Deep Vadose Zone Applied Field Research Center: Transformational Technology Development for Environmental Remediation – 11074

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ABSTRACT

The U.S Department of Energy –Environmental Management (DOE-EM) Office of Groundwater and Soil Remediation and DOE Richland Operations (DOE-RL), in collaboration with the Hanford Site and the Pacific Northwest National Laboratory, have established the Deep Vadose Zone Applied Field Research Center (DVZ-AFRC). The DVZ-AFRC leverages DOE investments in basic science from the DOE Office of Science, applied research from DOE-EM Office of Technology Innovation and Development, and remediation application by the site operation (e.g., site contractors [CH2M HILL Plateau Remediation Contractor and Washington River Protection Solutions], DOE-EM, -RL, and Office of River Protection) in a collaborative effort to address the complex region of the deep vadose zone. Although the aim, goal, motivation, and contractual obligation of each organization is different, the integration of these activities into the framework of the DVZ-AFRC focuses the resources and creativity of many to provide the knowledge and capabilities required to create viable alternative remedial strategies to current baseline approaches for persistent contaminants and deep vadose zone contamination challenges. This cooperative strategy removes stovepipes, prevents duplication of efforts, maximizes resources, and facilitates development of the scientific foundation needed to make sound and defensible remedial decisions that will successfully meet the targeted cleanup goals for one of DOE EM's most intractable problems, in a manner that is acceptable to regulators.

INTRODUCTION[1]

Many subsurface vadose zone environments within the DOE complex consist of complex stratified layers of unconsolidated and water-unsaturated sediments that are in many places contaminated with radionuclides, metals, organics, and, in some cases, complex mixtures. This contamination originated from a number of sources, including intentional disposal to the ground surface through the use of cribs, retention basins, and trenches, and from unintended tank waste releases. In many cases, minimal historical information exists regarding the magnitude, timing, and content of contaminant releases, thus necessitating estimation of the source terms.

Inorganic and radionuclide contamination in the deep vadose zone is isolated from the surface environment, such that direct contact is not a factor in its risk to human health and the environment. Rather, the deep vadose zone serves as both a present and potential future source of groundwater contamination. Movement of contamination from the deep vadose zone to the groundwater creates the potential for exposure and risk to receptors through contaminant uptake from water withdrawn from wells or discharge to water resources. Thus, remediation solutions for the deep vadose zone target protection of the groundwater, specifically by preventing contaminants from exceeding established concentration limits once in the groundwater. The magnitude of the contaminant discharge (mass per time) from the vadose zone to the groundwater must be maintained low enough to meet the groundwater concentration goals by natural attenuation (e.g., adsorption processes or radioactive decay) or through remedial actions (e.g., contaminant mass or mobility reduction).

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The deep vadose zone poses unique problems for characterization, monitoring, and remediation. The heterogeneous nature of the deep vadose zone makes detailed characterization of the distribution and extent of contamination very difficult; thorough characterization using traditional sampling and analysis is not cost-effective. Functionally, the methods for addressing subsurface contamination must remove contamination and/or reduce transport of contaminants through the vadose zone. However, because pore spaces are unsaturated, conventional remediation technologies such as pump-and-treat are ineffective. In addition, much of the contamination is too deep for conventional surface excavation and below the depth at which a surface infiltration barrier would sufficiently retard contaminant migration and protect groundwater. These issues and others combine to make the deep vadose zone one of the most challenging environmental remediation problems in the DOE complex today. Development of *in situ* remediation technologies or defensible technical data and justification for relying on natural attenuation may be the only way to remediate contamination in the deep vadose zone. Minimizing the flux of contaminants from the vadose zone to the groundwater with *in situ* techniques may be the only viable path to long-term stewardship of sites contaminated with metals and long-lived radionuclides other than physical removal techniques, which are costly and often simply move the problem from one location to another.

Given the sheer magnitude and cost associated with addressing this first-of-a-kind technical challenge, no single organization or entity has the financial and/or technical resources available to solve the complex issues facing DOE in the deep vadose zone. For DOE to successfully address remaining cleanup problems, it will require 1) partnering and leveraging with other relevant organizations and 2) integrating basic science and “needs-driven” applied research activities with DOE cleanup operations to facilitate the transition of scientific results into applied solutions.

DEEP VADOSE ZONE – APPLIED FIELD RESEARCH CENTER

DOE recognizes that finding solutions to deep vadose zone challenges is unlikely to have immediate solutions and is a long-term problem. DOE plans to use a “***defense-in-depth***” remediation approach that includes the following components (Fig. 1):

- Implementation of ***surface remedies*** to mitigate the potential effects of deep vadose zone contamination.
- Inclusion of an ***integrated groundwater and vadose-zone monitoring system*** designed to provide an early warning of significant contaminant movement or impact to groundwater.
- Implementation of ***groundwater treatment systems*** that can be expanded or redesigned to address emerging plumes, when necessary.
- Continued investment in ***treatability tests*** to evaluate innovative approaches to remediate deep vadose zone contamination.
- Sustained investment in ***advanced science and technology solutions*** to address deep vadose zone challenges including characterization, prediction, remediation, and monitoring.
- Periodically revisiting the ***effectiveness of remedies*** and possible changes in environmental conditions from natural or anthropomorphic sources to maintain effective and efficient remediation.

This defense-in-depth approach requires a sufficient technical basis to understand, predict, control and monitor contaminant movement. An integrated, science-based and problem-driven research effort is needed to fill existing gaps in knowledge and capabilities.

