Incorporating Climate and Regional Setting into Realistic-Efficacious End States for Contaminated Sites – 15386

Brian Looney *, Miles Denham *, Carol Eddy-Dilek *, Bill Dam **, Mark Kautsky **
* Savannah River National Laboratory, Environmental Stewardship Directorate, Aiken SC 29808
** U.S. Department of Energy, Office of Legacy Management, Grand Junction CO

ABSTRACT

Data from former uranium milling and processing sites in the arid western United States (US) exemplify a conceptual model in which climate and geomorphology, and the associated geochemical and hydrological conditions, control the fate and transport of contaminants. In arid settings, shallow groundwater is transferred to the vadose zone and atmosphere via evaporation, transpiration and seepage. During these transfers, dissolved constituents accumulate in the vadose zone near the capillary fringe and around the roots of phreatophyte plants. In areas where the water table is shallow, minerals will also accumulate at the surface as a result of capillarity and evaporation. Two western-semiarid former milling sites, Tuba City AZ and Riverton WY, represent deep and shallow water table cases, respectively. At Tuba City, hydrological and geochemical factors limit the size of the groundwater plume and reduce the potential for contaminated water to crop at the locally significant Moenkopi Wash. At both sites, minerals that have formed in the groundwater and vadose zone in the near-field and mid-field areas will sustain elevated groundwater concentrations of anthropogenic contaminants, such as sulfate and uranium, for an extended timeframe. The presence of secondary source mineral accumulations in the vadose zone in the mid-field area at Riverton is significant because episodic flooding occurs at this site. The contaminant distributions and plume dynamics observed at arid sites contrast with sites in the eastern US with stronger hydrologic driving forces and weaker evaporative processes. In both arid and non-arid settings, long term contaminant behaviors and concomitant exposure, risk and remediation timeframes are controlled by regional conditions and climate and by local modifying factors. As a result, incorporating these conditions and factors into environmental management decisions provides new opportunities to develop more robust, sustainable and technically-defensible end states.

INTRODUCTION

The U.S. Department of Energy (DOE) is responsible for environmental stewardship of sites and facilities in diverse settings. The DOE Offices of Environmental Management (EM) and Legacy Management (LM) manage any contaminated soil and groundwater at most of these sites. Figure 1 summarizes the magnitude of the challenge. DOE EM was initially responsible for 107 contaminated sites in 35 states. Of these, major cleanup activities have been completed at 90 sites leaving the current portfolio of active DOE EM sites – 17 sites in 13 states. Some of the challenges at these active EM cleanup sites (e.g., $6.5x10^{12}$ L of contaminated groundwater and $4x10^{7}$ m^3 of contaminated soil and debris) are listed in Figure 1. Following major cleanup activities under EM, a number of the completed sites require additional time, active management and (sometimes) additional remedial actions to reach their final environmental remediation goals. DOE LM is responsible for completing the cleanup at these sites as well as for the early uranium mining, milling and processing sites that were associated with defense programs. DOE LM is currently responsible for 90 contaminated sites in 26 states and Puerto Rico. Notably, LM manages extensive quantities of mill tailings and debris (in stabilized and protected tailings cells) and is responsible for identifying and addressing the safety of thousands of abandoned large and small uranium mines. The DOE LM portfolio will increase over time as additional sites are transferred from EM. Together, the DOE EM and LM efforts represent the largest and most challenging environmental cleanup program in the world. To help meet this challenge DOE has emphasized safety and risk reduction, effectively utilized existing engineering and technology, and incorporated innovative science and emerging methods, as appropriate, to address some of the most difficult "intractable" problems. For soil and groundwater, a small technical assistance program has proven to be an effective paradigm for putting science to work in solving environmental challenges. In the past eight years, 25 teams have visited 11 DOE sites and made recommendations that yielded an estimated cost savings over $100M, generating a return on investment of 30:1 for the program [1]. Importantly, innovative technologies have been implemented across the nation to cost-effectively protect and restore the environment. In performing the technical assistance activities, two recurring themes emerged: 1) the importance of matching technologies to site specific needs and spatial-temporal conditions, 2) the over-riding role that hydrologic and geochemical boundary conditions play in governing the behavior and associated risks of contaminants in soil and groundwater. Further, recent work at LM mining and milling sites in the arid to semiarid western U.S. highlights the important role of regional characteristics (climate and geology) and the value of considering these factors in developing cleanup strategies and defining stable-sustainable end states that are compatible with the regional conditions. A brief description of the matching process, the role of regional factors and how these can be used in developing end-states are provided in the following sections.

