

Alternate Endpoints for Deep Vadose Zone Environments: Challenges, Opportunities, and Progress - 13036

Dawn M. Wellman, Mark D. Freshley, Michael J. Truex, and M. Hope Lee

¹Pacific Northwest National Laboratory, 902 Battelle Blvd, Richland, WA, 99352, USA

ABSTRACT

Current requirements for site remediation and closure are standards-based and are often overly conservative, costly, and in some cases, technically impractical to achieve. Use of risk-informed alternate endpoints provide a means to achieve remediation goals that are permitted by regulations and are protective of human health and the environment. Alternate endpoints enable establishing a path for cleanup that may include intermediate remedial milestones and transition points and/or regulatory alternatives to standards-based remediation. A framework is presented that is centered around developing and refining conceptual models in conjunction with assessing risks and potential endpoints as part of a system-based assessment that integrates site data with scientific understanding of processes that control the distribution and transport of contaminants in the subsurface and pathways to receptors. This system-based assessment and subsequent implementation of the remediation strategy with appropriate monitoring are targeted at providing a holistic approach to addressing risks to human health and the environment. This holistic approach also enables effective predictive analysis of contaminant behavior to provide defensible criteria and data for making long-term decisions. Developing and implementing an alternate endpoint-based approach for remediation and waste site closure presents a number of challenges and opportunities. Categories of these challenges include scientific and technical, regulatory, institutional, and budget and resource allocation issues. Opportunities exist for developing and implementing systems-based approaches with respect to supportive characterization, monitoring, predictive modeling, and remediation approaches.

INTRODUCTION

Remediation of subsurface contamination is a significant challenge facing the nation [1]. A substantial part of this challenge is owned by the U.S. Department of Energy Office of Environmental Management (EM). EM manages one of the largest soil and groundwater cleanup programs in the world. At the end of fiscal year 2011, EM had completed cleanup activities for 90 sites in 30 states, meaning that a remedy is in place. However, remaining cleanup activities encompass some of the most technically complex challenges ever faced and include an estimated 1.7 trillion gallons of contaminated groundwater and more than 40 million cubic meters of contaminated soil and debris at 17 sites in 11 states [2]. EM's goal is to reduce the legacy footprint to 90 percent by the end of 2015 and for the Hanford Site—the largest site within the DOE complex that contains the largest inventory of contaminated groundwater and soil—to be the only site remaining to be cleaned up by the end of 2020. The anticipated cost to complete soil and groundwater remediation across the DOE complex ranges from \$17.3 billion to \$20.9 billion [3].

Deep vadose zone inorganic and radionuclide contamination is a significant issue facing EM and other federal agencies. The deep vadose zone is defined as the depth of sediments below the zone of practical excavation and removal but above the water table [4]. Contamination in deep vadose zone environments is isolated from exposure by direct contact and does not pose a risk to human health and the environment from direct exposure. The primary exposure pathway is downward transport and discharge (flux) to the groundwater and downgradient receptors. Thus, limiting flux of contaminants from the vadose zone to groundwater is critical for protection of human health and the environment.

Previous efforts have reviewed the technical and policy challenges of deep vadose zone remediation [4,5]. The technical approaches for deep vadose zone remediation are based on 1) contaminant mass reduction; 2) contaminant stabilization; and/or 3) reduction of transport and the flux of contaminants to the groundwater. Each of these methods leaves residual contaminant mass in the vadose zone. Therein lies the integrated technical and policy challenge for remediation of deep vadose zone environments: determining acceptable levels of residual deep vadose zone contamination for which the mass flux of contamination to groundwater is low enough to meet regulatory goals that are protective of human health and the environment [4]. The approach described herein builds on these concepts developed for groundwater plumes [6] and presents a framework to achieve risk-informed endpoints for deep vadose zone remediation.

DISCUSSION

Alternate Endpoints

The following definitions are used in this paper. An **end state** is a standards-based cleanup objective associated with closure of a waste site and/or long-term management that is permitted by regulation and is protective of human health and the environment. It is the final product of a remediation or management scenario. A familiar example of an end state is a condition where contaminants at a site are at or below the maximum concentration limits (MCL) established by regulation for contaminants in drinking water. An **alternate endpoint** is a risk-informed remediation goal permitted by regulations that is protective of human health and the environment. The concept of an **alternate endpoint** enables establishing a path for cleanup that may include intermediate remedial milestones and transition points and/or regulatory alternatives to standards-based remediation. Alternate endpoints can be used to determine technology development needs as described in [7] for management of waste tanks at the Hanford Site.

