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Evapotranspiration and Geochemical Controls on Groundwater Plumes at Arid Sites: Toward Innovative Alternate End-States for Uranium Processing and Tailings Facilities

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Authors:
Brian B. Looney*, Miles E. Denham*, Carol A. Eddy-Dilek*,
Margaret R. Millings*, Mark Kautsky**

* Savannah River National Laboratory, 773-42A, Aiken SC
** US DOE Office of Legacy Management, LM-20, Grand Junction CO

Presented by:
Brian B. Looney
Savannah River National Laboratory

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Brian B. Looney *, Miles E. Denham *, Carol A. Eddy-Dilek *,
Margaret R. Millings *, Mark Kautsky **
* Savannah River National Laboratory, 773-42A, Aiken SC
** US DOE Office of Legacy Management, LM-20, Grand Junction CO

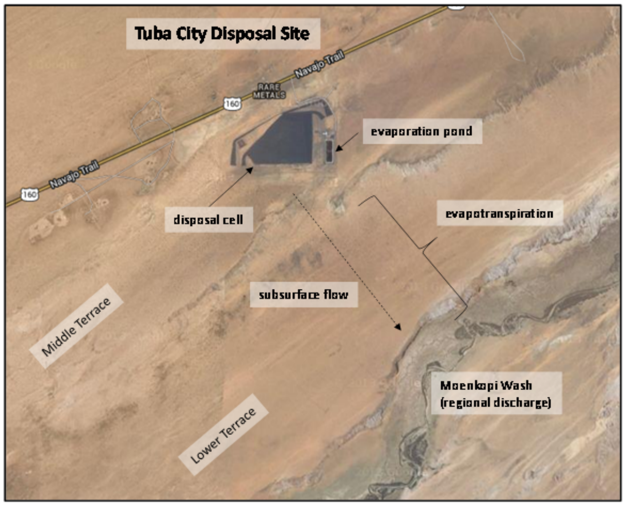
ABSTRACT

Management of legacy tailings/waste and groundwater contamination are ongoing at the former uranium milling site in Tuba City AZ. The tailings have been consolidated and effectively isolated using an engineered cover system. For the existing groundwater plume, a system of recovery wells extracts contaminated groundwater for treatment using an advanced distillation process. The ten years of pump and treat (P&T) operations have had minimal impact on the contaminant plume – primarily due to geochemical and hydrological limits. A flow net analysis demonstrates that groundwater contamination beneath the former processing site flows in the uppermost portion of the aquifer and exits the groundwater as the plume transits into and beneath a lower terrace in the landscape. The evaluation indicates that contaminated water will not reach Moenkopi Wash, a locally important stream. Instead, shallow groundwater in arid settings such as Tuba City is transferred into the vadose zone and atmosphere via evaporation, transpiration and diffuse seepage. The dissolved constituents are projected to precipitate and accumulate as minerals such as calcite and gypsum in the deep vadose zone (near the capillary fringe), around the roots of phreatophyte plants, and near seeps. The natural hydrologic and geochemical controls common in arid environments such as Tuba City work together to limit the size of the groundwater plume, to naturally attenuate and detoxify groundwater contaminants, and to reduce risks to humans, livestock and the environment. The technical evaluation supports an alternative beneficial reuse (“brownfield”) scenario for Tuba City. This alternative approach would have low risks, similar to the current P&T scenario, but would eliminate the energy and expense associated with the active treatment and convert the former uranium processing site into a resource for future employment of local citizens and ongoing benefit to the Native American Nations.

INTRODUCTION

The Department of Energy Office of Legacy Management (DOE-LM) is responsible for a number of former uranium milling and processing sites and the associated tailings disposal areas. These are collectively designated “Uranium Mill Tailings Radiation Control Act” (UMTRCA) sites. A majority of the sites are in arid regions in the western United States. The former uranium processing and tailings disposal site in Tuba City AZ (Fig. 1.) is an effective case study site that exemplifies the conditions, challenges and opportunities at UMTRCA sites.

Active management of legacy wastes and groundwater contamination is ongoing at Tuba City. The tailings have been consolidated and effectively isolated using an engineered cover system. For the existing groundwater plume, a system of 37 recovery wells extracts approximately 100 gpm (375 L/min) of contaminated groundwater which is then treated using an advanced distillation process. The treated water is reinfiltrated and the brine/reject is sent to a lined evaporation pond. This groundwater treatment system has operated for over 10 years. While the system has removed contaminants such as uranium, sulfate, nitrate, selenium and molybdenum from the subsurface, the operations have had limited impact on the plume.



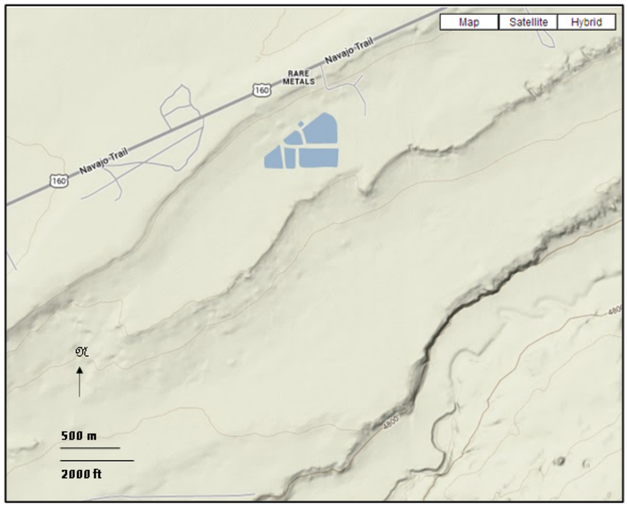


Fig. 1. Tuba City: former processing facility and disposal cell -- current configuration (left) and regional topography (right)

DOE-LM recently sponsored an independent technical evaluation of the status and prognosis for the cleanup at Tuba City [1]. The evaluation applied a range of scientific frameworks, including spatial, temporal, hydrologic, and geochemical frameworks, to the site specific conditions. The results of the hydrologic and geochemical framework evaluations are notable and provide a compelling basis for a protective and cost effective alternative end state.