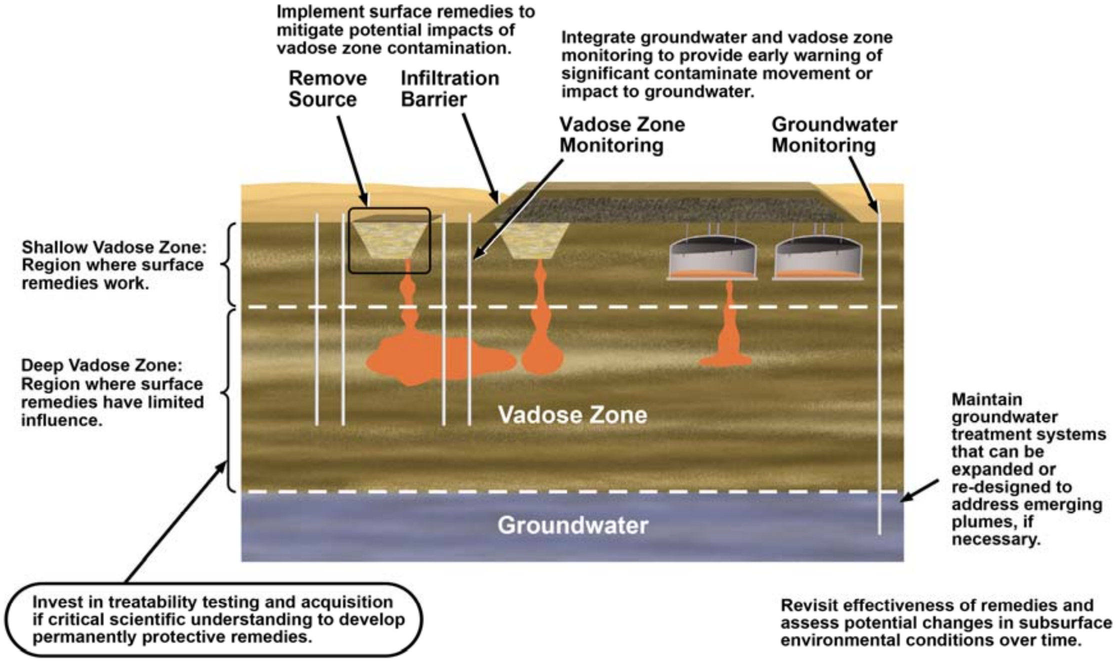


Fig. 1. Elements of the defense-in-depth approach to groundwater protection

A sustained effort is needed to seek and develop new transformational approaches to this challenging problem. The desired programmatic outcomes needed to support DOE's defense-in-depth approach include:

- Identify key regulatory and cleanup objectives that successful remedies will need to meet.
- Identify and prioritize critical gaps in knowledge and capability.
- Develop the technical and scientific bases to address deep vadose zone contamination where existing capabilities and knowledge fall short.
- Integrate basic research with applied science through field-scale engineering activities to test and develop remediation approaches.
- Focus research upon those problems or opportunities that offer the greatest potential payoff.
- Use a portfolio of restoration approaches tailored to provide the most effective and efficient strategies to achieve cleanup goals.
- Identify opportunities to enhance data collection and analysis by building a collaborative relationship with the subsurface remediation program.
- Link research and innovative treatability activities to the subsurface remediation program, and leverage knowledge, capabilities, and funding sources across multiple subsurface cleanup programs.
- Build validated conceptual and predictive models capable of simulating the subsurface environment and performance of remedial actions.

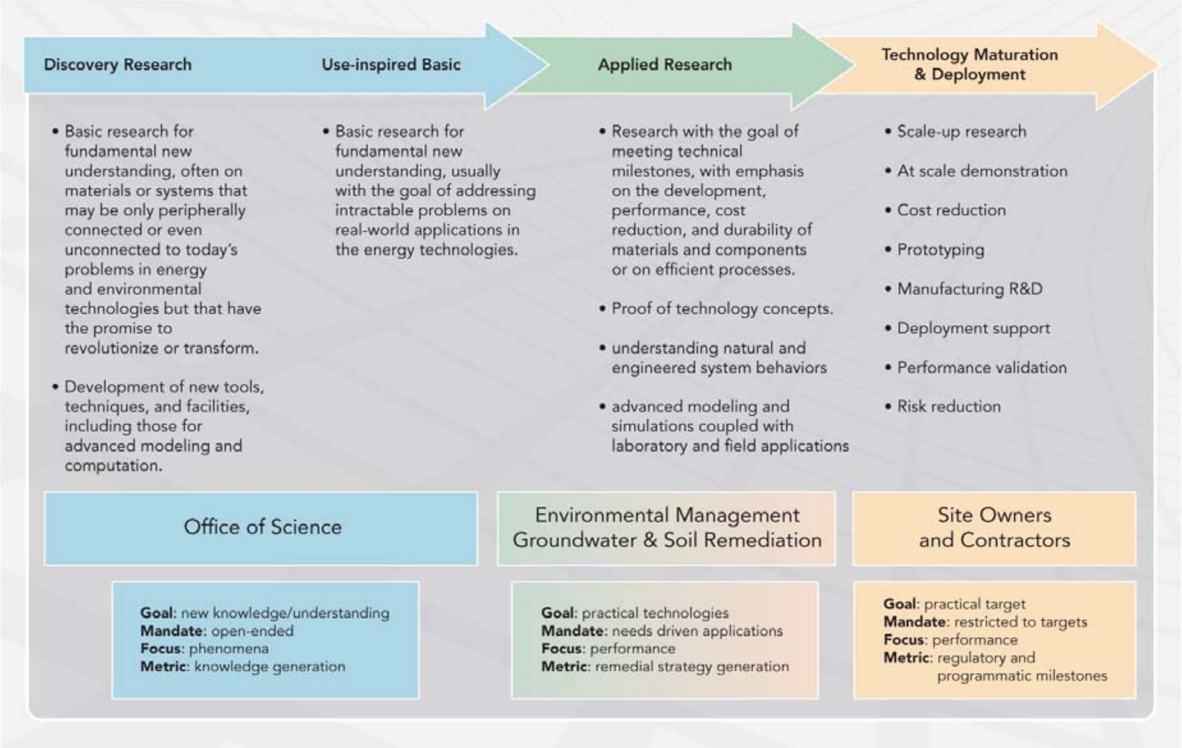
In recognition of the need to find a broader set of effective methods for characterizing, remediating and monitoring the deep vadose zone, DOE has prepared a long-range plan that identifies scientific and technological needs and opportunities related to the deep vadose zone at Hanford [1]. This planning effort focused on developing a set of basic and applied science gaps and opportunities that will be used to guide the integrated, collaborative Deep Vadose Zone Applied Field Research Center. The overall vision for DVZ-AFRC is to provide a technical basis to quantify, predict, and monitor natural and post-remediation contaminant discharge from the vadose zone to the groundwater and to facilitate developing *in situ* solutions that limit this discharge to acceptable levels and protect water resources.