Current end states and requirements for site remediation and closure are standards-based. This approach leads to remediation goals that often are overly conservative, costly—and in some cases—technically impractical to achieve. There is growing recognition that there are a number of complex sites where active remedies will not be successful and alternate endpoints will be required [8]. There are multiple currently acceptable alternate endpoints that apply to groundwater [6] including attenuation approaches, adaptive site management, groundwater reclassification, alternate concentrations, and Applicable or Relevant and Appropriate Requirements (ARAR) waivers. Attenuation approaches include monitored natural attenuation (MNA) and enhanced attenuation (EA) that are implemented based on robust conceptual models with adequate site characterization, long-term monitoring, and limited active remedies. Attenuation is important to consider in most remedial strategies for distal portions of a plume or remnant contaminants from an active remedy. EA involves either source reduction or actions to enhance the attenuation rate to stabilize or shrink a contaminant plume. Adaptive site management involves an iterative approach, with actions implemented over time in response to site conditions. Groundwater reclassification involves regulatory changes so that groundwater at a site is no longer designated as drinking water. Alternate concentration limits replace or modify cleanup standards; for example, where contaminated groundwater discharges to surface water. ARAR waivers are used where compliance with a regulatory limit is technically impractical. Of these approaches, attenuation methods, adaptive site management, and ARAR waivers potentially apply to deep vadose zone contamination.

The process of defining and implementing alternate endpoints is risk informed. This decision process is based on analysis of the potential for a contaminant to cause immediate and/or long-term harm to a receptor resulting from exposure and the likelihood of this occurrence. Comparable to end states, alternate endpoints must be scientifically and technically defensible and based on systematic, objective understanding of the contamination issue and impact of proposed solutions to provide justification for the site remediation decisions.

Alternate Endpoints Framework

Fig. 1 presents a systems-based framework for implementing remediation at a site where an alternate endpoint is expected. The framework provides a means to define the nature and extent of the problem to determine which risks are most critical and establish alternative endpoint cleanup decisions. The framework is based on a strong mass flux-based conceptual model in conjunction with assessing risks and potential endpoints as part of a system-based assessment that integrates site data with scientific understanding of processes that control the distribution and transport of contaminants in the subsurface and pathways to receptors. This system-based assessment and subsequent implementation of the remediation strategy with appropriate monitoring are targeted at providing a holistic approach to addressing risks to human health and the environment. Goals of the framework are to provide the following:

- Deeper insight into the *important* remedial/transport processes
- Platform for integrating new knowledge into flux-based conceptual site models that are significantly more predictive to provide defensible criteria/data for making long-term decisions
- Holistic assessment of risk to human health and the environment
- Flexible approach for application to a range of sites, from simple to complex
- Appropriate path for transitioning to long-term monitoring and stewardship.

Implementation of this framework and alternative approaches therein requires cooperative involvement from technical experts, site owners, federal and state regulators, and stakeholders.