APPROACH

In reviewing the available data, the independent team developed an overarching set of frameworks that potentially dovetail with existing conceptual models/approaches for UMTRCA sites and might provide areas of opportunity for the DOE-LM program. The frameworks provide a consistent way of organizing and interpreting complex data in a manner that supports environmental decision making. The individual frameworks include:

- Spatial Framework – places plume data within the spatial context of the sites from source to plume fringe; different locations within the spatial framework require different approaches to characterization, remediation and monitoring
- Temporal Framework – relates plume data to events in the history of the site from initiation of the processes that caused contamination, through remedial action with implications for effective long term monitoring
- Hydrological Framework – relates plume data to the physical forces driving plume movement including boundary conditions such as streams and, in arid climates, the capillary fringe
- Geochemical Framework – describes the interactions of plume constituents with aquifer materials and uncontaminated groundwater, as well as other geochemical processes affecting contaminant migration

When site information is organized into these frameworks the interrelationships become more apparent, facilitating holistic conceptual model development. As summarized below, Tuba City exemplifies the interrelations between frameworks reflecting the complicated history of the site, the location of the site relative to important geomorphic features, the influence of an arid climate, and geochemical interactions that have impact contaminant migration and hydrologic properties. The hydrologic and geochemical frameworks are particularly significant at Tuba City. These frameworks help clarify why the P&T system has underperformed and provide a technical basis for understanding the nature and extent of groundwater contamination.

RESULTS

Hydrologic framework – Where does the contaminated groundwater flow at Tuba City?

In all groundwater systems, water flows from sources (areas where water enters the subsurface) toward sinks (areas where water exist the subsurface). Flow lines and flow nets visualize the movement of water from sources to sinks. Within the subsurface the precise path of each flow line is driven by the strength and relative locations of the sources and sinks and the hydro-lithological characteristics of the aquifer. In many settings, nested flow nets develop [2]. Shallow flow lines move from local sources to local discharges and deeper flow lines move toward regional discharges. Importantly, there is little mixing of the flow lines so that water in the shallow flow zone will not interact with the deeper regional flow, resulting in the potential for groundwater to be vertically stratified. If contamination is present in the shallow flow zone, it will flow toward the local discharge and not the regional discharge. In Fig. 2, this general flow net concept has been overlain on the existing LM Tuba City site cross section. The former processing facility and zone of highest groundwater contamination are depicted along with the topography. The figure is annotated to show various areas of water discharge or loss. In this figure, some of the key flow lines are depicted – the potentially contaminated flow lines are color coded. The figure also demonstrates the general power of a flow net approach in providing site specific insights. Local flow is depicted as water moves from the former processing site to seepage and evapotranspiration (ET) areas in the lower terrace and in the terrace transition areas. Deeper regional flow lines are not impacted by the contaminants – unimpacted water is projected to flow beneath the site toward Moenkopi Wash.