The contaminant discharge from the vadose zone to the groundwater is affected by processes that occur in the vadose zone and affect contaminant transport. Investments from DOE Office of Science (SC) are being applied to understanding these processes and their relation to the biogeochemical and hydrologic conditions in the vadose zone. Site contractor resources are being applied to characterize the nature and extent of contaminants in the vadose zone, conduct treatability tests to quantify how technologies change the site and contaminant conditions, and to evaluate remediation options. One part of the DVZ-AFRC applied research effort integrates with these other efforts by providing laboratory- through field-scale data with a focus on relating vadose-zone process descriptions, contaminant nature and extent, and processes for treating the contaminant(s) discharge. These activities support development of site conceptual models and evaluating the long-term performance of remedies and are also related to DOE-EM's Advanced Simulation Capability for Environmental Management (ASCEM) program efforts. ASCEM will aid EM in transitioning scientific results into applied solutions that will be developed through the necessary framework of the DVZ-AFRC, which facilitates integration of resources and creativity to provide sites with viable alternative remedial strategies to current baseline approaches for persistent deep vadose zone contamination. The DVZ-AFRC will provide ASCEM information on site-specific hydrogeology and biogeochemistry defining contaminant source characteristics and controlling processes, and remedial strategies including remedial and monitoring technology implementation and performance metrics. ASCEM will use this information to assess the performance of remedial strategies and, through integration with the DVZ-AFRC, facilitate development of the scientific foundation, applied technologies, and remedial strategies necessary to make sound and defensible remedial decisions that will successfully meet the target cleanup goals in a manner that is acceptable to regulators. The DVZ-AFRC program also includes technology development efforts to enable application of innovative remediation techniques and improved amendment delivery processes for the deep vadose zone, to enhance remedy performance monitoring through application of geophysical techniques, and to develop flux-based monitoring for the vadose zone.

The technical objectives of the DVZ-AFRC effort are focused on four research and development categories. Within each of these categories, critical research and development lines of inquiry and opportunities are being identified and collaborative relationships with subsurface characterization and remediation activities at Hanford are being established. The four research and development categories are:

- **Remedial Design -** Perform fundamental and applied research supporting the design of surface and subsurface techniques to access and remediate DVZ contamination.
- **Controlling Processes -** Quantifying coupled hydrologic, geochemical, and microbial processes functioning in the DVZ is key to developing reliable conceptual models of moisture flux, contaminant movement, and remediation process efficacy in deep vadose zone environments.
- **Monitoring** – Develop and deploy efficient and effective monitoring methods for assessing the performance of remedies and for determining any long-term threat of contaminants reaching groundwater. Advance subsurface monitoring technologies including novel sensors, detectors, and data transmission techniques.
- **Predictive Modeling & Data Integration** - Simulate the integrated processes controlling moisture flux, contaminant transport, and remediation performance

As illustrated in Fig. 2, this effort must link all stages of the scientific, technology development, engineering and remediation processes. This effort will maintain a working interface with the ongoing site characterization and technology development and testing program, and will extend and focus scientific efforts toward achieving the outcomes necessary to support viable remediation strategies for the Hanford Central Plateau and deep vadose zone environments throughout the DOE complex.



- Basic research for fundamental new understanding, often on materials or systems that may be only peripherally connected or even unconnected to today's problems in energy and environmental technologies but that have the promise to revolutionize or transform.
- Development of new tools, techniques, and facilities, including those for advanced modeling and computation.

- Basic research for fundamental new understanding, usually with the goal of addressing intractable problems on real-world applications in the energy technologies.

- Research with the goal of meeting technical milestones, with emphasis on the development, performance, cost reduction, and durability of materials and components or on efficient processes.
- Proof of technology concepts.
- understanding natural and engineered system behaviors
- advanced modeling and simulations coupled with laboratory and field applications

- Scale-up research
- At scale demonstration
- Cost reduction
- Prototyping
- Manufacturing R&D
- Deployment support
- Performance validation
- Risk reduction

Goal: new knowledge/understanding
Mandate: open-ended
Focus: phenomena
Metric: knowledge generation

Goal: practical technologies
Mandate: needs driven applications
Focus: performance
Metric: remedial strategy generation

Goal: practical target
Mandate: restricted to targets
Focus: performance
Metric: regulatory and programmatic milestones

Fig. 2. Linkage of Use-Inspired Basic Research and Applied Science to Support Technology Deployment

This effort will include studies that span sufficient space and time scales to represent processes relevant to deep vadose zone applications. This includes molecular-scale functioning at solid/liquid interfaces through small and large field-scale investigations covering cubic meters to cubic kilometers, examined by borehole tests and geologic-formation, waste-site, and watershed-size modeling. Close collaboration will be required between the applied engineering, technology development, and science programs to translate scientific and advanced treatability findings into improved, realistic models of migration and swiftly use these new capabilities to meet the deep vadose zone remediation program goals.

HANFORD SITE DEEP VADOSE ZONE CHALLENGES – EXTENT OF THE PROBLEM

From the 1940s to the early 1990s, the Hanford Site (Fig. 3) released nearly 1.7 trillion liters (450 billion gallons) of contaminated liquid into the soil at the central portion of the site, known as the Central Plateau (Fig. 4). These liquids ranged from high-volume slightly contaminated cooling water to more concentrated effluents contaminated with chemicals and radionuclides resulting from spent-fuel reprocessing and plutonium recovery operations. These operations resulted in widespread contamination of the vadose zone and groundwater. Some of these discharges have reached the groundwater and migrated to the Columbia River, seven miles or more away. Unintentional discharges of more highly concentrated waste from Hanford's single-shell tanks (up to 3.8 million liters) also occurred. Today, much of that contamination remains in the vadose zone, though some radionuclides and hazardous chemicals have the potential to contaminate the underlying groundwater. This groundwater eventually discharges to the Columbia River.