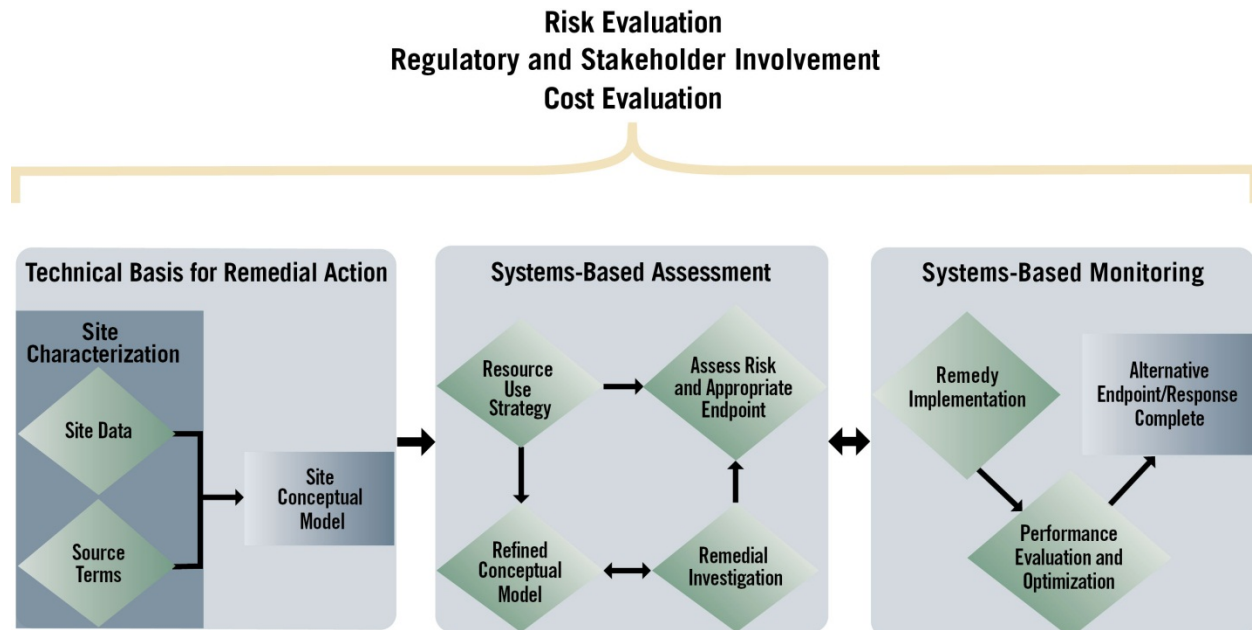


Fig. 1. Systems-based framework for endpoints evaluation

Alternate Endpoints for Vadose Zone Environments

Contaminants in the vadose zone often are long-term sources of groundwater contamination. Remediation decisions for the vadose zone typically are based on projected impacts to groundwater, emphasizing the need for a flux-based evaluation approach [4]. Significant natural attenuation processes control vadose zone contaminant transport and discharge to groundwater. Attenuation processes include both hydrobiogeochemical processes that serve to retain contaminants within porous media and physical processes that mitigate the rate of water flux. The physical processes controlling fluid flow in the vadose zone are quite different and generally have a more significant attenuation impact on contaminant transport relative to groundwater systems. Truex and Carroll [9] present a remedy evaluation (alternate endpoint) framework that is based on an adaptation of the established EPA MNA evaluation approach and a mass flux-based conceptual model approach focused on identifying and quantifying features and processes that control contaminant flux through the vadose zone. The groundwater-based MNA evaluation framework was extended to natural attenuation for contaminants within the vadose zone and consideration of alternate endpoints.

The EPA technical protocol for MNA of inorganic contaminants in groundwater [10] is based on a tiered approach (Table I); the first three tiers constitute progressive evaluation of the subsurface contamination and site conceptual model and the fourth tier is focused on implementation.

Table I. Summary of Assessment Criteria Typically Used for Groundwater Contamination Sites and Proposed for the Vadose Zone [9].

Tier	Objective	Assessment
I	Demonstrate active contaminant removal	Is the plume expanding, static, or contracting?
II	Determine mechanism and rate of attenuation	Is the MNA rate sufficient for attaining cleanup in a reasonable time frame?
III	Determine system capacity and stability of attenuation	Is the MNA capacity sufficient and sustainable to attenuate contaminant mass to below regulatory objectives?
IV	Design performance monitoring program and identify alternative remedy	Can monitoring be implemented to verify performance and identify condition changes that may lead to failure?

As with groundwater contamination, MNA may be sufficient as a sole vadose zone remedy or can be quantified for inclusion as part of a remedy along with other measures. Along with providing information to define a site conceptual model and support a baseline risk assessment, evaluating whether MNA is sufficient as a remedy is a key initial step in evaluating the need for and scale of other remedial measures. Similarly to groundwater assessments, examining alternative endpoints for the vadose zone requires making projections of future contaminant behavior (the first three tiers of the EPA protocol) and providing a basis for monitoring design (the final tier). Truex and Carroll [9] modified the EPA protocol for evaluating vadose zone remediation needs based on a scientific understanding of contaminant mass flux and discharge to groundwater (Fig. 2).

