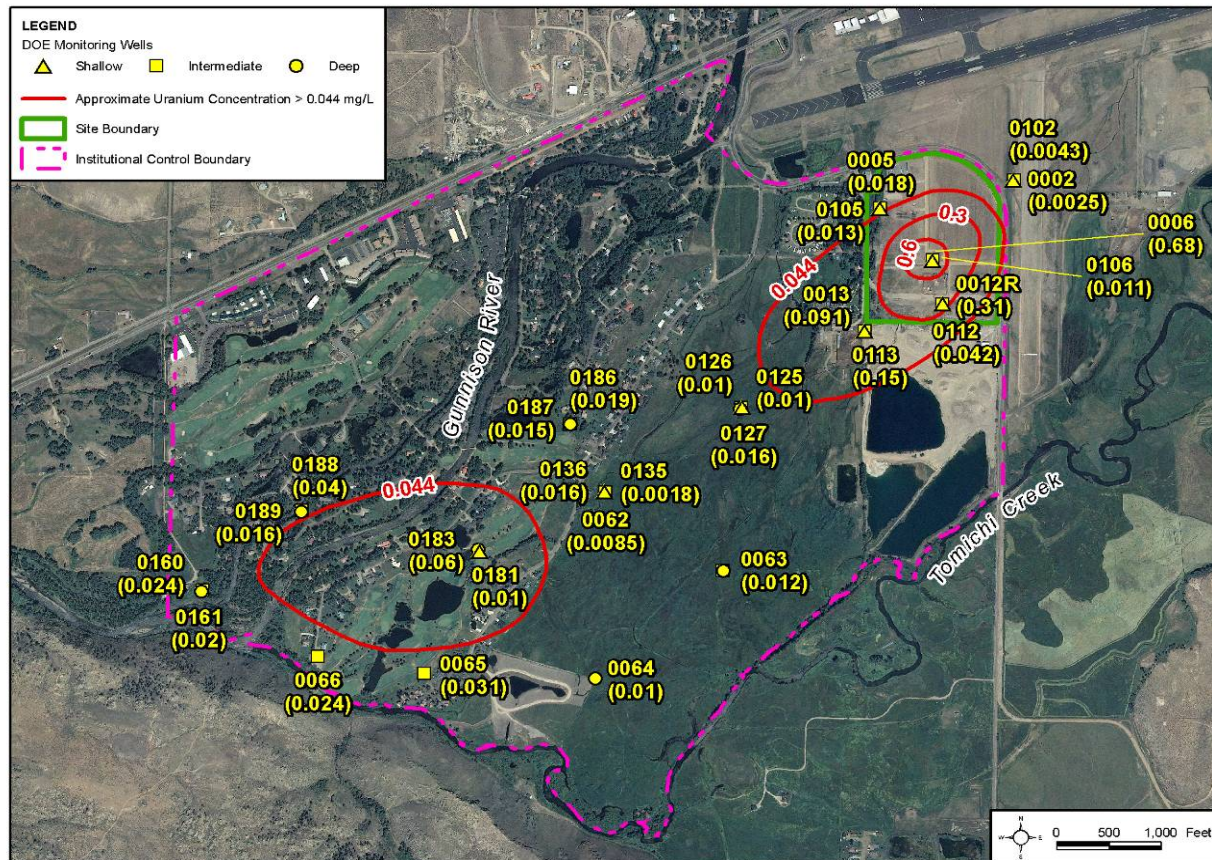


Fig. 5. Uranium concentrations at Gunnison site groundwater wells in April 2010.

As at the New Rifle site, contaminated soils were left in place on the Gunnison mill site because of difficulties in excavating below the water table. Transport modeling for uranium, the main contaminant of concern, took into account this residual source term. Monitoring wells for the site are screened at shallow (1.8–7 m), intermediate (11–18 m), and deep (26–30 m) intervals. Today, most uranium contamination beneath the mill portion of the site is observed in shallow wells. The downgradient contaminant plume appears to migrate to increasingly greater depths with distance from the mill location, which may reflect the effects of recharge from flood irrigation. As a consequence, shallow downgradient wells are largely uncontaminated. In 2010, the only downgradient well with uranium levels that have exceeded the uranium standard is a deep well (well 0183 on Fig. 5), and the uranium concentration in an intermediate-depth well located even farther downgradient is slightly below the standard (well 0188).

During several years following surface remediation, uranium levels in shallow wells at the former mill area appeared to be trending downward from concentrations that were once as high as 1.2 mg/L. More recently, however, mill-area uranium levels appear to have leveled off somewhat, but fluctuations in concentration between 0.2 and 0.4 mg/L are common. Deeper

wells at and adjacent to the mill area appear to show slightly increasing trends in uranium concentration (wells 0112 and 0113 in Fig. 5). The two downgradient wells with the highest uranium concentrations (0183 and 0188) do not show any well-defined trends. Thus, it is unclear if they represent the migration of an attenuating plume. In contrast, intermediate-depth well 0186, a short distance upgradient from these locations, shows slightly declining uranium concentrations, as does shallow well 0181. Wells on the downgradient end of the monitoring network (0160 and 0161) show slightly increasing uranium concentrations, but observed concentrations here are well below the applicable uranium standard (0.044 mg/L).