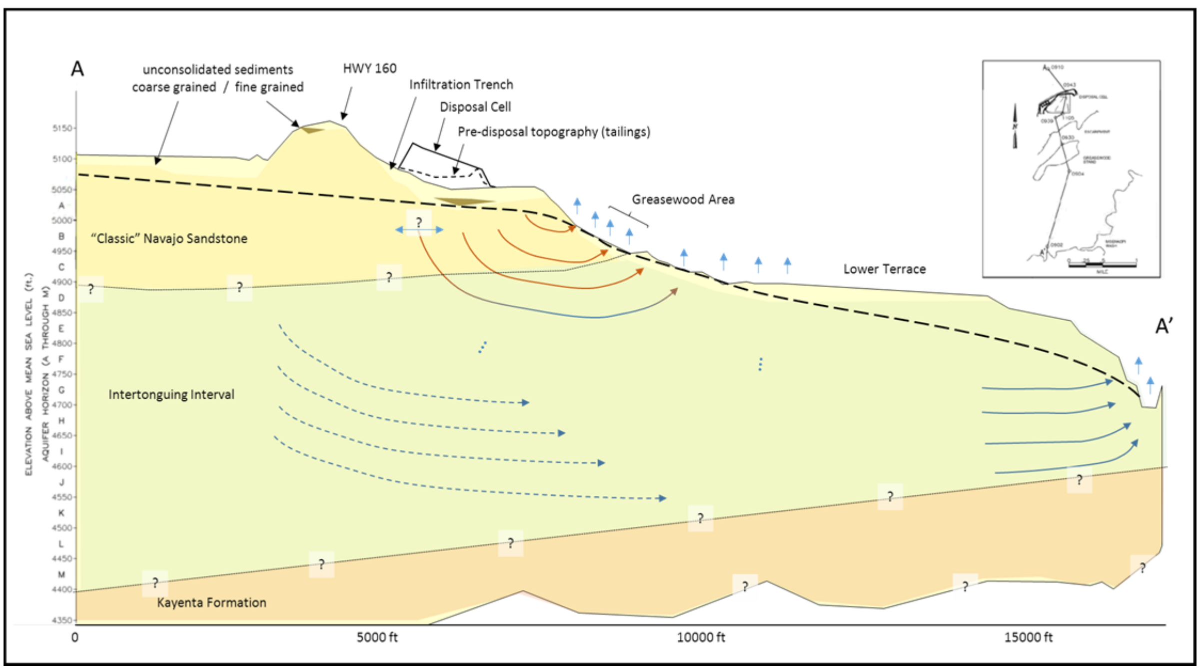


Fig. 2. Hydrogeologic cross section with key flow lines depicted; brown lines approximate the path of contaminated water.

Based on this conceptualization, evapotranspiration (ET) and seepage along the path of the plume are important parameters that control and limit the extent of the plume in this arid environment. This simple analysis is consistent with the *Final Site Observational Work Plan* (SOWP) [3]: "…the entire area between Moenkopi Wash and the middle terrace is essentially a ground-water discharge area via ET. The ET is dominated by transpiration across the lower terrace and the slope between the middle and lower terraces…." These conclusions were based on a variety of studies, including surveys of desert phreatophytes (plants that extract groundwater from depths up to about 10 m), climate data, the scientific literature, and engineering calculations. The SOWP data were organized into a water balance. The primary input of water into this aquifer system (>85%) flows laterally into the cross section from the left and flows under the former Tuba City Site. Note that only 23% of the water in the cross section exits at Moenkopi Wash (so that all of the local infiltration and the bulk of the lateral inflow are lost to evapotranspiration between the middle terrace and Moenkopi Wash). Thus, evapotranspiration is dominant component of the water balance at this particular site – accounting for about 77% of the water leaving the aquifer. The available information indicates that the extent of the contaminated plume will be limited and that it is improbable for milling site related contaminants to reach Moenkopi Wash. This conclusion would be unchanged if the fraction of water lost to evapotranspiration is anywhere in the range of approximately 20% to 100%.

Geochemical framework – Where do the groundwater contaminants go?

As described above, the basics of hydrology theory and results of early technical studies at Tuba City indicate that water originating at the former processing site is expected to discharge as evaporation, transpiration and seepage – an important related question is the fate of the constituents dissolved in the water. Fortunately, the behaviors of dissolved constituents in this scenario have been described and documented in a number of journal articles and reports [see 1]. This supporting literature identifies precipitation and accumulation of minerals such as calcite and gypsum in arid and semi-arid settings as a dominant process. Fig. 3 summarizes a conceptual model of the dynamic processes that occur in the vicinity of a "near-surface" water table in areas of evapotranspiration and outcrop.

In arid to semi-arid climates, desert phreatophytes extract water and associated dissolved constituents. For areas located above the plume at Tuba City, the extracted water would contain nitrate (a nutrient), sulfate, calcium and sodium (elements familiar to desert plants), and trace elements including uranium. Data from plant uptake studies presented in the SOWP [3] indicates that the groundwater contaminant concentrations observed in the lower terrace will not adversely impact plant growth and will not accumulate to harmful concentrations in plants. A survey of the plants present in the lower terrace [3] documented the presence of both obligate phreatophytes (plants that must access groundwater to live) and facultative phreatophytes (plants that can, but do not have to, access groundwater). These desert plants have evolved a number of mechanisms to limit the uptake and accumulation of dissolved constituents extracted from groundwater; for example, Purvis and Wright [5] document that dissolved constituents are liberated from the water during transport to the surface and that significant mineral accumulation occurs in the vicinity of the deep roots of desert phreatophytes sometimes forming "rhizocrete" deposits associated with root masses (Fig. 3).