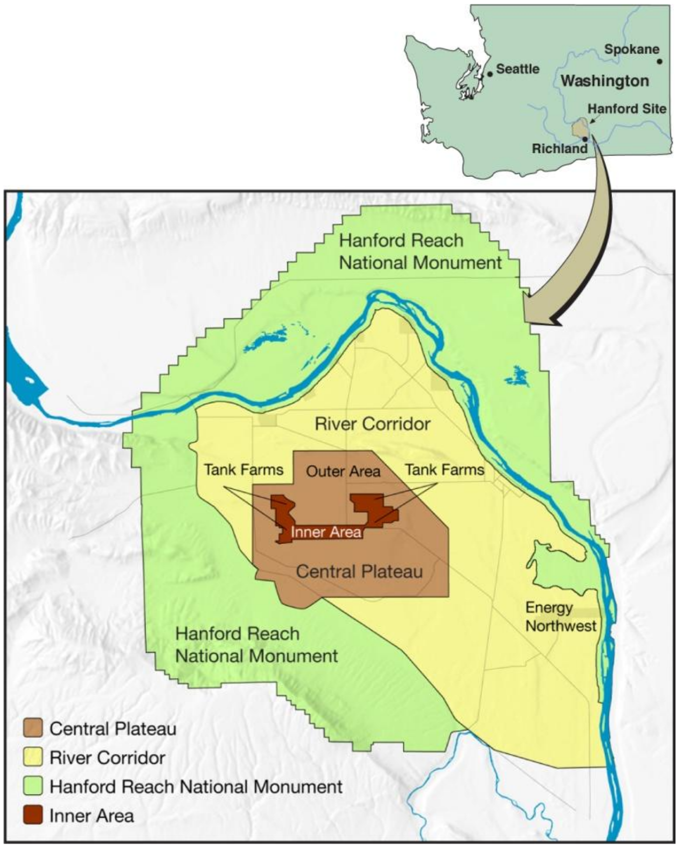


Fig. 3. Location of the Hanford Site and the Central Plateau

A vast majority of Hanford's remaining in-ground contaminants reside in the vadose zone of the Central Plateau, where reprocessing operations occurred. The vadose zone comprises 50 to 100 meters of water-unsaturated sediments above groundwater. The two principal deep vadose zone contaminants of concern are Tc-99 and uranium [2]. Other contaminants such as I-129 and nitrate are also prevalent in the deep vadose zone and groundwater. There is also a large carbon tetrachloride plume in the groundwater; however this problem is being addressed through a combination of soil vapor extraction and groundwater extraction and treatment [3].

The sites described below represent a broad range of problem types that are examples of key areas of interest for deep vadose zone investigations addressed through Hanford's deep vadose zone project efforts. Much of this contamination resides in the deep vadose zone, which is defined as the region below the practical depth of surface remedy influence (e.g., below excavation or surface barrier influence) and above the local water table (Fig. 4). If left untreated, these contaminants could reach groundwater and could remain a threat for centuries. Additional information on Central Plateau waste sites, release inventories and potential effects on groundwater can be found in these references [4,5,6].

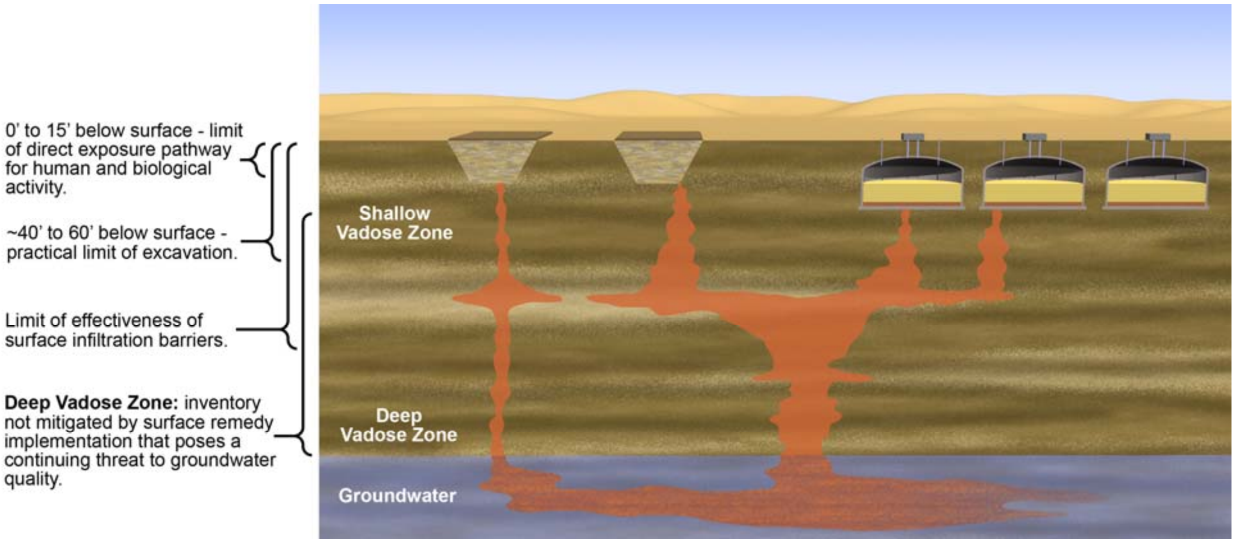


Fig. 4. Major subsurface strata and associated contaminant and remediation challenges

- **Laterally extensive subsurface plumes of mobile contaminants**. The BC cribs and trenches are characteristic of vadose zone contamination in the Central Plateau. This group of 26 cribs and trenches in the southern portion of the 200 East Area received about 115 million liters (30 million gallons) of liquid waste cascaded[2] between tanks from 1956 to 1965. This liquid contained about 410 curies of Tc-99 [6]. While groundwater monitoring wells are sparse in this area, there is no evidence that the contamination has reached groundwater which is about 90 meters deep in this area. Initial characterization efforts indicate that the Tc-99 inventory is located mostly at a depth in the vadose zone of between 30 and 45 meters below ground surface (bgs) and is spread across an area of nearly 0.14 km^2. Transport model predictions, however, indicate the potential for this contamination to adversely affect groundwater in the future if remedial actions are not successful [7].
- **Commingled tank farm and non-tank farm plumes**. There are several locations on the site where tank farms and their associated engineered disposal structures have both resulted in deep vadose zone contamination and even groundwater contamination. These include the areas around the B-BX-BY Waste Management Area (WMA), the S-SX WMA and T WMA. The commingling of the plumes makes source identification difficult. The principal contaminants of concern in these areas are Tc-99 and uranium. In 1951, the single-shell tank BX-102 was inadvertently overfilled and released about 265,000 liters (70,000 gallons) of tank waste containing about 10,000 kgs of uranium [6]. In the mid- to late-1990s, a breakthrough of uranium to the groundwater about 100 meters northeast of this tank was detected [8]. Analyses of uranium isotopic signatures point to BX-102 as the likely source of much of the uranium entering the groundwater in this area [9]. Groundwater concentrations of uranium have risen steadily since the mid-1990s and have reached more than 5,000 µg/L [8] – the highest concentration detected in Hanford groundwater.
- **Deep vadose zone plumes from processing facilities**. Hanford's five processing canyons have liquid disposal sites associated with them – several of which have resulted in existing groundwater plumes. These disposal sites have residual deep vadose zone inventories that have the potential to continue releasing contaminants into the groundwater.