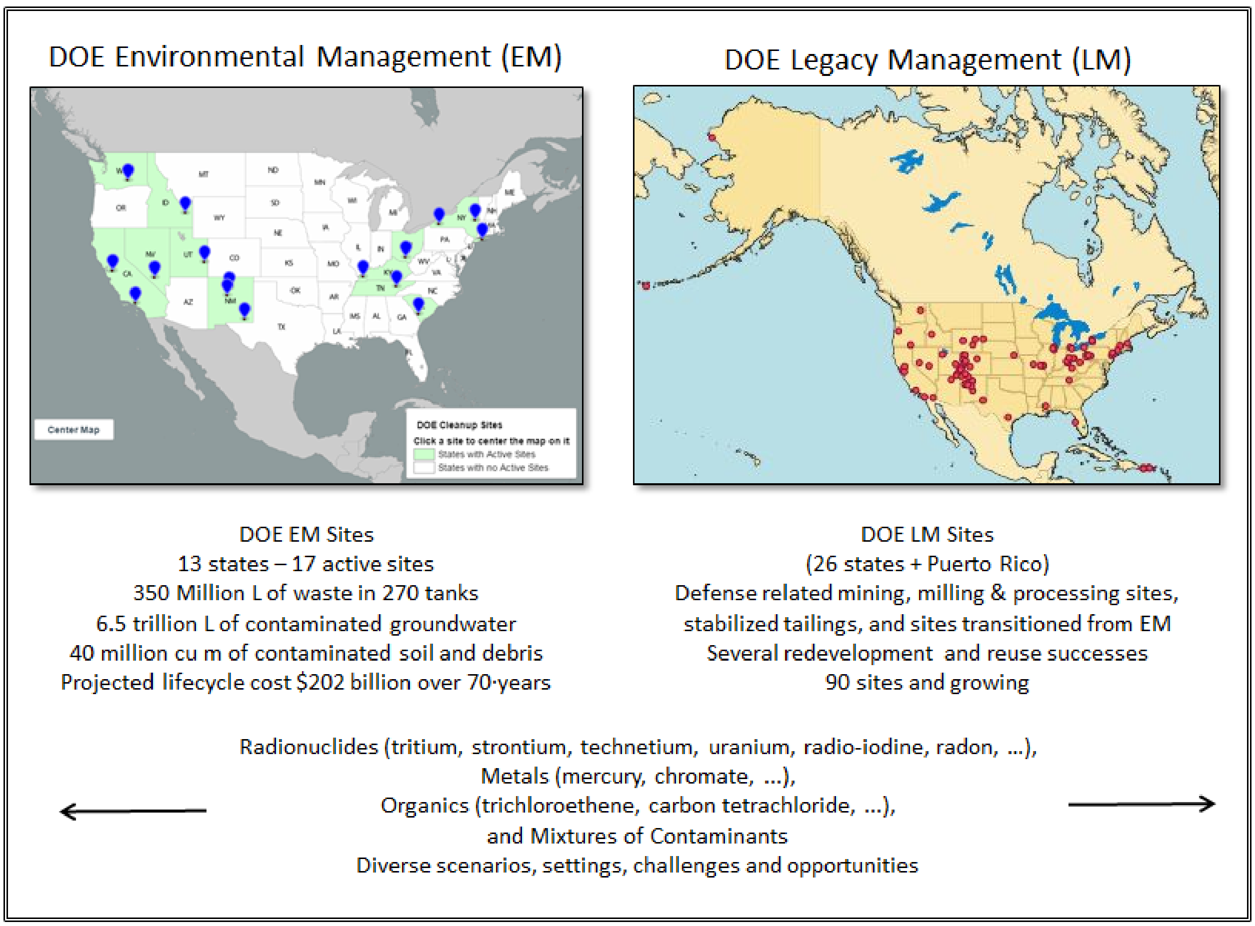


Fig. 1. Scope of the U.S. Department of Energy Environmental Management and

Legacy Management Challenge.

Figure 2 provides a simplified conceptual plan view diagram of a facility that has impacted the surrounding environment. The three ovals – the disturbed zone, the impact zone, and the transition/baseline zone – represent different portions of the affected environment. Each of these zones has a different character and provides opportunities for technology matching. The disturbed zone received relatively high levels of contaminant. The impact zone often manifests as a primary contaminant plume that contains lower levels of pollutants than the disturbed zone but still represents a potentially significant present or future risk. The transition/baseline zone contains contamination at relatively low concentrations but impacts relatively large volumes of water (or air or soil). For a real-world target problem, the contaminated areas are not simple ovals. Instead, contamination occupies a complex three-dimensional geometry and encounters multiple geochemical conditions and geological materials as it travels through subsurface (vadose zone and groundwater), surface water (e.g., wetlands and streams), and/or the atmosphere. A site-specific technology assessment process considers these multiple levels of complexity to identify areas of opportunity.