At this time, it does not appear likely that uranium levels in groundwater wells beneath the mill area will meet the uranium compliance standard within a reasonable time period (e.g., 100 years). It is unclear what effects residual contamination in this area may have on the downgradient portion of the plume. The very limited extent of elevated aqueous-phase uranium levels observed to date suggests that the bulk of the downgradient aquifer may achieve standards. However, the progress of natural attenuation in downgradient portions of the uranium plume is difficult to gauge because of the relatively large thickness of the aquifer and short lengths of monitoring well screens. It appears that the average uranium concentration at most or all wells in the aquifer would meet the standard for this constituent within a reasonable time, but it is uncertain whether an average concentration, in lieu of the concentration at each monitoring location, would be acceptable for demonstrating regulatory compliance.

MONTICELLO MILL SITE

The Monticello mill site is a former uranium mill site located near the town of Monticello (Fig. 6) in southeastern Utah, about 400 km southeast of Salt Lake City. The 32-ha mill property came under the jurisdiction of CERCLA and the regulatory authority of EPA in 1989, and LM assumed management of the site in 2003. Surface cleanup involving the removal of tailings and contaminated soils and sediments on the mill property occurred in 1999 and 2000. Components of Operable Unit I established for the surface remediation included the relocation of contaminated tailings and sediments from the mill area to a disposal cell located about 1.6 km to the south, revegetation after removal of tailings, realignment of a local creek, and reestablishment of wetland areas. Unlike at the UMTRCA Title-I mill sites, soils and sediments associated with the Monticello operation were remediated to meet cleanup levels for constituents in addition to radium-226. In particular, uranium in soils was of concern as a source of groundwater contamination.

A local stream, Montezuma Creek, passes through the former mill area in an east-west direction. A shallow and thin (3–4.5-m-thick) layer of alluvium consisting of fluvial sands, gravels, and some fine-grained sediments fills a 150-m-wide valley that was formed by the creek (Fig. 6). The alluvium is underlain by and bordered on its margins by low-permeability bedrock.

During and subsequent to the Monticello mill's operation between 1942 and 1960, leaching of soils and tailings on and near the mill site resulted in on-site and downgradient contamination of groundwater in the alluvium by several inorganic constituents, including arsenic, molybdenum, nitrate, uranium, and vanadium. Uranium was the most widespread contaminant and represented the greatest potential risk to human health. Montezuma Creek was also affected by the contamination because of groundwater–surface water exchanges.