HANFORD DEEP VADOSE ZONE REMEDIATION DECISION MAKING AND COMPLETION STRATEGY

The U.S. Department of Energy (DOE) and its regulatory agencies, the Washington State Department of Ecology (Ecology) and the U.S. Environmental Protection Agency (EPA) have developed a new approach for reaching remedy decisions for deep vadose zone areas on the Central Plateau [10]. This approach creates a single

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[2] Cascading refers to the practice of allowing waste to flow through a series of three or more single-shell tanks. This provided time for most transuranic-containing solids to settle. Often the resulting supernatant was discharged to the soil through cribs and trenches designed to aid in the dispersal and containment of the discharged liquid within the vadose zone.

Comprehensive Environmental Response, Compensation, Liability Act (CERCLA) operable unit encompassing multiple deep vadose zone sites.

The deep vadose zone operable unit has been initiated with an initial set of waste sites (cribs and trenches) that are primarily located adjacent to tank farms and that are known to have or are strongly suspected to have deep vadose zone problems. As new deep vadose zone sites are identified, these will be assigned to the deep vadose zone operable unit for remedy selection. DOE anticipates that decisions for past releases from single-shell tanks that have reached the deep vadose zone will also be addressed through this operable-unit approach. Previously these sites were assigned to many different operable units and each operable unit would have had its own characterization and remedy selection process.

This new approach allows deep vadose problems with common characteristics and challenges to be addressed by a single focused project. In addition, DOE has begun implementing a "defense-in-depth" approach to protect groundwater at Hanford so as to:

- **Provide project-like focus for the deep vadose zone** – A "project" approach to deep vadose zone challenges offers more efficiency and clear requirements for deep vadose zone treatability testing.
- **Integrate consistent investigations and decision making** – Single, integrated investigation efforts in areas with commingled deep vadose zone plumes can evaluate the future threats to groundwater and yield more complete and consistent remedy decisions.
- **Enable joint or concurrent decisions (for adjacent or commingled areas)** – Remedy selection decisions that provide protection of groundwater will be made concurrently, rather than staggered in time as would have resulted from the previous approach.
- **Allow surface, groundwater and deep vadose zone remediation to proceed on reasonable schedules** – Surface remedy decisions for near-surface waste sites can proceed uninhibited by uncertainties in the deep vadose zone, and provide more complete remedies for the deep vadose zone once sufficient understanding of the threat and potential remedies is gained. The new approach allows time for the benefits of treatability testing to unfold.

Consistent with the "Defense in Depth" approach, the major elements of DOE's Central Plateau cleanup strategy include: (1) contain and remediate contaminated groundwater; (2) develop and implement a cleanup strategy to guide remedy selection from a plateau-wide perspective; (3) identify, evaluate, and deploy viable remediation methods for deep vadose contamination to provide long-term protection of the groundwater; and (4) conduct critical waste management operations in coordination with cleanup actions [11]. The objectives of DOE's overall integrated strategy for the deep vadose zone are to:

- Develop sufficient understanding of the nature and extent of deep vadose zone contamination and processes that affect fate and transport;
- Develop predictive capabilities for describing contaminant fate and transport as well as flux from the vadose zone to the groundwater under natural and remediated conditions;
- Develop, test and deploy effective methods for remediating contaminated areas; and
- Develop and deploy effective monitoring methods for assessing the performance of remedies and for determining the long-term threat of contaminants to the groundwater.

New technologies and innovative ideas must be developed, tested and applied to resolve Hanford's deep vadose zone problems. However, it is unlikely that a single technology will adequately solve contamination problems in the deep vadose zone. Effective long-term remediation and protection of the underlying groundwater and vadose zone must rely upon a combination of approaches that collectively will control the flux of contaminants to the groundwater.

The following sections provide examples of current collaborative efforts among basic and applied researchers and Hanford's subsurface remediation program. These collaborations provide initial efforts that DOE plans to expand upon in the coming years.

DOE's Office of Science – Science Focus Areas

The DOE Office of Science funds Scientific Focus Areas (SFAs) [12] to resolve critical Hanford and basic subsurface science issues through integrated and multi-disciplinary research focused on the role of microenvironments and transition zones in reactive transport of technetium, uranium, and plutonium. Microenvironments are small domains within larger ones that exert a disproportionate influence on subsurface contaminant migration. They may be internal fractures or microbiologic niches within porous media lithic fragments; grain coatings, biofilms, or microcolonies on larger mineral particles; or compact silt/clay stringers in gravel-dominated subsurface sediments. Transition zones are field-scale features in which chemical, physical, or microbiologic properties change dramatically over relatively short distances (e.g., $\leq$ 1 m). They exhibit steep, transport-controlled gradients of system-controlling chemical species such as O_2, $H+$, or organic carbon. Microenvironments and transition zones frequently dominate subsurface contaminant reactivity, with strong effects resulting from the coupling of chemical reactions, physical transport (advection, diffusion), and microbiologic processes. Past research has documented the importance of these zones at the Hanford Site [12].

The overall goals of the SFA are to develop: 1) an integrated conceptual model for microbial ecology in the Hanford subsurface and its influence on contaminant migration, 2) a fundamental understanding of chemical reaction, biotransformation, and physical transport processes in microenvironments and transition zones, and 3) quantitative biogeochemical reactive transport models for technetium, uranium and plutonium that integrate multi-process coupling at different spatial scales for field-scale application. The SFA emphasizes lab-based, coupled computational and experimental research using relevant physical/biologic models, and sediments and microbial isolates from various Hanford subsurface settings to explore molecular, microscopic, and macroscopic processes underlying field-scale contaminant migration. The SFA also pursues the refinement of geophysical techniques to define, characterize, and map spatial structures and reactive transport properties of microenvironments and transition zones in the field [12].

The SFA supports DOE's cleanup mission and long-term stewardship responsibilities by providing new insights into the behavior of contaminants. These insights, which are derived from micro-scale studies or from laboratory tests, ultimately require validation in natural materials and at the field scale. This is an important part of assessing the accuracy of conceptual and/or computational models of subsurface contaminant transport and of determining the relative importance of various biogeochemical mechanisms postulated to affect contaminant transport and/or transformation. *In situ* field investigations also provide an opportunity to test measurement and monitoring tools developed to describe subsurface processes. The Hanford Site provides a unique environment for applying a wide array of fundamental research concepts and tools to critical environmental problems.