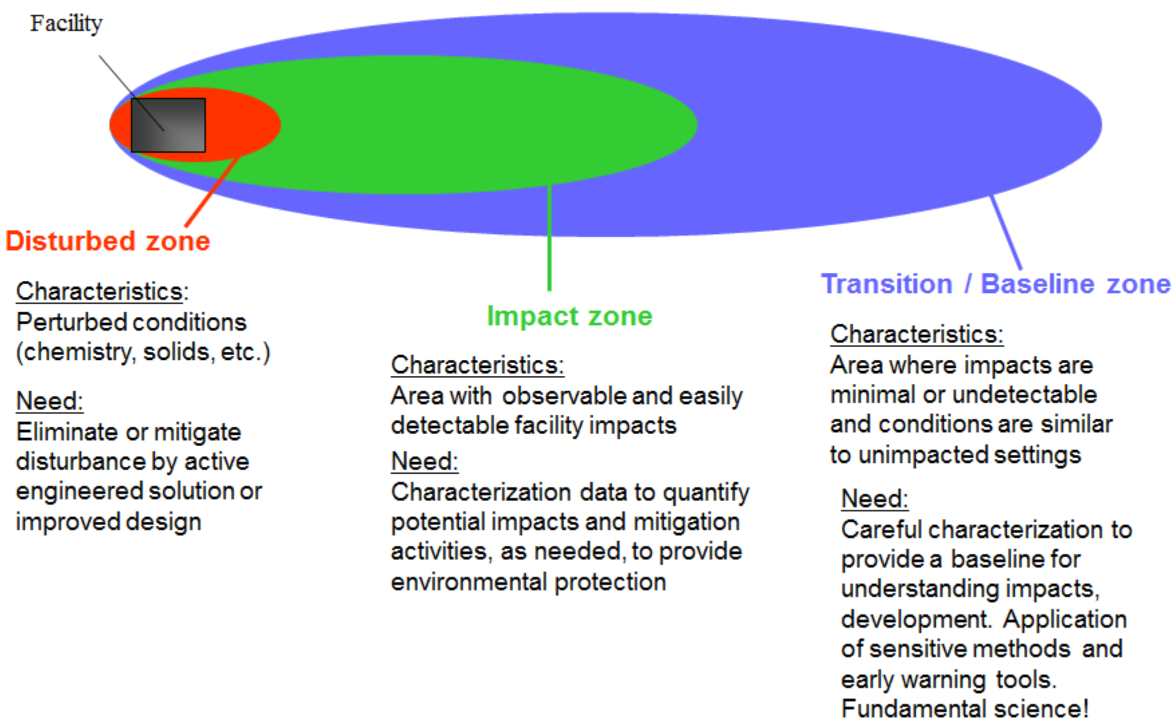


Fig. 2. Simplified conceptualization of facility impacts on the surrounding environment and technology matching principles developed by SRNL.

In the case of groundwater contamination, the changing size and structure of a contaminant plume is a dynamic process with conditions that change in both space and time. Figure 3 schematically depicts the general trends of plume expansion stabilization and shrinkage and overlays the examples of potential matches of remedial technologies for application – this approach is used to help apply appropriate technologies and to transition technologies (e.g., from active to passive) at appropriate times.

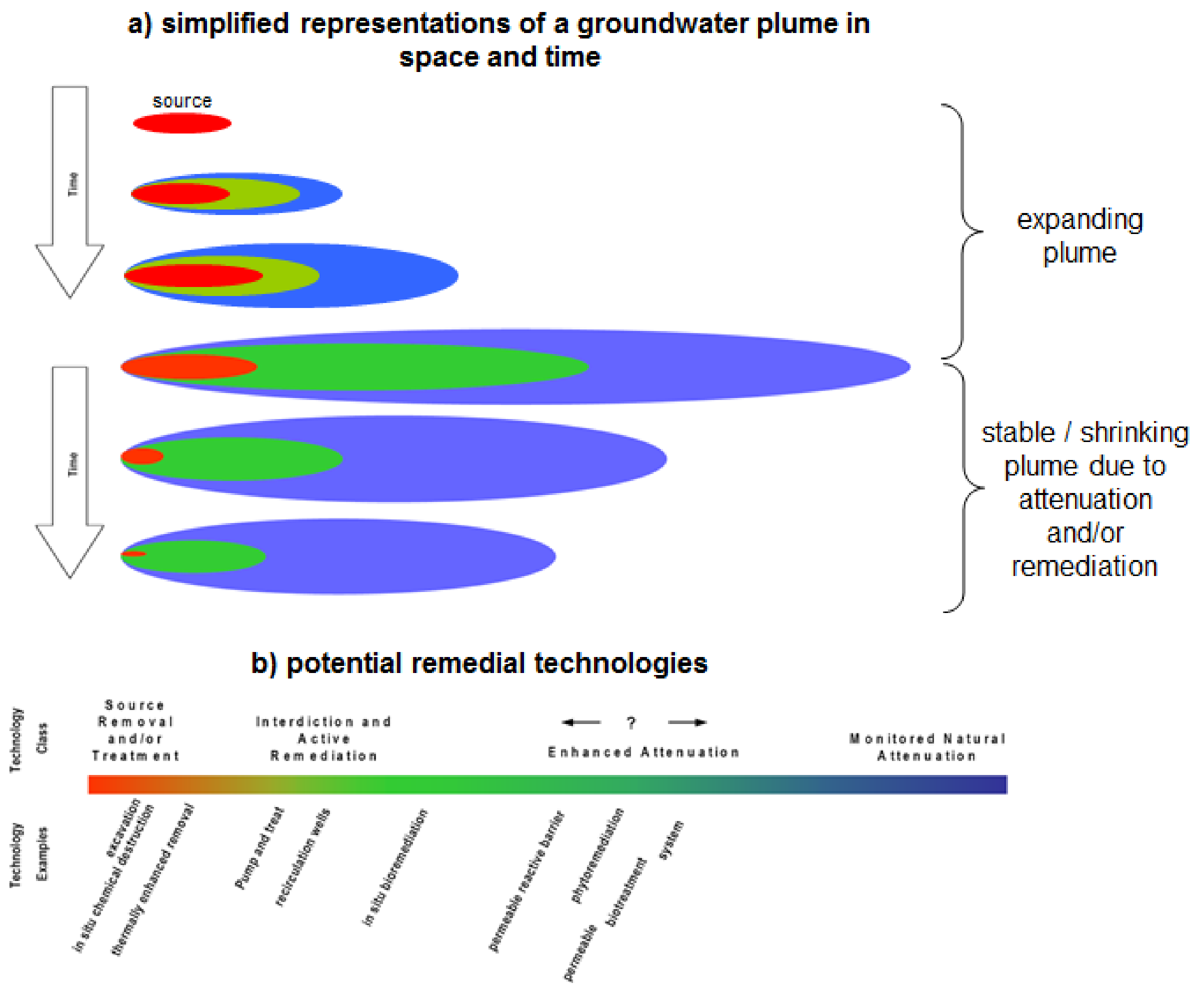


Fig. 3. Simplified depiction of plume structure over time and the matching of potential remediation technologies.