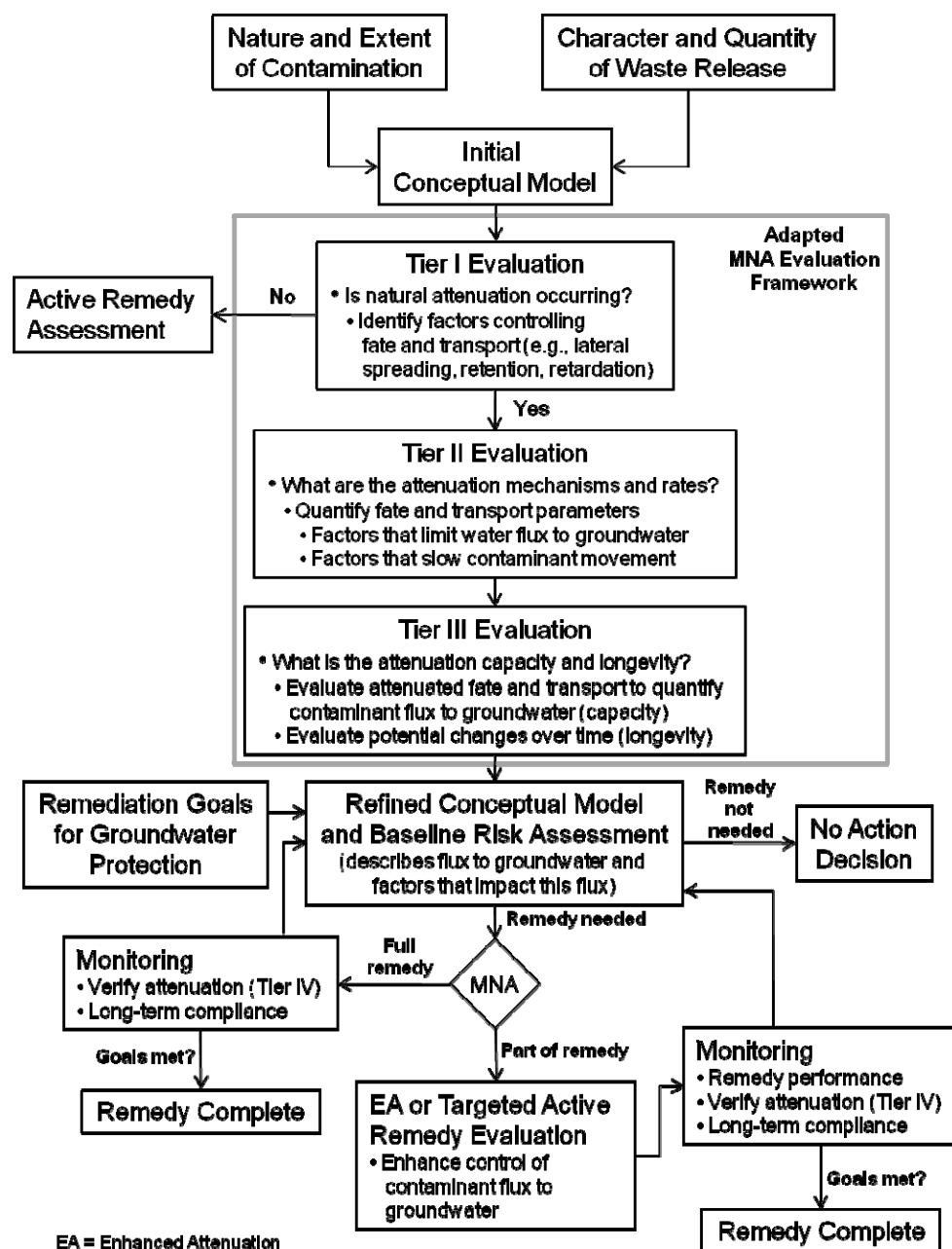


Fig. 2. Vadose Zone Remedy Evaluation and Implementation Flow Chart [9].

The approach to vadose zone remediation and alternate endpoints shown in Fig. 2 is based on an overall conceptual model framework of contaminant movement in the vadose zone. In this conceptual model, waste material released to the vadose zone is a perturbation of pre-existing conditions and results in an increase of contaminants and/or moisture in the vadose zone. Because of the nature of moisture movement in the vadose zone, the post-contamination moisture conditions will return to pre-existing conditions, which are governed by the recharge rate and unsaturated flow processes near the ground surface. Under long-term equilibrium conditions, the rate of water movement is effectively constant throughout the vadose zone and equal to the average recharge rate. Movement of inorganic and

radionuclide contaminants as solutes may be slower than vadose zone water movement because of processes such as sorption, solubility, degradation and decay, and dispersion. Contaminant discharge to groundwater over time is controlled by the combination of the moisture flux and biogeochemical processes.

Tier I in the EPA protocol evaluation for groundwater plumes is based on evidence that natural attenuation is occurring. Evidence that a plume is shrinking or calculations that show a plume is not posing a risk to receptors are examples of natural attenuation. For the vadose zone, there is no direct exposure risk; thus, movement toward underlying groundwater is the primary concern. Natural attenuation can limit movement through the vadose zone and is a mechanism for reducing flux to groundwater. In this case, a Tier I assessment consists of demonstrating that the rate of contaminant movement toward the groundwater is decreased by factors that limit vertical flux of contaminants relative to water flow. The assessment can make use of direct evidence or a transport analysis of contaminant movement, data from sediment cores that shows partitioning of contaminants from pore water to sediment, and monitoring data such as subsurface geophysics that demonstrate declining contaminant flux rates.

In Tier II, the mechanisms and rates of attenuation are evaluated and quantified. For the vadose zone, there are two major contributors to attenuation of contaminant flux: 1) hydrologic factors that limit water and contaminant flux to the groundwater; and 2) biogeochemical processes that slow contaminant movement relative to water movement. Hydrologic factors include heterogeneities that cause lateral moisture movement, preferential pathways, and nonlinear moisture retention characteristics. Contaminant dilution, dispersion, and degradation impact attenuation in the vadose zone. Biogeochemical phenomena include sorption, solubility, and degradation or decay. The Tier II approach is to quantify the processes identified above to estimate or measure the rate of flux and vadose zone mass balance over time.

Tier III of the EPA protocol consists of evaluating two elements of natural attenuation that are important in defining alternate endpoints and demonstrating that remedial strategies will meet remediation goals. First, the capacity of an aquifer system to attenuate contaminants is evaluated. The subsurface (vadose zone and aquifer) system within the zone identified for treatment (e.g., before reaching a compliance location) must be able to attenuate contaminants that are present to reach targeted concentration goals. Second, the longevity of natural attenuation is assessed. This is done by demonstrating that attenuation mechanisms will be able to maintain acceptable concentrations in the groundwater over time under the range of hydraulic and geochemical conditions that are expected. For the vadose zone, demonstrating attenuation capacity and longevity points to the need to quantify the contaminant flux to groundwater over time and demonstrate this flux will be acceptable over the long term. Determination of the “acceptable” vadose zone flux must be linked to meeting endpoint goals for the site.

Tier IV of the EPA protocol for groundwater plumes specifies designing a monitoring program using a network of wells to 1) provide adequate areal and vertical coverage to verify the groundwater plume remains static or shrinks; and 2) monitor groundwater chemistry necessary to ensure that attenuation mechanisms are being sustained. The approach for groundwater is also relevant to the vadose zone in that it points to the need for verifying attenuation capacity and longevity, although the specific monitoring approach for groundwater using monitoring wells and groundwater chemistry assessment is not applicable in the vadose zone. In the vadose zone, multiple elements identified in the conceptual site model of contaminant fate and transport are monitored as “lines of evidence” in a systems-based conceptual model approach [11]. Lines of evidence can include items such as the following:

- Recharge rate and changes over time
- Moisture/pressure distribution monitoring as an indicator of moisture flux or retention

- Contaminant distribution monitoring over time using methods such as geophysics
- Monitoring of geochemical conditions related to stability of sorption, solubility, and/or degradation phenomena
- Monitoring of changes in groundwater conditions or active remedy impacts relevant to vadose zone attenuation.