Advanced Remedial Methods for Metals and Radionuclides in Deep Vadose Zone Environments

In 2008, the U.S. Department of Energy initiated deep vadose zone treatability testing to seek remedies for Tc-99 and uranium contamination. These tests include applying desiccation for Tc-99 and reactive gas technologies for uranium. The defense-in-depth approach will implement multiple approaches to understand and control contaminant flux from the deep vadose zone to the groundwater. Among these approaches is an increased investment in science and technology solutions to resolve deep vadose zone challenges including characterization, prediction, remediation, and monitoring.

DOE's Office of Environmental Management, Groundwater and Soil Remediation (EM-32), has initiated efforts to develop advanced remedial methods for metals and radionuclides in the vadose zone at DOE sites. EM-32 is seeking to develop transformational technologies and innovative remedial strategies in order to meet remedial action objectives and long-term stewardship goals. These efforts advance the understanding of fundamental controlling processes described through Office of Science research to provide viable solutions that complement the Hanford Site deep vadose zone science and technology development activities.

One such advanced remedial strategy is described in these proceedings in Paper No. 11026 [13]. Efforts within this initiative are working to transform foam technology into a viable method for delivering remedial amendments to vadose zone environments. In contrast to solution flow in water-based delivery systems, foam flow under vadose zone conditions is not dominated by gravity; rather, it can be directed by pressure gradients in the sediments. This avoids the problem of uneven remedial fluid distribution in heterogeneous subsurface environments, and, in particular, facilitates lateral migration and penetration into low-permeability zones, which generally contain the majority of the contamination. Furthermore, the use of foam also provides better control on the volume of fluid (~ < 20% vol.) required for remedy delivery, thereby reducing the potential for unintended contaminant mobilization.

Integral to the successful development of any remedial technology is the ability to monitor, and eventually predict, the delivery, emplacement, and long-term performance of the treatment. In general, such monitoring is complicated by subsurface heterogeneity and by the disparity of scales that the hydrological properties span. These properties control the distribution of the remedial amendment and thus the location of subsequent transformations. Conventional techniques for subsurface hydrologic, geochemical, and biologic monitoring rely on wellbore-based approaches to collect samples or make measurements. Because of their limited spatial extent, these methods often cannot provide sufficient information to describe key controls on subsurface flow and transport. This is especially true in the vadose zone, where vertical infiltration pathways can form as a result of variable saturation and heterogeneity and where recovering fluid for sampling can be challenging.

The inability to conventionally characterize controlling properties and induced processes at a high enough spatial resolution, and over a large enough spatial extent, prohibits accurate assessment of foam-based delivery technologies for deep vadose zone treatments. To this end, we are advancing the application of radar and complex resistivity methods, which encompass three different geophysical attributes. Briefly, radar methods are expected to provide information about the dielectric constant, which is sensitive to soil moisture and may also respond to the reactive foam. Complex resistivity measures both the frequency-dependent electrical conductivity and induced phase response of the media to an external current. The electrical conductivity is expected to be useful primarily for monitoring the change in saturation and total dissolved solids associated with the reactive foam.

Finally, this initiative is developing the use of natural markers as a long-term monitoring approach to assess the effectiveness of remedial treatment and reaction of community dynamics. This profiling can be performed rapidly at the point source and at downstream gradients where microbial community changes may occur in advance of measurable geochemical metrics, thereby providing a highly sensitive “warning” of possible changes in contaminant plume behavior or the need for additional *in situ* remediation.

EM-32 has also prepared integrated research approaches for key problems within the DOE complex [14]. These approaches provide examples of how to effectively link basic and applied research activities with DOE-site field remediation projects. A specific example was prepared for the BC cribs and trenches at the Hanford Site. DOE’s current deep vadose zone planning effort will build upon this model.

CURRENT DEEP VADOSE ZONE TREATABILITY TESTING

To complement the preceding strategy for decision making, DOE has initiated a series of treatability tests to identify and evaluate potential remedies for deep vadose zone contamination [4]. The results of these tests will feed directly into remedy selection decisions. The overriding objective of the treatability testing is to evaluate specific vadose zone remediation technologies for Tc-99 and uranium, the primary long-term risk drivers at the site. The treatability test approach includes laboratory tests, modeling, and field tests of promising technologies. *In situ* treatment and surface barrier technologies offer promise for immobilizing contaminants in place, minimizing worker risk by eliminating the need to handle waste materials, and eliminating the need to transfer waste to another location where risks must be managed (e.g., to the Environmental Restoration Disposal Facility, often within a few miles of Central Plateau deep vadose zone sites). However, *in situ* technologies for application to the Hanford deep vadose zone are not yet sufficiently developed and tested to enable adequate evaluation as a remedial alternative. Initial efforts have focused on evaluation of soil desiccation, reactive-gas treatments, surface barrier effectiveness, grouting and soil flushing for the deep vadose zone. These efforts are described below.

- **Tc-99 Desiccation Testing -** Soil desiccation is a potential *in situ* technology for the arid conditions and the thick vadose zone at Hanford [4, 15, 16, 17]. In the past, contaminants were discharged to the soil along with significant amounts of water; this water continues to drive contaminants deeper in the vadose zone toward groundwater. Surface barriers can limit the rate that additional water moves into the vadose zone, and thereby slow contaminant movement. Desiccation could augment surface barriers by removing pore water, the driving force, to further slow the rate of contaminant movement to the groundwater. Field testing of desiccation is underway at the BC Cribs and Trenches waste site [17].

- **Pore Water Extraction Testing -** Recently, DOE initiated an investigation of applying vacuum to extract pore water based on observations during the initial activities for the desiccation field test. This work has initially included modeling to evaluate the area of influence for high-vacuum pore water extraction within the deep vadose zone. Current efforts are proceeding with laboratory validation of the pore water extraction processes and controlling factors, and with planning for future field tests.

- **Uranium Sequestration Testing -** Some reactive gases and gas-advected aqueous reactants (at low water content) can induce geochemical changes in sediments that act to render contaminants such as uranium less mobile. A range of potential amendments was tested in the laboratory [18]. The amendments targeted oxidation/reduction reactions, pH manipulation, or phosphate addition to induce precipitation reactions.

  Based on the laboratory test results, pH manipulation with ammonia gas proved to be effective in reducing uranium mobility and is amenable to application in the Hanford vadose zone [18]. When dilute ammonia gas is injected into vadose zone sediments, it rapidly partitions into the pore water. A portion of the ammonia dissociates and increases the pore water pH to nearly 12. Under these conditions, ions desorb and aluminosilicates dissolve. After cessation of ammonia injection, buffering and the loss of ammonia occur over time, the pH declines, and the ions in solution precipitate. These precipitates coat and bind uranium contamination, rendering it much less mobile. In this process, uranium is not chemically reduced, so the oxidation state does not affect treatment effectiveness. Laboratory experiments have shown this process to be robust in many Hanford sediments [18, 19]. Field testing is planned to further evaluate the process for application in the Hanford vadose zone.