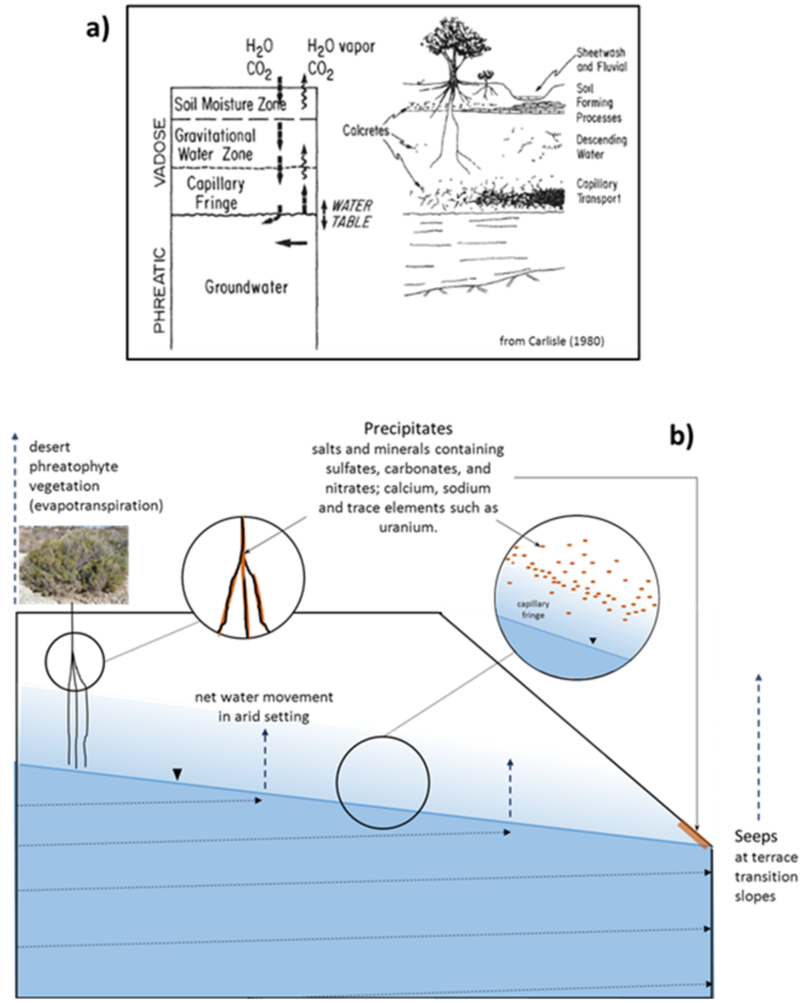


Fig. 3. Key geochemical processes associated with non-pedogenic mineral accumulations in arid and semi-arid settings – a) general concept [4] and b) annotated to highlight probable mineral accumulation zones for Tuba City

In addition to the phreatophyte extraction, groundwater can be lost as a result of capillary flow and evaporation. These abiotic physical processes result from the presence of moisture gradients, both liquid phase and vapor, in the vadose zone – moving from the capillary fringe toward the atmosphere. These gradients will result in a net loss of water to the atmosphere and chemical precipitation of dissolved groundwater constituents as solid salts and minerals. When the water table is close to the ground surface (such as areas near seeps), capillary forces will dominate and draw liquid water to the surface where it can evaporate and leave mineral solids. In areas where the water table is deeper (e.g., 5 to 10 m and below), capillary forces combined with vapor phase diffusion result in mineral precipitation in the vicinity of the capillary fringe. As noted above, the mineral assemblages would be expected to be similar to naturally occurring calcite and gypsum deposits.

Performance of the P&T System

The operation of the groundwater P&T system at Tuba City is currently the primary focus of on-site activities. The performance and cost of the system have emerged as overriding challenges, both in the near-term and projected into the future. The existing P&T system comprises 37 extraction wells that produce 100 gpm or about 375 L/min (nominal) for treatment using distillation; the distilled water is reinfiltrated into the subsurface using an trench installed on the NW edge of the stabilized tailings and the brine is discharged to a lined pond for evaporation. Implicit in an effective P&T remedy are the successful operation and efficient performance of the two major sub-activities: 1) in the subsurface, extraction of contaminated water and infiltration of clean water, and 2) above ground, robust and cost effective water treatment and disposal of secondary wastes. The Tuba City P&T system faces significant challenges, both in the subsurface and above ground.

The P&T system targets the near-field contaminant plume impacted by the historical operation at the former milling site. The disposal history for the Tuba City site is complicated resulting in a time dependent near-field geochemical framework that includes:

- Acid infiltration from the original uranium extraction process
- Alkaline infiltration from the later carbonate extraction process
- Precipitation of solid phases in the near-field aquifer as a result of the complex and perturbed geochemistry
- Natural groundwater flushing between the end of ore processing and the beginning of remediation
- Transient drainage of the tailings following construction of the disposal cell
- Flushing by treated groundwater water discharged to the infiltration trench

The aggressive fluids associated with the uranium processing and transient drainage dominate the near-field geochemical framework. The early acidic sulfate-rich fluids infiltrating from the tailings pile result in dissolution of calcite and precipitation of gypsum [see 1]. When the uranium extraction process switched from acid extraction to carbonate extraction, infiltration into the subsurface would have been a high pH solution with high concentrations of carbonate; under these conditions, calcite would precipitate. Thus, precipitation of various solid phases would clog pores and reduce aquifer permeability in the near field plume. Further, these solids impact reliable operations of pumping wells throughout the near-field area targeted by the P&T system.

Effective extraction of groundwater is a crucial step in operating a viable P&T system. Unfortunately, the subsurface portion of the P&T has underperformed in the initial years of system operation. The original design basis focused on extracting 100 gpm (375 L/min, increasing to about 200 gpm (750 L/min) in a second phase) with a goal to flush one or two “pore volumes” of contaminated groundwater beneath the stabilized tailings. The added driving force of the water from the infiltration trench is intended to increase hydraulic gradients and to accelerate the flushing. Field experience has tempered the original objectives. The bulk of the water for treatment is extracted from only a few of the recovery wells and a large number of the wells in the system do not produce significant amounts of water (e.g., 0.1 gpm or less). Over an extended timeframe, the uneven hydraulic performance would correlate with water flow through some areas (i.e., flushing) and not through other areas. Further, the uneven hydraulic performance is a manifestation of natural heterogeneity – active flow paths will clean up relatively rapidly while less active flow paths will clean up slowly and result in plume tailing and the need for many equivalent pore volumes to achieve remedial objectives throughout the groundwater.