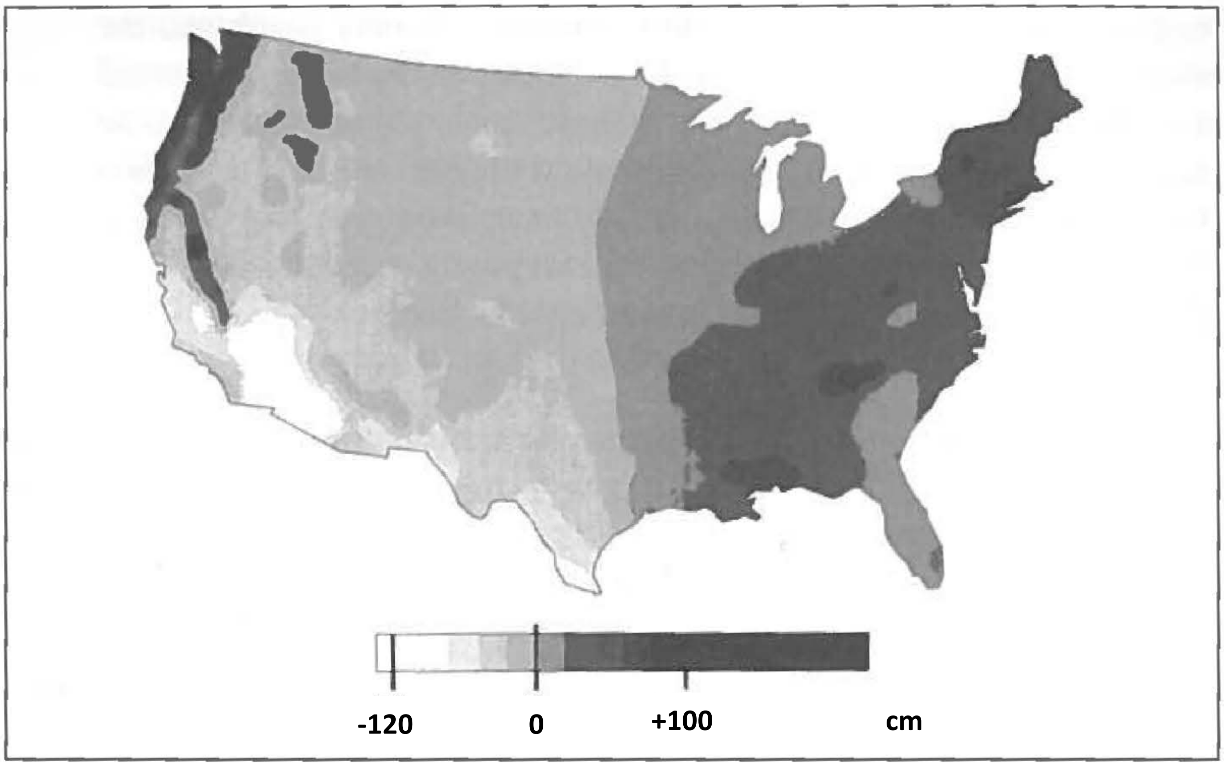
The DOE EM and LM technical assistance activities have applied the general matching concepts described above at numerous sites throughout the United States – generating technology listings that are generally binned into several key categories such as: “viable and recommended”, “viable but not recommended”, or “not viable”. Viable technologies are generally those that would physically work at the target site. Those viable technologies that were “recommended” are well matched to site conditions so that they would be expected to be relatively efficient and effective compared to those that are “not recommended”. Those technologies that are designated “not viable” typically will not work at the site due to some type of physical, chemical or hydrological constraint (e.g., technologies that require injecting liquid reagents into a clay zone). In most cases, there is more than one technology in each bin providing flexibility for the DOE-contractor-regulators-stakeholder team to further evaluate options in the context of local conditions and needs. Past technical assistance results highlighted the need for aggressive technologies such as steam enhanced extraction or excavation in the red (disturbed) zone, mass removal, in-situ stabilization and enhanced attenuation in the green (impact) zone, and natural or enhanced attenuation in the blue (transition) zone.

In most technical assistance evaluations hydrologic and geochemical boundary conditions are over-riding factors that govern the behavior and associated risks of contaminants in soil and groundwater. Hydrological and geochemical boundary conditions are regional in nature and are determined by climate, geology, and geomorphology. As shown in Figure 4, a simplified analysis of the role of climate in environmental contamination evaluation can be demonstrated using an estimate of water balance [3]. This map denotes the relationship between regional water inputs (average precipitation) and water outputs (potential evapotranspiration). Average precipitation is equal to the cm of water that fall on the land surface as rain, snow or sleet in an average year. Evapotranspiration is the reverse of precipitation resulting from transport of water from the earth back to the atmosphere through evaporation and plant transpiration (potential evapotranspiration is a maximum value assuming water is available for transfer at all times).