- **Surface Barrier Evaluation -** Surface barriers limit the flux of contaminants from the vadose zone to the groundwater by reducing the infiltration of water at the surface. Surface barriers have been shown to be effective for shallow contamination, but their effectiveness in the deeper vadose zone is still uncertain. To augment the desiccation testing and uranium sequestration testing, an in-depth evaluation of existing information on surface barriers was conducted to develop a strategy for evaluating use of surface barriers as an element of an integrated strategy to address deep vadose zone contamination [20].

  The hydrologic properties of the vadose zone are key factors influencing the effectiveness of surface barriers for deep contamination. In scoping simulations, for a simple homogeneous vadose zone a surface barrier significantly increases the travel time of water (and contaminants) in the vadose zone to the groundwater. For instance, with natural surface infiltration rate conditions, travel time to the groundwater from 25 m below ground surface was simulated to be about 1500 years [7]. With a surface barrier emplaced to impose a very low infiltration rate (0.5 mm/yr), travel time increases to over 7500 years; thus contaminants would be only slowly released to groundwater. However, heterogeneous conditions in the vadose can alter the effectiveness of the barrier by imposing changes to the way in which water moves, in preferential vertical paths or laterally, in the vadose zone. The surface barrier evaluation identified the additional information needed to predict the performance of barriers for the heterogeneous conditions at the Hanford Site and potential activities to obtain this information.

- **Grouting Technologies -** Although *in situ* grouting is a mature technology, identification of an appropriate grouting configuration, grout penetration in deep vadose sediments, and verification of proper placement are the principal challenges to implement *in situ* grouting technology [4]. An initial evaluation of *in situ* permeation grouting is being conducted with the goal of providing information for use in subsequent feasibility studies for the Hanford Site deep vadose zone. The focus of this initial evaluation is to review candidate grout materials/formulations and techniques for their application to the Hanford Site 200 Area and to model grout penetration as a function of the physical properties of candidate materials and example subsurface properties.

- **Soil Flushing -** Soil flushing operates through the addition of water, and if necessary an appropriate mobilizing agent, to mobilize contaminants and flush them from the vadose zone and into the groundwater where they are subsequently captured by a pump-and-treat system. There are uncertainties associated with applying soil flushing technology to contaminants in the deep vadose zone at the Hanford Central Plateau. Modeling and laboratory efforts have been conducted to provide a quantitative assessment of factors that affect water infiltration and contaminant flushing through the vadose zone and into the underlying groundwater [21]. Soil flushing was evaluated primarily with respect to applications for technetium and uranium contaminants in the deep vadose zone of the Hanford Central Plateau.

SUMMARY AND FURTHER WORK

The risk posed by deep vadose contamination creates an enormous environmental liability. DOE is committed to bringing forward the best capabilities possible to address the deep vadose zone at the Hanford Site and across the DOE complex. The impact of Deep Vadose Zone - Applied Field Research Center investments is to develop effective and economical solutions at the Hanford and other DOE sites, while building upon available knowledge and capabilities, to meet cleanup goals. This approach will leverage investments from different DOE organizations, including sites across the DOE complex, working in basic science, applied research, and site engineering activities. DOE will use expertise from agency-wide activities, national laboratories, academia, and industry to work in collaboration with the Tri-Party Agreement signatories, site contractors, the public, and others to provide viable remedial technologies and strategies targeting baseline needs.

This approach will rely upon multi-project teams focusing on coordinated subsurface projects across the Hanford Site, and will facilitate research investments by implementing a Deep Vadose Zone - Applied Field Research Center located at Hanford and relying upon scientific studies from other DOE sites. The Center will provide a technical basis to quantify, predict, and monitor natural and post-remediation contaminant discharge from the vadose zone to the groundwater and to facilitate developing *in situ* solutions that limit this discharge and protect water resources. This knowledge will be used to transform science innovation into practical applications deployed by site contractors at Hanford and across the DOE complex. Carefully selecting investments will yield useful results within time frames supporting Tri-Party Agreement milestones, and support development of documentation strengthening cleanup decisions. Investments will support both time-critical decisions and long-term, non-time-critical objectives. Balancing these competing drivers will sustain both “bias for action” and “scientific sufficiency” priorities for program implementation. This will support development of sustainable solutions that are broadly applicable throughout similar environments within the DOE complex (e.g., Los Alamos, New Mexico, Idaho, and Nevada).

During FY 2011, treatability tests will continue to evaluate potential approaches to remediate deep contamination, and more closely integrated working relationships between user-inspired research and field-applied engineering will be established. In addition, a multiyear implementation plan is being developed to focus resource allocation on the most critical needs and opportunities.

ACKNOWLEDGEMENTS

Funding for this project was provided by the U.S. Department of Energy – Richland Operations Office and by the DOE Office of Environmental Management, Office of Groundwater and Soil Remediation (EM-32). In addition, the DOE Office of Biological and Environmental Research within the Office Science is involved in the effort through leveraged funding focused on complex systems science for the subsurface. The work was conducted at Pacific Northwest National Laboratory, operated by Battelle for the U.S. Department of Energy under contract DE-AC06-76-RL01830.

REFERENCES

1. U.S. Department of Energy. 2010. *Long-Range Deep Vadose Zone Program Plan*. DOE/RL-2010-89, Revision 0. U.S. Department of Energy, Richland Operations Office, Richland, Washington. Accessed December 15, 2010 at http://www.hanford.gov/files.cfm/DOE-RL-2010-89_Rev.0.pdf.