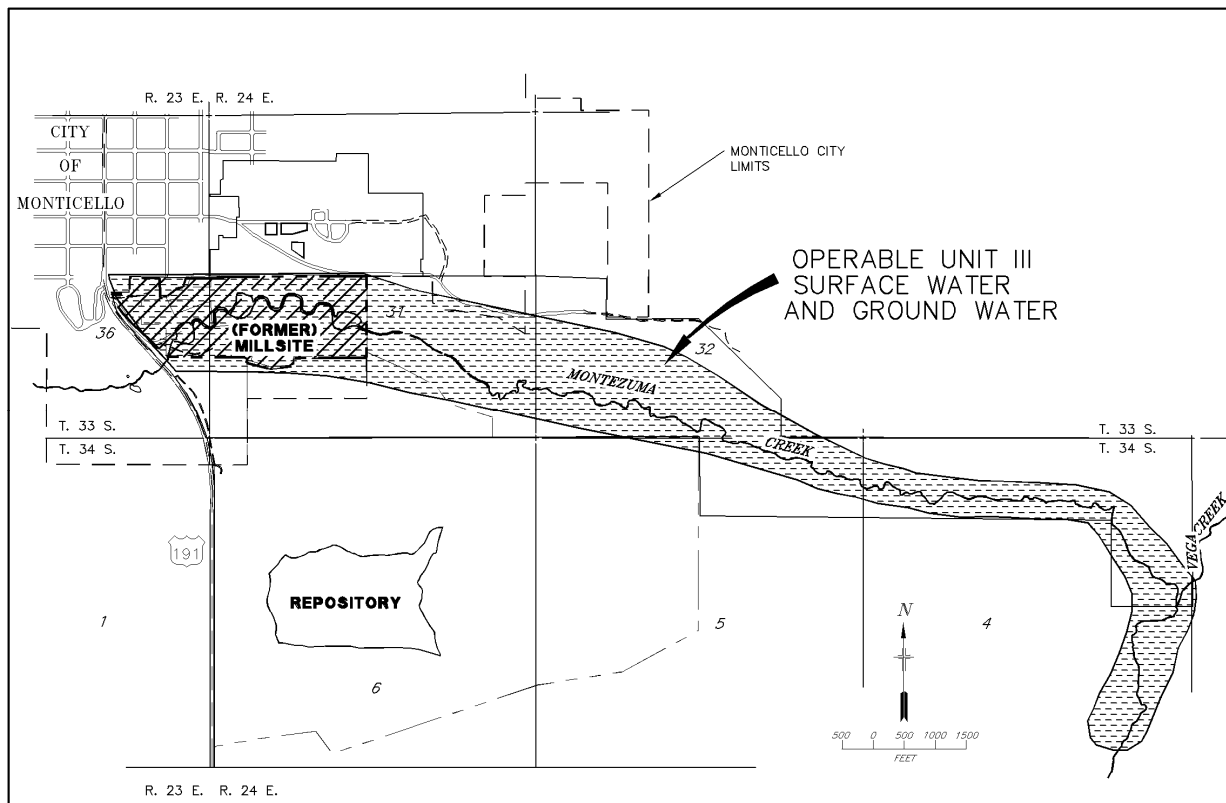


Fig. 6. Monticello mill site and the alluvial valley associated with Montezuma Creek.

Operable Unit III was established in early 2004, 4 years after completion of the surface remediation, for cleanup of surface water and groundwater. The remedy selected was MNA, which was based on a groundwater model developed to simulate flow and transport processes on and east of the mill property. The model suggested that ambient flushing processes would eventually produce uranium concentrations sufficiently low to meet applicable goals designated for both surface water and groundwater. The model predicted that the goals would be reached over a period of about 42 years, beginning in 2002.

The Record of Decision for Operable Unit III contained specific uranium concentration criteria that were expected to be met over a relatively short time frame in the interest of assessing whether MNA was progressing as predicted by the modeling. Groundwater monitoring during the initial 4 years following the modeling indicated uranium concentrations were not following projected declines and were maintaining constant levels that exceeded cleanup goals. In 2007, LM and EPA concurred that the criteria were not being met, which prompted an investigation of potential remedial alternatives other than MNA. A review of relevant data suggested that potential reasons for the divergence between model-predicted and observed trends included simulations based on a constant K_d , which ignores sorption into phases of varying mobility and dual-domain mass transfer.

Certain regions of the aquifer have shown significant restoration progress following contaminant source removal, but concentrations of uranium in these regions have since stabilized between

0.15 and 0.2 mg/L, well above the remediation goal under CERCLA (0.03 mg/L). In another region of the aquifer, restoration progress is marked by significant downward trends in uranium concentration that were once as high as about 0.5 mg/L but have now decreased to about 0.25 mg/L. On the basis of previously described physical and chemical obstacles to flushing and laboratory evidence (column studies) of a contaminant tailing effect in Monticello sediments, it appears that flushing in this latter area (and other locales as well) will eventually lead to uranium concentrations that stabilize between 0.15 and 0.2 mg/L.

LM recently committed to active groundwater remediation in a well-defined region of the alluvial aquifer where uranium concentrations are about 1 mg/L. One objective of this effort is to assess whether more aggressive remediation efforts followed by natural attenuation as a polishing mechanism can lead to concentrations that meet regulatory standards within a reasonable time.

CONCLUSION

Data collected at MNA sites within LM's jurisdiction are being interpreted on contaminant-specific and site-specific bases to assess the progress of groundwater cleanup. The MNA remedy depends on a combination of discharge to nearby waterways and ambient hydrologic and geochemical processes to decrease contaminant levels. Some generalizations about the progress of natural attenuation can be made using the monitoring information collected to date:

- It is unlikely that MNA will achieve unrestricted use standards for groundwater in areas where contaminated soils remain in place (e.g., on former mill sites where only radium-226 soil standards were met).
- Uranium, which is a major groundwater contaminant at many LM sites, was thought to be a good candidate for MNA due to its apparent mobility. However, complexities in the physical and chemical behavior of uranium in the subsurface appear to limit the effectiveness of MNA; cleanup to meet drinking water standards may be impracticable.
- Results of LM efforts identify a need for collecting rigorous data to support selection of MNA as a groundwater remedy. However, the complexity of subsurface systems may render adequate data collection infeasible.
- It is possible that downgradient portions of contaminant plumes may clean up through MNA; however, the degree to which residual contamination associated with source areas will affect plume dispositions in the future is uncertain.
- Although monitoring results at most LM natural attenuation sites do not closely match model-predicted trends, contamination at the sites poses no immediate risk to human health or the environment. Contingency measures may be necessary if conditions change adversely.
- Processes other than discharge of groundwater to surface water are likely responsible for some of the observed contaminant attenuation at LM sites. While these can result in marked reductions of dissolved contaminant mass in groundwater, contaminants might not be fully removed from aquifer media and could be available for remobilization.

ACKNOWLEDGMENT

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