Dark shading (positive numbers) on Figure 4 indicates regions where more water is available than can be returned to the atmosphere by evaporation and plants. Such regions have substantial natural driving force (downward water flow) for moving contamination through the vadose zone and into groundwater. The importance of other sources of water (e.g., water line leaks or outfalls) is reduced.

Light shading (negative values) on Figure 4 indicates regions where the general climactic balance is reversed (less rain and/or more evaporation and transpiration). Anthropogenic sources of water or spatial/temporal changes in surface water infiltration/management (e.g., after a wildfire) are relatively more important in these regions and can dominate downward water migration and contaminant hydrology. Characterizing and controlling the sources of water can be significant in reducing environmental impacts at such sites.

The trends in the map are useful in making engineering judgments about technologies. For example, a number of innovative cap systems rely on slope, capillary barriers and plants to maximize runoff, evaporation and transpiration. Since these processes are fundamentally controlled by large-scale climactic driving forces, the map provides a good idea where the innovative caps will be viable and provide robust performance (light shading) and where they will be less appropriate and more subject to failure (dark shading). An initial evaluation of this type can be supplemented by a more detailed evaluation of climate variability/dynamics and specific design features.



Shading represents water balance = (precipitation minus potential evapotranspiration)

Fig. 4. General Climactic Water Balance of the Continental United States [2] [3].

The simple map shown in Figure 4 does not directly predict vadose zone thickness. In "arid" eastern Washington, the vadose zone ranges from 0 to approximately 100 m thick at the Hanford Site; in "humid" South Carolina, the vadose zone is similar in thickness (0 to 60 m thick) at the Savannah River Site. The map does not provide a complete water balance because it does not account for runoff, excursions in climate, periods where potential evapotranspiration is not possible due to moisture limitations, or localized factors. Negative values on the map do not indicate areas where there is no recharge (water is still present in the vadose zone and moving in response to gravity and capillary forces). Even in this highly simplified form, however, the Figure 4 does provide interesting information on the general nature of environmental conditions in various regions, the relative importance of possible water sources at contaminated sites, and the relative applicability of alternate environmental strategies. This map also indicates why regional climate factors are important to federal agencies like the DOE and the Department of Defense (DoD), large industries, and others. These organizations have facilities located throughout the climactic regime and their environmental policies and procedures must be appropriate for the entire range of conditions.

EXTENSION OF THE TECHNOLOGY MATCHING PROCESS

Based on the experiences of the technical assistance teams, the technology matching process can be extended to more explicitly incorporate regional factors. Figure 5 depicts the relative risk associated with sites that range from arid (on the left) to humid (on the right). The quantitative measure of water balance and shading of the scale bar are consistent with Figure 4. The vertical scale represents the mass discharge (or another relevant measure of the level of contaminant source). The quantitative nature of this scale will be contaminant specific (e.g., [4]). The highest risks are generally associated with the lower right corner (high driving force for contaminant migration and large contaminant source) and the lowest risks are associated with the upper left corner (low driving force and small contaminant source). The color coding for relative risk in the main field of Figure 5 align with the red (disturbed), green (impact) and blue (transition) zones described above. The right side of the figure provides examples of some of the most common modifying factors that influence risk. These include depth to groundwater, distance to receptor, contaminant toxicity and mobility, attenuation processes, and interactions with the surrounding ecosystems/environment. The modifying factors would move a site either upward or downward in the risk field. For example, increasing the distance to a receptor or significant biogeochemical attenuation would tend to reduce risks and move a site upward on the graph (e.g., A to A'). Conversely, if the contaminants are highly toxic or mobile, the position of a site on the graph would move downward (e.g., B to B').

Figure 6 overlays remediation technologies (the technology classes defined above) onto the risk field. For sites with significant sources and risks, source zone contaminant destruction, removal or stabilization are appropriate. This type of action is important to long-term success and the sustainability of EM/LM responses. Further, such action is explicitly required in most regulatory paradigms (e.g., removal of source material and principal threat wastes to the extent practicable). The area near the bottom of the figure is shaded to emphasize these general principles. Active treatment of the contaminated media or interdiction of the contamination are identified in the center of the risk field. Enhanced and natural attenuation, respectively, occupy the upper (lowest risk) portion of the figure. Performing the identified remedial action moves a site upward on the graph (by reducing the quantity or mass discharge of contaminant). This movement is depicted using dashed arrows. Note that the amount of progress resulting from a cleanup action can vary. Further, all remediation classes will not be needed at all sites. For example, it may be possible to move from source removal to enhanced or natural attenuation at some sites. In some cases, an extended period of active treatment is appropriate; where possible, however, actions to implement a passive-stable-sustainable enhanced attenuation are generally desirable. Such technologies can perform better than the active treatments and provide a path to natural attenuation (e.g., [5] [6] [7]).