For some of the target contaminants, such as sulfate, the aquifer currently contains a large quantity of precipitated solid source material that would extend the remediation timeframe "indefinitely" (i.e., until the precipitated minerals are dissolved and removed). The timeframe for other contaminants, such as uranium, may also be extended since these elements are often incorporated in/on gypsum, calcite and silicate minerals. Significantly, the portions of the plume that have been impacted by the highest levels of contamination would have been subject to highest levels of solids precipitation and would be the most important to flush. But these areas would produce less water while requiring many more flushing volumes to achieve remedial goals. This discordance between fundamental natural controls/processes in the subsurface and "remediation performance needs" is a significant and overriding challenge.

The above ground portion of the P&T system is also challenging. The objective of the above-ground treatment operations is to separate the contaminated groundwater into two effluent streams. The bulk of the influent volume (approximately 90 %) is converted to clean water for reinfiltration. The remainder of the volume contains all of the contaminants and other dissolved constituents from the original groundwater along with any chemicals added during treatment. As noted above, this brine is directed to a lined basin for evaporation. Currently, the water separation is performed using a state-of-practice distillation system (a "falling-film vapor recompression distillation process"). Further, the contractor team has incorporated a number of components/actions to optimize the distillation, including: various types of heat recovery, solar heat collection system (installed for use in the future), supplemental solar photovoltaic energy, groundwater pretreatment (softening), and frequent process monitoring to minimize scaling. Use of an innovative distillation process and attention to energy recovery, in particular, have partially mitigated the high energy requirements associated with evaporating large volumes of water.

Despite the high quality efforts, the treatment system has significantly underperformed (operating less than 20% of the time in 2012) and has incurred high operation and maintenance costs. Large-scale distillation systems are complex and subject to shut down due to failures in any of the numerous processing steps, scaling or clogging of equipment/heat exchangers/transfer lines, solids build-up on the "falling-film" distillation membranes, leakage of vacuum/vapor or water, scheduled or unscheduled cleaning or maintenance, and other factors. Thus, compared to a number of alternative water treatment methods, large-scale distillation systems are relatively labor intensive and typically are not as robust or reliable. If P&T is continued at Tuba City, two viable treatment alternatives that could be considered include reverse osmosis and spray evaporation [1]. Regarding the P&T system, the independent technical review concluded:

- The P&T system will require an extended timeframe to achieve remediation objectives; if P&T is determined to be necessary, many decades of operation would be needed
- If P&T is continued for an extended timeframe, DOE should initiate an orderly process to transition the above ground treatment technology to an alternative technology such as reverse osmosis and/or spray evaporation
- Reliable installation and continuous operation of recovery wells represent an ongoing challenge; a program of well replacement/relocation/rework will be needed if long term operations are required
- The issues and challenges associated with all aspects of the P&T system, both above ground and in the subsurface, increase the urgency and potential significance of efforts to document natural attenuation processes occurring along the flow path.

Next Steps – Confirming the Alternative Conceptual Model

Confirming and incorporating the evapotranspiration discharge boundary zone into the site conceptual model are keys to the future management of Tuba City and to the development of protective and cost effective site strategies. A focused characterization study of the critical transition region from the middle to lower terrace and the lower terrace to the Greasewood area would support both conceptual model development, and provide a strong technical basis for productive stakeholder and regulatory interactions. Processes at such boundaries produce distinctive geochemical signatures. In some cases, the required characterization would be straightforward, such as surveying the area for surficial mineral signatures associated with seeps. Characterization of the depth profiles in the vadose zone (in both plume areas and nearby background areas) would define the presence and nature of non-pedogenic minerals above the water table.

An updated interpretation of existing aerial gamma survey provides an illustrative case study of the application of one of characterization methods recommended by the technical team. A high resolution dataset collected for the U.S. Environmental Protection Agency (Fig. 4) provides a detailed depiction of the local terrestrial gamma exposure rates/patterns. The overflights for these datasets were performed after 1990; therefore, remediation of localized surface contamination on/near the site was complete and the tailings were consolidated and covered at the time of the high resolution data collection. Note that all of the data on the local high resolution terrestrial gamma exposure map are within the background data range from the North American NURE data and all of the data are below UMTRCA soil guidelines (e.g., 40CRF192) and unrestricted public access guidelines from NRC and international organizations (e.g., 1 mSv/yr as designated in 10CFR2).

Despite the low levels of radioactivity, the pattern of gamma exposure levels provides significant insight into background conditions and potential transport pathways for milling-related radionuclides in the vicinity of the Tuba City site. One of the most notable features in Fig. 4 is the band of high natural gamma exposures along the entire surveyed reach of Moenkopi wash. This gamma is associated with the evaporative accumulation of minerals in a regional discharge area and the accumulation of heavy minerals due to physical sorting and concentration in a surface water environment that is subject to flash flooding. The background gamma exposure levels are the weakest in areas of surficial sand dunes and relatively higher in other areas.