Transitioning to long-term monitoring configurations that build on near-term lines-of-evidence and use diagnostic elements of the overall environmental system and/or threshold monitoring approaches can be used to demonstrate compliance in a streamlined approach that is sustainable over long time frames.

Alternate Endpoint Evaluation Examples

Two examples from the Hanford Site with respect to application of the framework outlined by Truex and Carroll [9] (2012) are described: 1) for Tc-99 at the BC Cribs and Trenches, and 2) evaluation of plutonium and americium at multiple waste sites. These two examples are at different stages of the evaluation process.

Tc-99 at BC Cribs and Trenches. Technetium contamination at the BC Cribs and Trenches site is being evaluated as part of the Deep Vadose Zone Operable Unit. Data, including analysis of borehole samples (Fig. 3) and geophysical electrical resistance surveys (Fig. 4), show that lateral spreading of contamination and associated waste water has occurred. The lateral spreading diminishes moisture conditions as an attenuation mechanism, and an assessment of moisture conditions and porous media properties can be used to assess current fluxes in comparison with those expected under long-term recharge-driven conditions. The BC Cribs and Trenches site has been the focus of efforts associated with deep vadose zone investigations (e.g., [12,13,14,15]). These efforts have included simulation of water and Tc-99 flux to groundwater [15]. These simulations predict that most Tc-99 will reach the water table under recharge-driven conditions (Fig. 5) at fluxes that cause the groundwater concentrations to exceed the drinking water standard. Additional simulations and investigation of Tc-99 transport in unsaturated sediments [16] are underway to refine the understanding of potential future groundwater impact at the BC Cribs and Trenches site.

The remedy evaluation framework evaluation is not likely to support the feasibility of using MNA as the sole remedy. However, the refined conceptual model and evaluation of groundwater impacts do support decisions regarding other active remedies. While it is recommended the structured remedy evaluation framework be applied to this site, these initial efforts demonstrate the type of information that could feed into remediation decisions. For example, current information suggests that efforts to mitigate the flux of Tc-99 to the water table would be needed to meet the drinking water standard. Surface barriers and soil desiccation have been examined as a potential mitigation measure for this site (e.g., [12,13,14,15]). This case study is an example of a Tier 1 investigation.

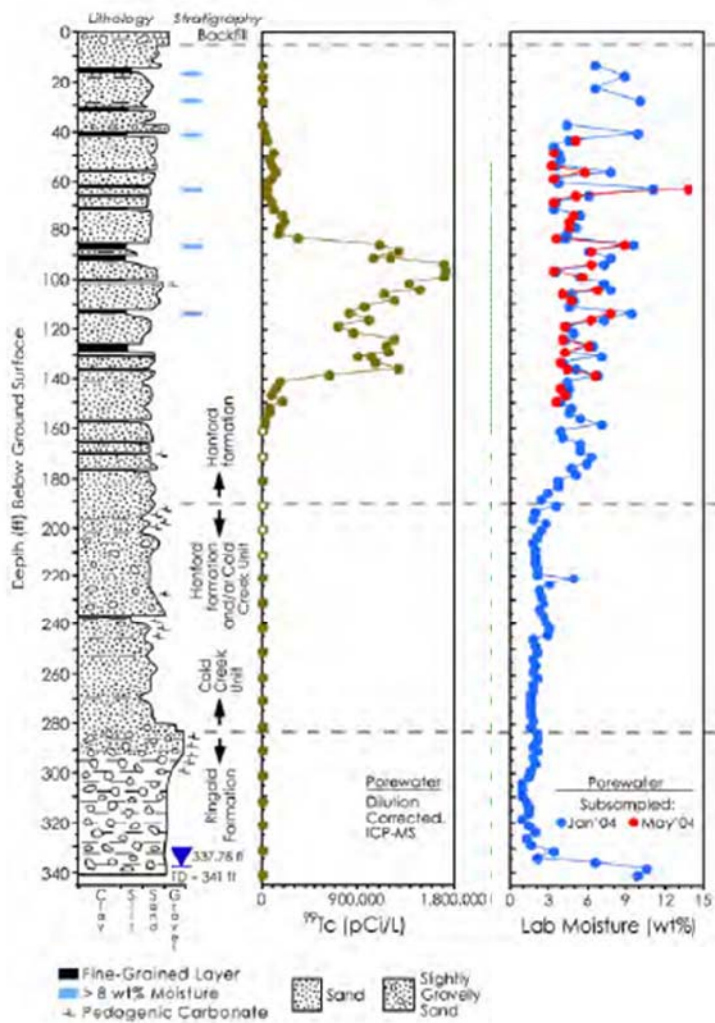


Fig. 3. Laboratory-sample-derived moisture and Tc-99 profiles for a borehole located adjacent to the B-26 trench at the BC Cribs and Trenches site [17]. Tc-99 in water was released to the subsurface in a short duration discharge into the trench and water and Tc-99 pulse have dispersed during transport in the vadose zone.