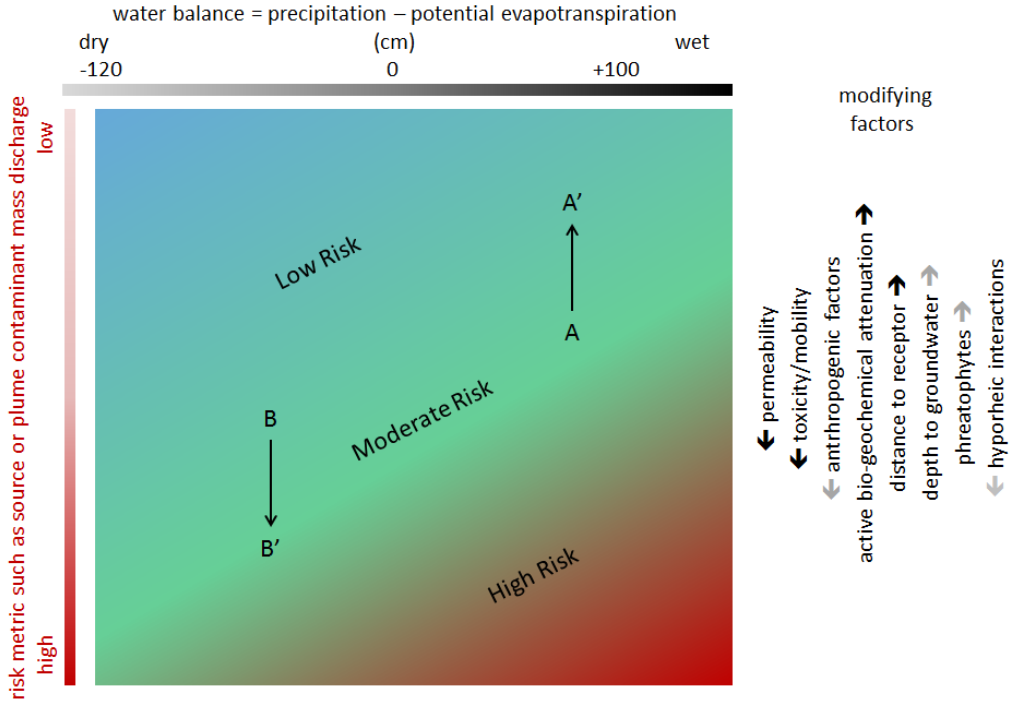


Fig. 5. Key factors determining relative risk at contaminated sites.

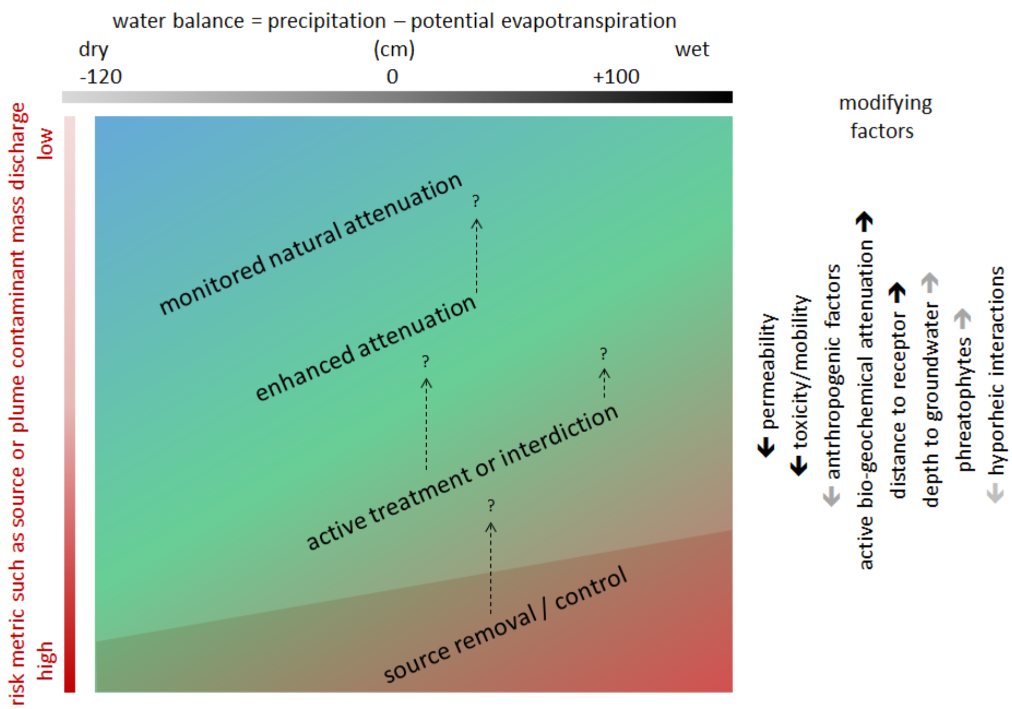


Fig. 6. Extending technology matching process to include climate and source magnitude

LM ARID SITE CASE STUDY EXAMPLES

The role of regional conditions – climate, hydrology, geology and geochemistry – is exemplified by the behavior of contaminants at the Tuba City (AZ) and Riverton (WY) Uranium Mining and Milling Sites. These LM sites in the arid to semiarid western U.S. highlights both the overarching regional influences and the significance of modifying factors in assessing cleanup strategies and defining stable-sustainable end states that are compatible with the regional and local conditions.

The behaviors of dissolved constituents at arid and semi-arid sites have been described and documented in a number of journal articles and reports [see 8]. This supporting literature identifies precipitation and accumulation of minerals such as calcite and gypsum as a dominant process. Figure 7 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 these settings, desert phreatophytes extract water and associated dissolved constituents. For areas located above the plumes at Tuba City and Riverton, 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 Tuba City Site Observational Work Plan [9] indicates that the groundwater contaminant concentrations will not adversely impact plant growth and will not accumulate to harmful concentrations in plants. Surveys of the plants present at both case study sites 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; 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.