In the vicinity of the mill site, there are detectable gamma signatures toward the east and toward the south (Fig. 5). The eastern gamma signature was previously observed [see 1] and attributed to windblown dust from the mill site during the period when the tailings were not consolidated and covered. The gamma exposure levels are notably low over the area of the tailings cell. Importantly, the data indicate that the LM cleanup actions – collection/consolidation and covering of contaminated tailings and soils – were relatively effective since the terrestrial gamma exposure levels measured in the post-1990 aerial gamma were significantly below the historical values measured in the 1960s. The gamma signature to the south of the mill site is particularly interesting because it is indicative of groundwater transport and discharge (either at seeps or evapotranspiration boundaries) and/or indicative of erosion and overland transport of contaminated surficial soils from the middle terrace to the lower terrace. The southern transport pathway is limited in scale and is consistent with the expected short flow distances for the uppermost groundwater flow lines or limited erosion. Evaluation of gamma signatures associated with specific radionuclides (e.g., uranium) confirms and further supports the interpretation of the total terrestrial gamma patterns [1]. Note that there is no southern transport pathway observed for the windblown area (the surficial contaminants to the east) – possible evidence that supports the subsurface/groundwater pathway from the main mill site area. For the southern transport direction, however, the aerial gamma surveys alone do not allow the alternative pathways to be differentiated and additional lines of evidence (e.g., study of contaminant profiles above the water table) would be needed to further refine and quantify the conceptual model.

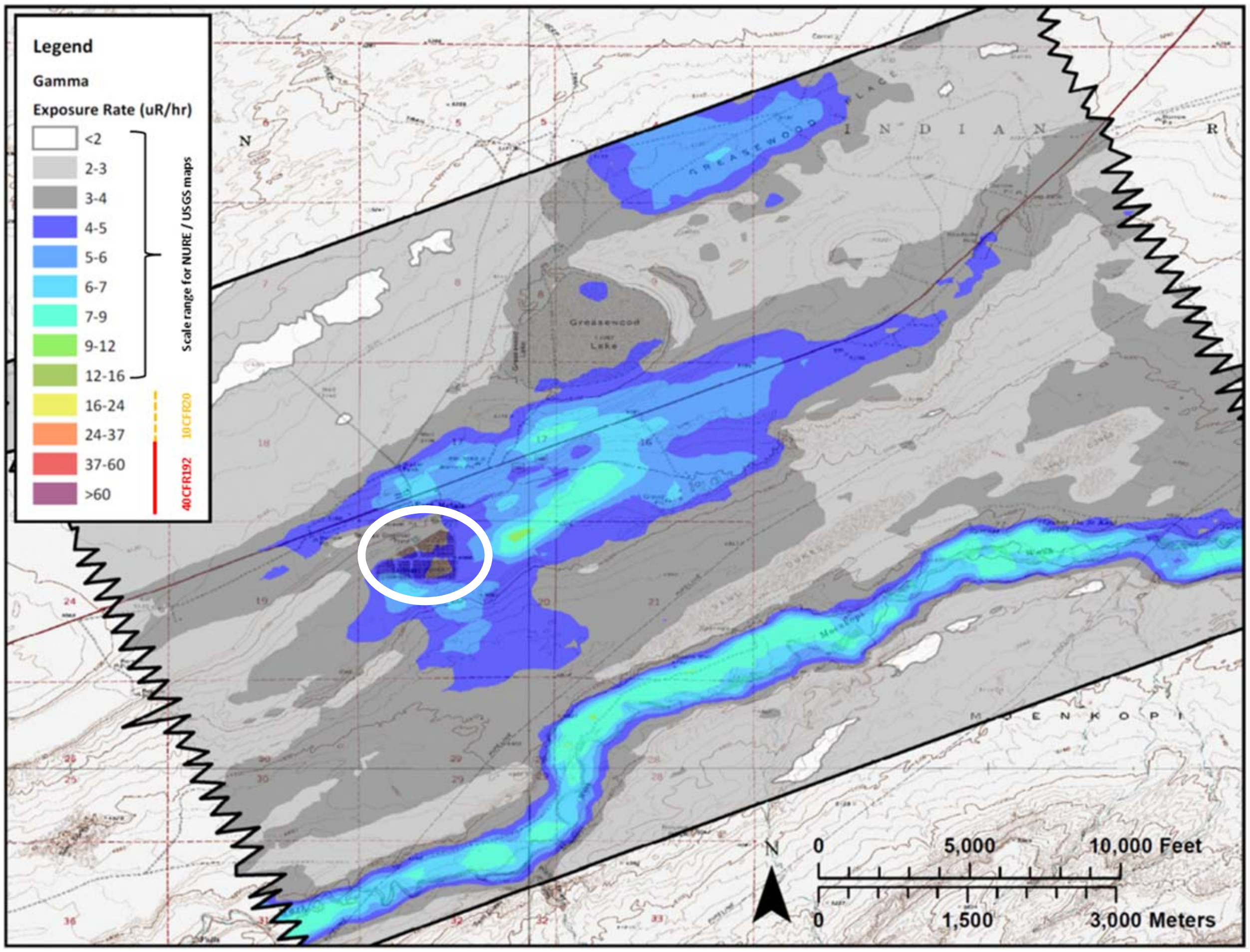


Fig. 4. High resolution terrestrial gamma exposure map (uR/hr) in the vicinity of the Tuba City Mill Site (circled)