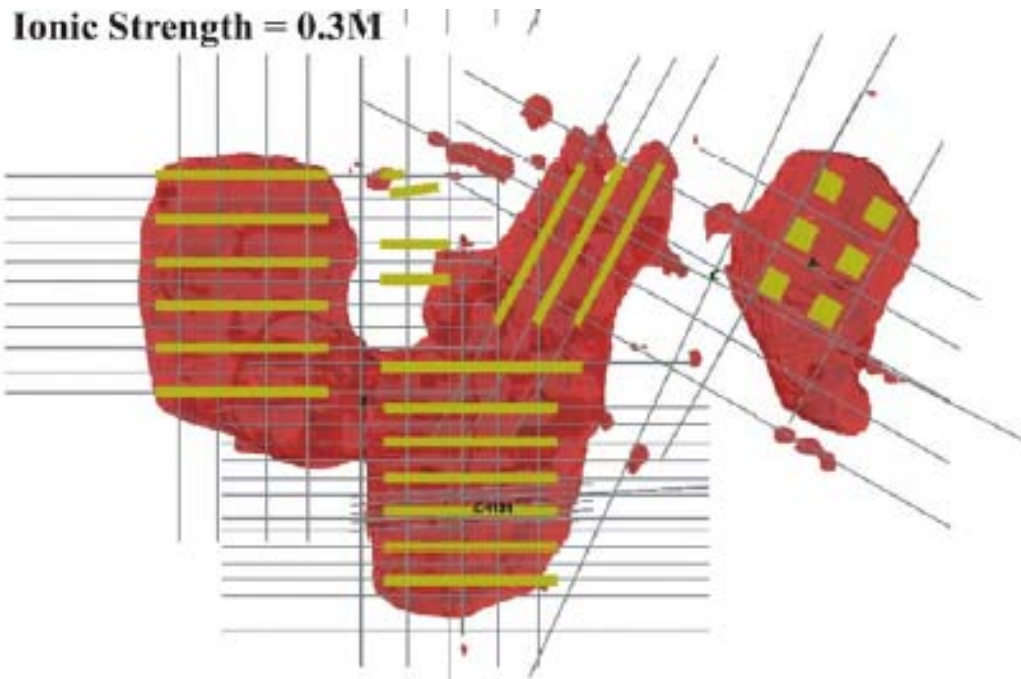


Fig. 4. Estimated lateral distribution of ionic strength (used as a surrogate for contamination) in the vadose zone at the BC Cribs and Trenches (adapted from [17]). The total area of the figure is approximately 35 acres.

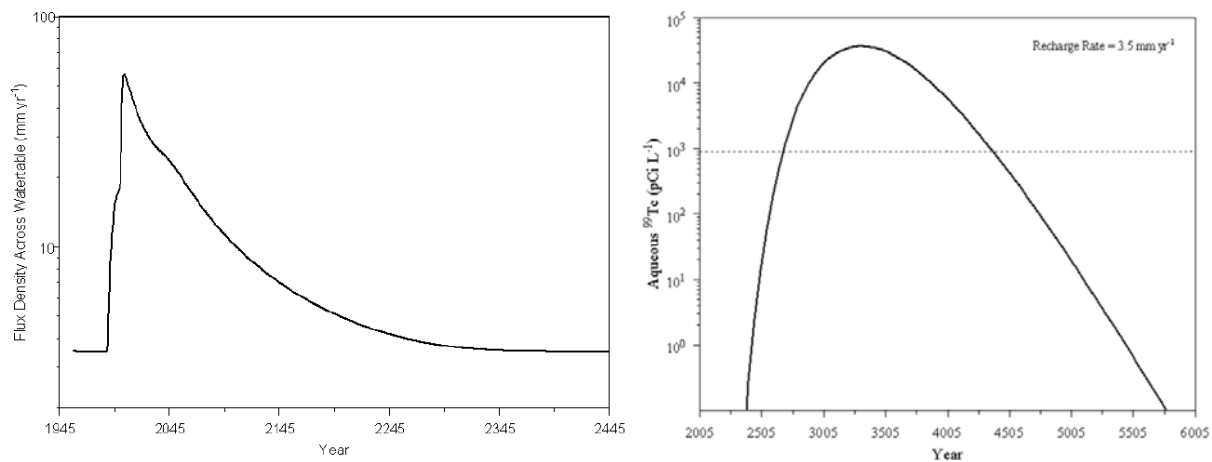


Fig. 5. Simulated water flux across the water table (left) and predicted Tc-99 concentrations over time in the groundwater (right) beneath the trenches portion of the BC Cribs and Trenches site [15]. The dashed line in the right plot is the Tc-99 drinking water standard.