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.

The available field data (e.g., the groundwater plume shape and size, the observed presence of evaporite minerals on escarpments and terrace transitions, etc.) confirm the overarching role of regional arid site controls on contaminants at Tuba City and Riverton. However, some of the modifying factors at these sites significantly alter how the regional influences manifest in terms of potential risks and EM/LM response. Figure 8 summarizes the composite conceptual model for Tuba City [8]. In this figure, 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. Shallow contaminated groundwater beneath the former processing site is subject 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. The Riveton situation (Figure 9) is analogous, but with a shallow water table, shorter distance to receptors, rapid later flow beneath the site (from the Wind River toward the Little Wind River), and episodic flooding.

At Tuba City the regional processes provide a relatively robust condition in which natural attenuation mechanisms limit the extent of the subsurface plume. While these regional processes are also operating at Riverton, the local modifying factors result in more potential downgradient impacts and the likelihood of contaminant remobilization by the episodic floods that flush the vadose zone. Consistent with the LM decisions and past activities at the two sites: a) the conditions at Riverton support an end state strategy that aggressively removes original source material, while b) the natural attenuation processes and conditions at Tuba City support leaving stabilized waste in place. The arid site conditions at both sites, and particularly at Tuba City, work against efficient performance of active groundwater treatment and suggest consideration of alternative natural and enhanced attenuation strategies [8] [11].

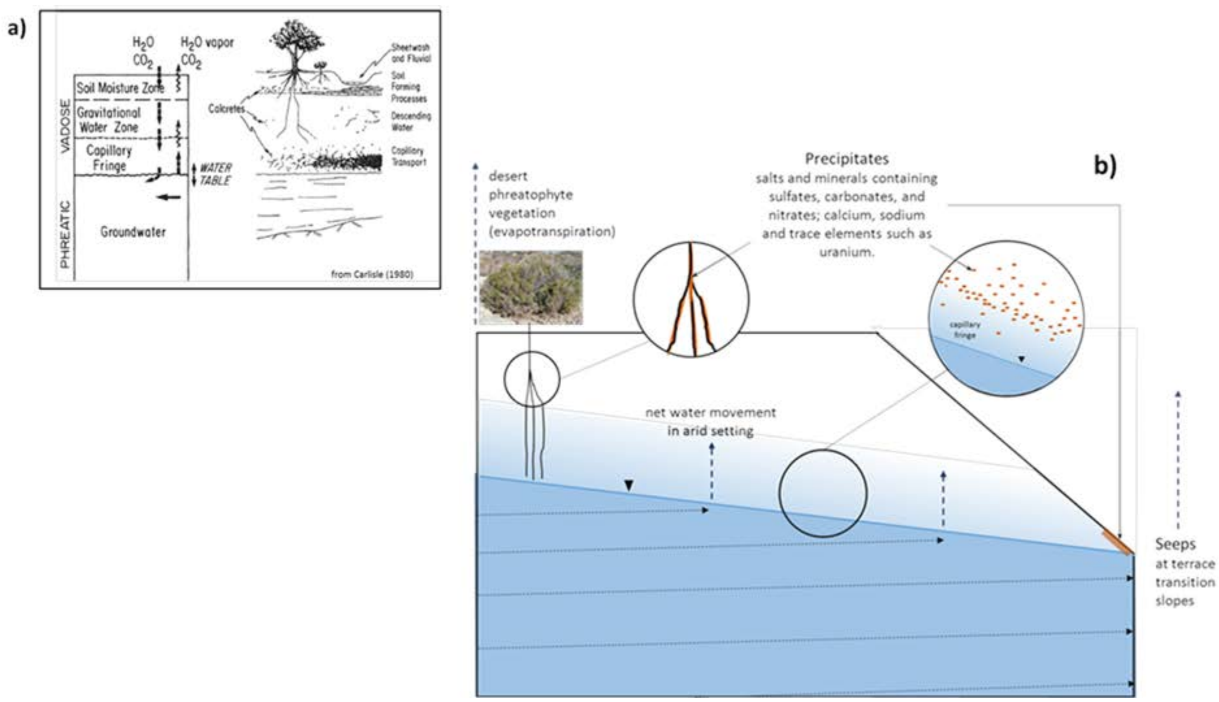


Fig. 7. Key geochemical processes associated with non-pedogenic mineral accumulations in arid and semi-arid settings – a) general concept [10] and b) annotated to highlight probable mineral accumulation zones for Tuba City and Riverton [8].