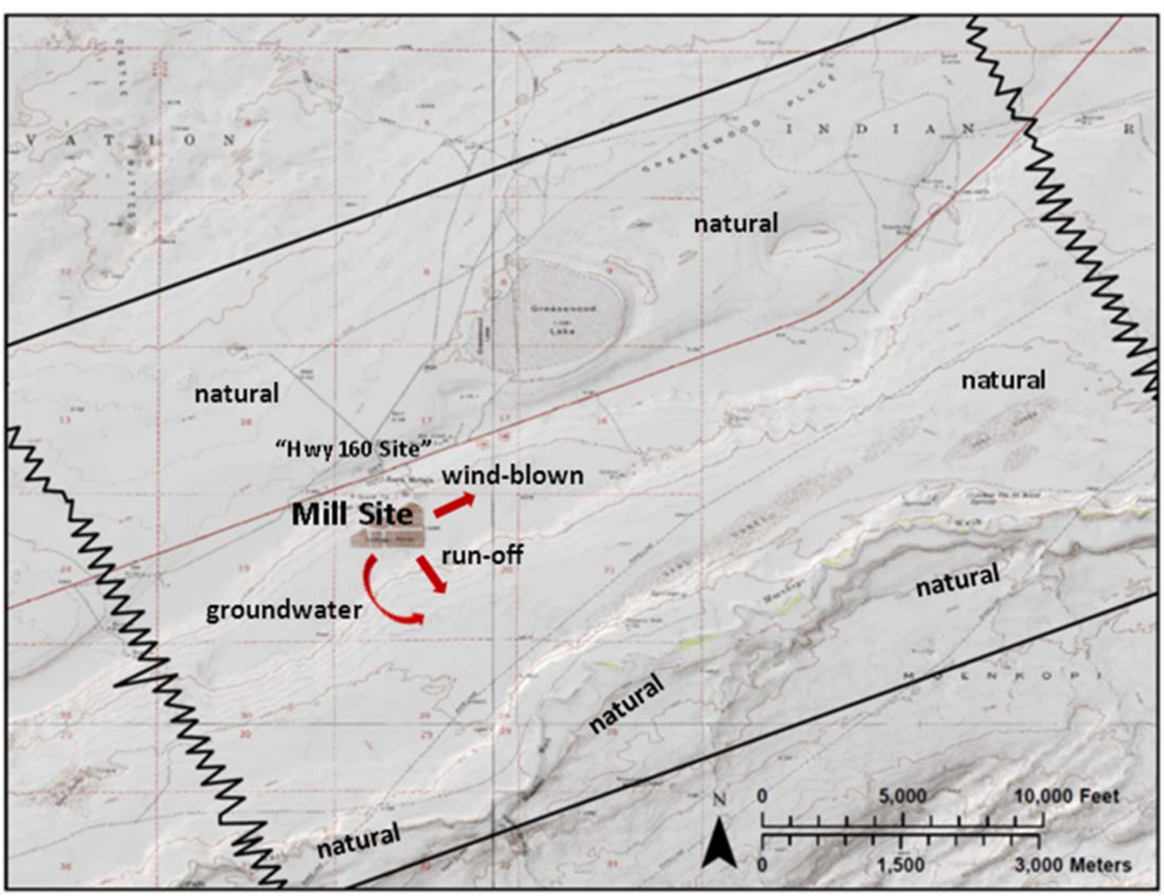


Fig 5. Summary of predominant transport vectors in the vicinity of the Tuba City Mill Site

Long Term Strategy Development

Developing a safe, viable and productive long-term future use (end-state) for the Tuba City Site is an important component in strategic planning. At several DOE-LM sites, the land and facilities are transferred to the local community for transition to beneficial reuse and development. For example, at the former Mound Site, the "brownfield" land and facilities have been transitioned to a local public private partnership, the Mound Development Corporation, that operates the successful Mound Advanced Technology Center. The Mound Site transfer to beneficial reuse took many years and involved a careful-stepwise multi-party process to assure the safety of the new site occupants and the surrounding community. The results of the significant effort, however, have been positive for both DOE and the community. Currently, the Mound Advanced Technology center houses 11 businesses with 256 employees that perform research, development, testing and production of high-tech products and processes. A similar redevelopment process is underway for a former uranium milling site in Gunnison CO.

Tuba City Site appears to be a good candidate for reuse and represents a potentially significant resource for future employment and ongoing benefit to the Native American Nations. A schematic of the steps for such a process is depicted in Fig. 6. The initial stage of the effort would be formulation of a shared vision for redevelopment with the local community and regulators. As shown, the redevelopment process is contingent on confirming that onsite risks are low and that the risks can be controlled. Further, redevelopment would be contingent on confirming the conceptual models from the hydrologic and geochemical frameworks – specifically, that the natural processes of the Tuba City system will effectively attenuate and limit the extent of the plume and result in low offsite risks. If these contingencies are met, then the site can be prepared for the transfer by discontinuing pump and treat and transitioning the groundwater strategy to monitored natural attenuation, removing infrastructure that would not be transferred, and general clean-up (e.g., removing/isolating the solids collected in the evaporation pond). Some of the on-site infrastructure would be highly desirable to a potential redevelopment candidate, particularly the large photovoltaic power array, the solar heat collection system and the buildings. It is likely that the availability of such resources would increase the competition for candidate tenants. Based on scale of the Tuba City, it is feasible that one or more small businesses could occupy the site and perform small-scale green manufacturing with the potential to employ a significant number of local citizens.