Plutonium and americium in Hanford Site wastes. The state of knowledge for plutonium and americium at the Hanford Site was reviewed and summarized [18] to provide a basis for remediation decisions. This assessment is an example of information that can be compiled to support evaluation of geochemical attenuation and transport processes in the vadose zone. Spent fuel was reprocessed at the Hanford Site

from 1944 through 1989 for production of plutonium for nuclear weapons [19]. Over the lifetime of the Hanford Site facilities, 96,900 metric tons of uranium in the form of spent fuel was reprocessed to recover 67.4 metric tons of plutonium (4.2×10^6 Ci) [19] (Gephart 2010). The reprocessing operations produced waste streams that were disposed of in facilities ranging from single- and double-shell tanks to trenches, cribs, and ponds. It is estimated that 11,800 Ci (189 kg) of Pu-239; 2,900 Ci (12.6 kg) of Pu-240; 37,500 Ci (0.34 kg) of Pu-241; 28,700 Ci (9.0 kg) Am-241; and 55 Ci (78 kg) Np-237 were disposed across the Hanford Site [20]. The vast majority of transuranic contaminants disposed to the vadose zone at the Hanford Site (10,200 Ci [86%] of Pu-239; 2,560 Ci [88%] of Pu-240; 33,100 Ci [88%] of Pu-241; 27,900 Ci [97%] of Am-241; and 41.8 Ci [78%] of Np-237) were disposed at sites adjacent to the Plutonium Finishing Plant.

Waste sites that received plutonium and americium at Hanford can be classified into two major categories based primarily upon the type of wastes that were received [21] (Cantrell and Riley 2008). The two waste categories are low-salt near-neutral wastes and acidic high-salt waste with which organic complexants were codisposed [22]. At some waste sites where plutonium- and americium-rich wastes were disposed, measureable concentrations of both plutonium and americium have reached considerable depths within the vadose zone (in excess of 40 m below ground surface). In general, much higher concentrations of plutonium and americium were transported deep into the vadose zone at sites that received acidic high-salt waste co-disposed with organic solvents.

The geochemistry of plutonium is impacted by the oxidation state [18]. The oxidation state can significantly impact complexation with dissolved ligands, solubility, and sorption. As a result, the mobility of americium can be different than that of plutonium and these differences can be significantly impacted by local redox chemistry. The precipitation of plutonium and americium in Hanford Site sediments is linked to the concentrations in the disposed solutions, oxidation state (plutonium), the acidity or pH of the discharge solutions, presence of ligands (such as PO₄) that can form solid phases, and changes in chemistry that can occur when waste solutions contact subsurface sediments. In this regard, acidic wastes discharged to some of the cribs and trenches are examples of disposal sites where precipitation of plutonium—and possibly americium—solids would occur as the acid waste solutions contact the underlying sediments. When plutonium and americium concentrations are below the solubility limits of applicable solubility controlling phases, adsorption to mineral surfaces can be important. Adsorption is particularly important in the far field region of a waste site. The degree of adsorption can vary considerably and is dependent upon oxidation state (for plutonium), pH, complexation with dissolved ligands, and mineral surface type and surface area. Some of the more important minerals that can adsorb plutonium and americium include various metal oxides (iron oxides, manganese oxides, aluminum oxides), clay minerals, calcite, and silica. All of these minerals occur in Hanford Site sediments.

The significance of colloidal transport of plutonium and americium within the Hanford Site vadose zone where plutonium and americium wastes were disposed remains unclear. Based on data from a single study, it appears that for typical far-field conditions, colloidal transport through Hanford Site groundwater is not an important transport mechanism. The situation for waste sites significantly impacted by plutonium processing wastes is much less certain. Several lines of evidence suggest that colloidal transport may have played a role in the migration of plutonium through the vadose zone beneath waste sites during periods of active disposal. These lines of evidence include the fact that acidic wastes containing plutonium and americium can produce colloidal particles during acid-induced weathering of sedimentary minerals in the vadose zone.

The study of Cantrell and Felmy [18] is an example of an investigation in Tier II where the attenuation mechanisms are identified and are in the process of being evaluated. Cantrell and Felmy [18] also

identified two issues that need to be resolved: 1) the mechanism responsible for migration of plutonium and americium into the deep subsurface at disposal sites in the Hanford Site 200 West Area; and 2) whether plutonium and americium present at Hanford Site disposal sites can be remobilized and transported through the vadose zone. To address these issues, three research areas were identified:

1. Determine the transformations of Hanford Site sediments in response to changes in waste/groundwater composition. Significant mineralogical transformations can occur when acidic wastes contact Hanford Site sediments. These transformations can impact the solubility and adsorption of plutonium and americium and possibly result in the generation of colloids that could facilitate plutonium and americium migration.
2. Assess the impact of changes in waste and groundwater chemistry on the potential for plutonium and americium solubilization or colloid formation. Changes in waste or groundwater chemistry can greatly impact the chemical form or speciation of plutonium and americium. Knowledge of plutonium and americium speciation is important for evaluating the potential for solubilization, adsorption, or remobilization of adsorbed complexes, colloid formation, and colloid interactions with sedimentary minerals.
3. Establish the role of organic complexants and/or nonaqueous solvents in the transport of plutonium and americium in the deep subsurface. Plutonium has been found to be associated with organics and the presence of nonaqueous solvents, at least in certain sediment samples. It is important to establish the role of nonaqueous solvent in past movement of plutonium and its potential role in future mobility.