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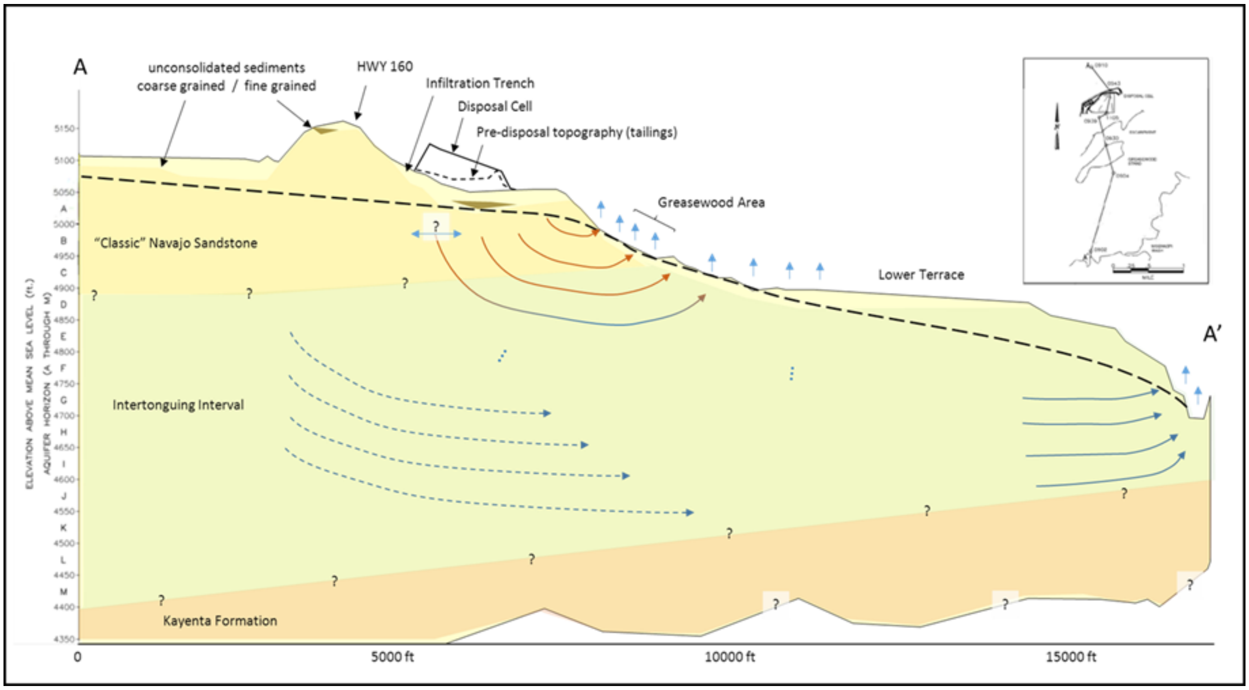

Fig. 8. Conceptual model for Tuba City showing the significance of regional arid site processes [8]

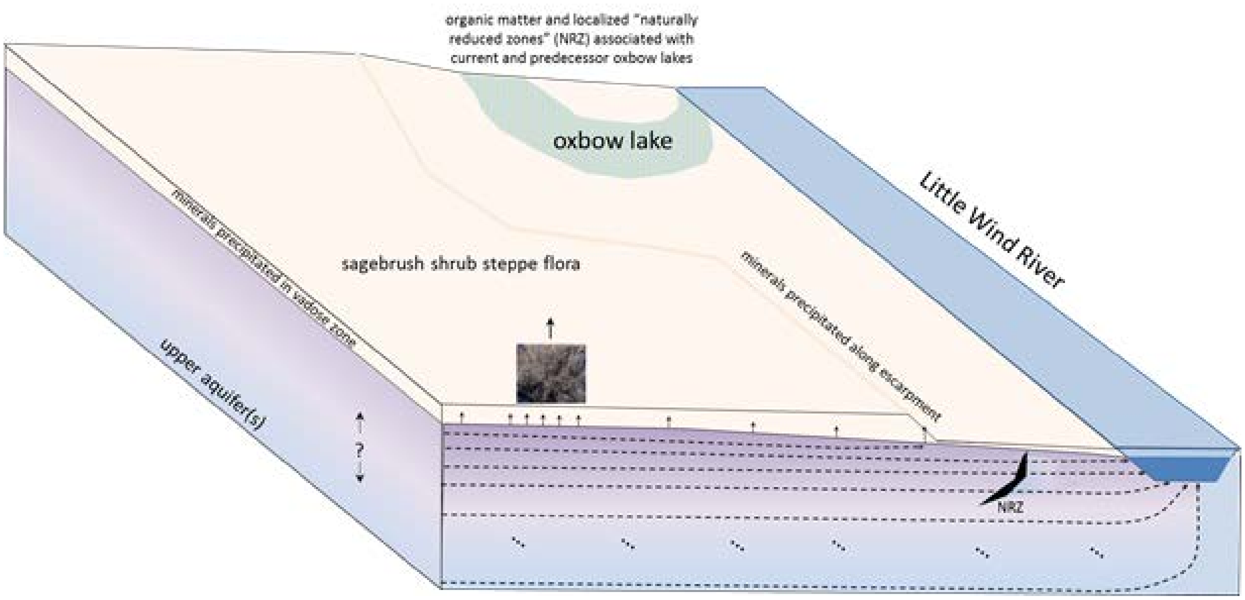

Figure 9. Midfield conceptual model for Riverton showing some of the significant modifying factors [11]

CONCLUSIONS

The contaminant distributions and plume dynamics observed at arid "western" DOE sites contrast with sites in the "eastern" US. DOE sites in the east generally have stronger hydrologic driving forces and weaker evaporative processes. In both arid and non-arid settings, long term contaminant behaviors and concomitant exposure, risk and timeframes are controlled by climate and regional conditions and by site specific modifying factors. Incorporating these conditions/factors into environmental management decisions provides new opportunities to develop more robust, sustainable and technically-defensible end states. The meta-data from the DOE technical assistance process at Tuba City and Riverton Sites, and the portfolio of DOE technical assistance sites across the country suggests that the technology matching paradigm described above is effective in helping to rapidly identify innovative environmental cleanup strategies/options that are aligned with site specific needs and conditions.

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