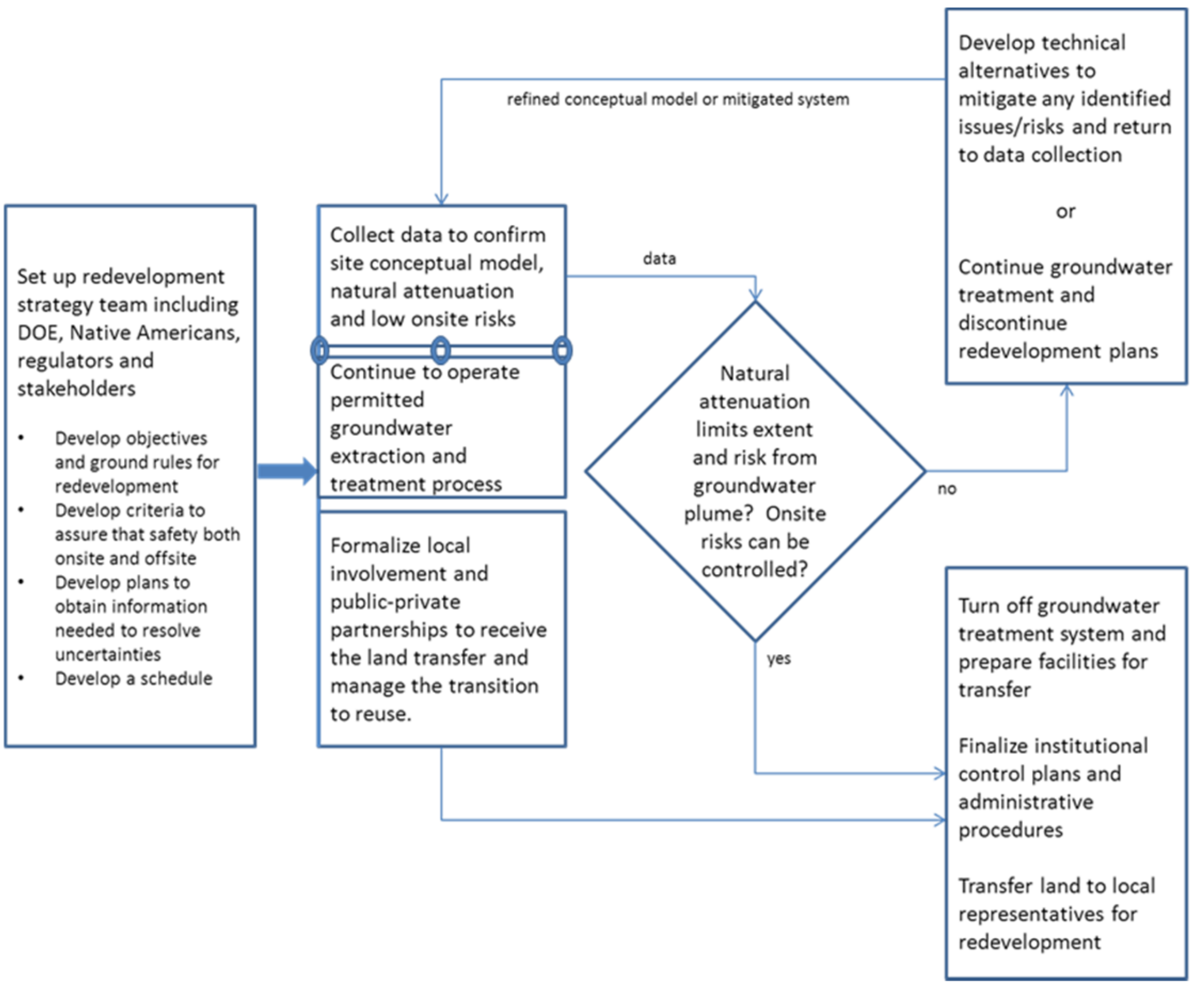


Fig. 6. Summary of a process to transition the Tuba City Site to beneficial reuse

CONCLUSIONS

The technical evaluation of Tuba City suggests that hydrologic and geochemical frameworks appear to be particularly relevant to contaminated sites in arid settings such as the western portion of the United States. A simple flow net analysis demonstrated that groundwater contaminants beneath the former processing site are in the uppermost portion of the aquifer, and exit the groundwater as it flows into and beneath a lower terrace in the landscape. The evaluation indicated that contaminated water will not reach the Moenkopi Wash, a locally important stream. Instead, shallow groundwater in arid settings such as Tuba City is transferred into the vadose zone and atmosphere via evaporation, transpiration and diffuse seepage. In the near-field, the geochemical evaluation demonstrated that mineral precipitation/dissolution in the aquifer would decrease the effectiveness and reliability of groundwater extraction and increase the remediation timeframe. In areas of downgradient groundwater evaporation, transpiration or seepage, dissolved constituents are projected to precipitate and accumulate as minerals such as calcite and gypsum that contain contaminants in the form of accessory minerals such as carnotite. These mineral accumulations will be in the deep vadose zone (near the capillary fringe), around the roots of phreatophyte plants, and near seeps. The hydrologic and geochemical controls common in arid environments such as Tuba City work together to limit the size of the groundwater plume, to naturally attenuate and detoxify groundwater contaminants, and to reduce risks to humans, livestock and the environment. Further, the evaluation provides the basis for an alternative end state for this site.

At Tuba City, this framework approach has a high potential to be transformational providing the technical bases: to document protection of human health and the environment, for safe shutdown of an “ineffective” P&T system, and to support sustainable and beneficial future use. If successfully transitioned, equal or improved protection of the environment could be achieved with a significant potential for cost savings and an improved benefit of the land and its resources to the local community.

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