CONCLUSIONS

The current approach of traditional active engineered remediation works at “simple” sites to achieve remediation and closure goals, but has proven ineffective at complex sites where regulatory milestones are often missed and little has been done to diminish actual risk. The remaining challenges facing EM are complex and require holistic systems-based approaches that integrate research and understanding between technical areas, take into account the entire ecosystem, and advance from standards-based remediation to managing actual risks to ecological and human receptors.

Challenges and Opportunities

Alternate endpoints and the associated implementation framework provide an improved mechanism for EM to address challenges, risks, and remediation costs of contamination at complex waste sites in place of more traditional approaches of contaminant removal and disposal. Developing and implementing an alternate endpoint-based approach for remediation and waste site closure presents a number of challenges and opportunities (Fig. 6). Categories of these challenges include scientific and technical, regulatory, institutional, and budget and resource allocation issues. Opportunities exist for developing and implementing systems-based approaches for determining remediation approaches and enabling implementation of alternate endpoints. Characterization, monitoring, predictive modeling, and risk assessments are critical components of the implementation framework. Technology development and evaluation, as well as attenuation-based approaches, are foundational elements supporting the ability to achieve remediation goals and close waste sites using alternate endpoints. Communication with regulators, Tribal Nations, and stakeholders is critical for implementation of alternate endpoint approaches, particularly with respect to risk assessments and choices for prioritizing resources. The transition of sites to long-term monitoring and stewardship is also a key component of an alternate endpoint approach. While some development and policy efforts are needed to enable broad implementation of alternate endpoints for EM, the alternate endpoint approach has the potential to

expedite cleanup and reduce cost through understanding what should be accomplished through cleanup efforts, what endpoint(s) or condition(s) constitute progress or completion of Hanford Site cleanup, and schedule commitments with a defensible and credible technical scope of work, including clear requirements to achieve risk-informed endpoints.

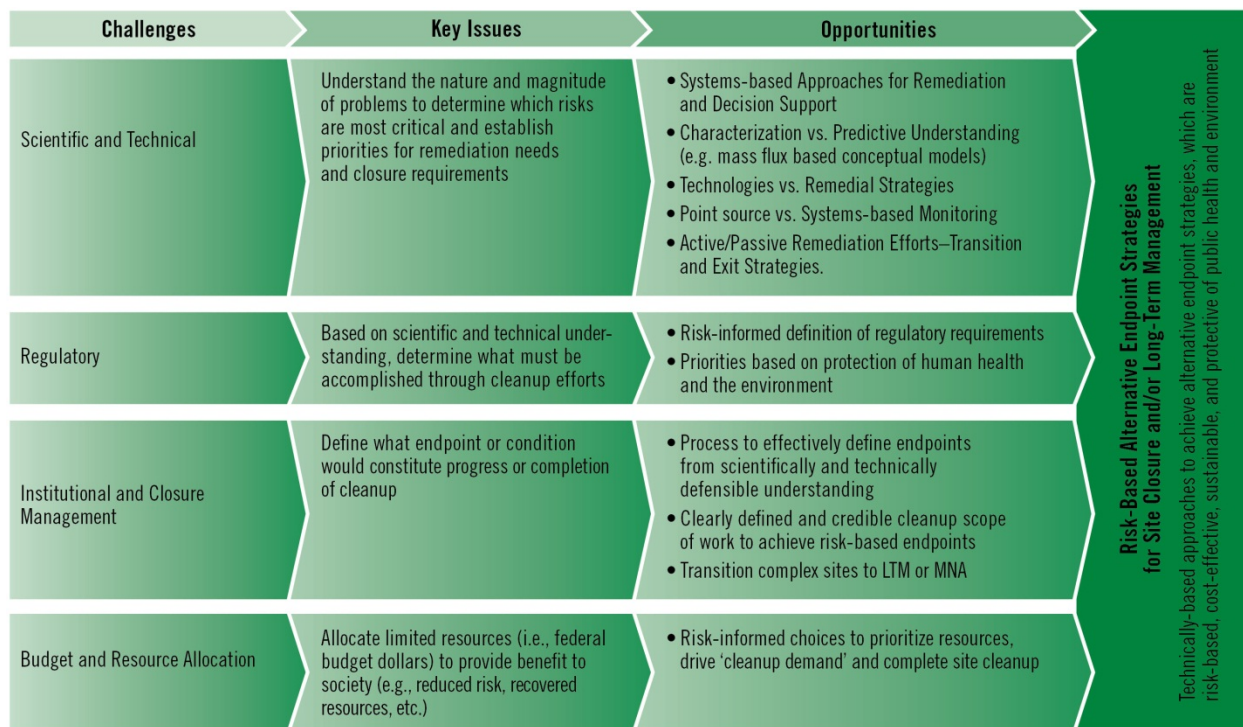


Fig. 6. Challenges, issues, and opportunities associated with risk-based alternate endpoint strategy.

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