2. Morse JG, BL Charboneau, RW Lober, and MB Triplett. 2008. "Challenges for Deep Vadose Zone Remediation at the Hanford Site." *Proceedings of Waste Management 2008*, Paper No. 8407, Phoenix, Arizona. Accessed December 15, 2010 at http://www.wmsym.org/archives/2008/pdfs/8407.pdf.
3. U.S. Environmental Protection Agency. 2008. *Record of Decision Hanford 200 Area 200-ZP-1 Superfund Site, Benton County, Washington*. U.S. Environmental Protection Agency, Washington, D.C. Accessed December 15, 2010 at http://yosemite.epa.gov/r10/CLEANUP.NSF/sites/hanford2/$FILE/Hanford-200-ZP-1-ROD.pdf.
4. U.S. Department of Energy. 2008. *Deep Vadose Zone Treatability Test Plan for the Hanford Central Plateau*. DOE/RL-2007-56, Rev. 0, U.S. Department of Energy, Richland Operations Office, Richland, Washington. http://www5.hanford.gov/arpir/?content=findpage&AKey=0804110065.
5. Kincaid CT, MP Bergeron, WE Nichols, PW Eslinger, and MB Triplett. 2006. *A Ranking of Waste Sites Supporting the Prioritization of Groundwater Remedial Actions on Hanford's Central Plateau*. PNNL-16057, Pacific Northwest National Laboratory, Richland, Washington.
6. Corbin RA, BC Simpson, MJ Anderson, WF Danielson III, JG Field, TE Jones, and CT Kincaid. 2005. *Hanford Soil Inventory Model Rev. 1*. RPP-26744, Rev. 0, CH2M HILL Hanford Group, Inc., Richland, Washington.
7. Ward AL, GW Gee, ZF Zhang, and JM Keller. 2004. *Vadose Zone Contaminant Fate and Transport Analysis for the 216-B-26 Trench*. PNNL-14907, Pacific Northwest National Laboratory, Richland, Washington. Accessed December 15, 2010 at http://www.pnl.gov/main/publications/external/technical_reports/PNNL-14907.pdf.
8. Hartman MJ, JA Rediker, and VS Richie. 2009. *Hanford Site Groundwater Monitoring for Fiscal Year 2008*. DOE/RL-2008-66 Revision 0, U.S. Department of Energy, Richland Operations Office, Richland, Washington. Accessed December 15, 2010 at http://www.hanford.gov/c.cfm/sgrp/GWRep08/html/start08.htm
9. Christensen JN, PE Dresel, PG Conrad, K Maher, and DJ DePaolo. 2004. "Identifying the Sources of Subsurface Contamination at the Hanford Site in Washington Using High-Precision Uranium Isotopic Measurements." *Environmental Science & Technology* 38(12):3330–3337. Accessed December 15, 2010 at http://pubs.acs.org/doi/pdf/10.1021/es034700q.
10. U.S. Department of Energy. 2010. *Tentative Agreement on Hanford Federal Facility Agreement and Consent Order Change Forms Implementing Changes to Central Plateau Cleanup*. U.S. Department of Energy, Richland Operations Office, Richland, Washington. Accessed December 15, 2010 at http://www.hanford.gov/files.cfm/TPA_CP_Final_Approved_TA.pdf.
11. U.S. Department of Energy. 2009. *Central Plateau Cleanup Completion Strategy*. DOE/RL-2009-81, Revision 0, U.S. Department of Energy, Richland Operations Office, Richland, Washington. http://www2.hanford.gov/arpir/?content=findpage&AKey=1002180676.
12. U.S. Department of Energy. 2010. "Subsurface Science Scientific Focus Area (SFA) – Role of Microenvironments and Transition Zones in Subsurface Reactive Contaminant Transport." Project website at http://www.pnl.gov/biology/sfa/.
13. Wellman DM, SV Mattigod, SS Hubbard, A Miracle, L Zhong, M Foote, Y Wu and DP Jansik. 2011. "Advanced Remedial Methods for Metals and Radionuclides in Vadose Zone Environments." In press. *Proceedings of 2011 Waste Management*, Paper No. WM-11026, Phoenix, Arizona.
14. Pierce EM, MD Freshley, SS Hubbard, BB Looney, JM Zachara, L Liang, D Lesmes, GH Chamberlain, KL Skubal, V Adams, ME Denham, and DM Wellman. 2009. *Scientific Opportunities to Reduce Risk in Groundwater and Soil Remediation*." PNNL-18516, Pacific Northwest National Laboratory, Richland, Washington. Accessed December 15, 2010 at http://www.pnl.gov/main/publications/external/technical_reports/PNNL-18516.pdf.
15. Oostrom M, TW Wietsma, JH Dane, MJ Truex, and AL Ward. 2009. "Desiccation of Unsaturated Porous Media: Intermediate-Scale Experiments and Numerical Simulation." *Vadose Zone Journal* 8(3):643–650. Accessed

December 15, 2010 at https://www.soils.org/publications/vzj/articles/8/3/643?highlight=JmFydGljbGVfdm9sdW1lPTgmYXJ0aWNsZV9maXJzdHBhZ2U9NjQzJmxlbj0xMCZzdGFydD0xJnN0ZW09ZmFsc2Umc29ydD0%3D.

16. U.S. Department of Energy. 2010. *Characterization of the Desiccation Field Test Site as Part of the Deep Vadose Zone Treatability Test for the Hanford Central Plateau*. DOE/RL-2009-119, U.S. Department of Energy Richland Operations Office, Richland, Washington.

17. U.S. Department of Energy. 2010. Field Test Plan for the Soil Desiccation Pilot Test. DOE/RL-2010-04, Revision 0, U.S. Department of Energy Richland Office, Richland, WA.

18. Szecsody JE, MJ Truex, L Zhong, MD Williams, CT Resch and JP McKinley. 2010. *Remediation of Uranium in the Hanford Vadose Zone Using Gas-Transported Reactants: Laboratory-Scale Experiments*. PNNL-18879, Pacific Northwest National Laboratory, Richland, Washington. Accessed December 15, 2010 at http://www.pnl.gov/main/publications/external/technical_reports/PNNL-18879.pdf.

19. Szecsody, JE, MJ Truex, L Zhong, NP Qafoku, MD Williams, JP McKinley, CT Resch, JL Phillips, D Faurie, and J Bargar. 2010. Remediation of Uranium in the Hanford Vadose Zone Using Ammonia Gas: FY10 Laboratory-Scale Experiments. PNNL-20004, Pacific Northwest National Laboratory, Richland, WA.

20. Fayer MJ, AL Ward, and VL Freedman. 2010. *Technical Basis for Evaluating Surface Barriers to Protect Groundwater from Deep Vadose Zone Contamination*. PNNL-18661, Pacific Northwest National Laboratory, Richland, Washington.

21. Truex, MJ, M Oostrom, F Zhang, KC Carroll, J Schramke, TW Wietsma, GD Tartakovsky, and K Gordon. 2010. Evaluation of Soil Flushing for Application to the Deep Vadose Zone in the Hanford Central Plateau. PNNL-19938, Pacific Northwest National Laboratory, Richland, WA.