SUPPORT TO HANFORD ENVIRONMENTAL REMEDIATION AND LONG-TERM STEWARDSHIP





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Pacific Northwest National Laboratory (PNNL) provides continuity of scientific and technical expertise to support environmental remediation and site stewardship decisions at Hanford. Our partnership with the Hanford Site started with establishing the environmental monitoring programs that today protect critical water resources. PNNL provided the technical foundations for initiating the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA) process at Hanford and has developed numerous technologies and approaches for remediation that are being implemented. Building on these accomplishments, PNNL is well positioned to 1) support the U.S. Department of Energy (DOE) and site contractors in implementing the current baseline through technology maturation and technical guidance; and 2) develop new technology solutions and approaches through applied science to meet the challenges ahead, including defining and achieving risk-informed endpoints for active remediation. We are committed to addressing environmental issues with technically defensible solutions. As described in the following sections, PNNL stewards scientific and technical capabilities to provide four core competencies to support the DOE Office of Environmental Management (EM) mission at the Hanford Site:

- Nature and Extent of Contamination
- Remedial Design
- Remedial Action
- End States



Hanford Site-Wide Map



NATURE AND EXTENT

In 1965, PNNL established the environmental program and technical leadership and scientific expertise to conduct environmental surveillance of the Hanford Site, including terrestrial and river ecosystems, groundwater monitoring, and environmental and subsurface characterization. The program was successfully transferred to the site contractor in 2007. As the site begins to transition from remediation to closure and long-term stewardship, PNNL is working with DOE Richland Operations (RL) and the site contractor to help transition the program from characterization to remediation performance and long-term monitoring. In doing so, PNNL developed the Scientific Opportunities for Monitoring at Environmental Remediation Sites (SOMERS) approach, which is being applied and is projected to decrease the annual site monitoring budget by more than 30 percent. PNNL's characterization efforts provided the technical basis for initiating the Basalt Waste Isolation Project investigations of a potential repository in basalt rock.

In the late 1990s, serious challenges were presenting perceived threats to human health and the environment. The unexpected behavior of ¹³⁷Cs and other contaminants in the vadose zone and groundwater threatened progress on soil and groundwater cleanup at the Hanford Site. Contaminants were deeper in the vadose zone and more widespread in the groundwater than was predicted by conceptual and numerical models. The conceptual models were incomplete, in some cases incorrect, and could not be used to predict subsurface contaminant behavior. In response, DOE established the Hanford Groundwater/Vadose Zone Integration Project, where PNNL was a major partner with the DOE site office and site contractor. PNNL conducted investigations to provide the scientific knowledge regarding subsurface contaminant transport. These efforts led to development of conceptual and numerical models that more accurately predict past and potential future migration events in the vadose zone at Hanford. Results of these studies were



Environmental Molecular Sciences Laboratory (EMSL)

Capabilities for characterizing the features and processes controlling contaminant behavior, fate, and transport reside in the EMSL Facility. EMSL is a DOE Office of Science user facility equipped with state-of-the-art capabilities for acquiring a deep understanding of molecular-level processes necessary to gain a predictive, systems-level understanding of terrestrial systems.

published in nearly 150 papers. The following outcomes highlight the project impacts:

- Developed the Soil Inventory Model (SIM) to estimate the inventory of key contaminants at waste sites and associated tank leaks based on existing data and process knowledge. The SIM provides inventory estimates for all site-wide analyses of risk and impact at Hanford. The model provided the inventory basis for the recently completed *Final Tank Closure and Waste Management Environmental Impact Statement for the Hanford Site*, *Richland*, *Washington* (DOE/EIS-0391).
- Developed conceptual and numerical models that improved DOE's ability to describe previous migration events of ¹³⁷Cs, ⁹⁹Tc, U, and CrO₄²⁻ in the vadose zone beneath leaked single-shell tanks and to predict future transport for remedial assessment. This understanding led to improved reaction/transport models and formed the basis for planning future tank farm and vadose zone remedies.

- Developed a conceptual framework for understanding complex distributions of contaminants in the vadose zone resulting from interactions with fine-scale sediment texture and lateral spreading. These observations have provided a framework for interpreting complex contaminant distribution patterns in the subsurface as the basis for planning effective remedies at the Hanford Site, including BC Cribs and Trenches, one of the Hanford site's deep vadose zone (DVZ) contamination problems.
- Resolved uranium geochemistry at waste sites across Hanford; ongoing studies are updating the uranium conceptual model and providing input for designing a remediation approach. The uranium conceptual model provides the basis for planned remediation efforts under development for the Central Plateau.
- Identified and tested geophysical technologies to costeffectively interrogate the subsurface and delineate vadose zone plumes. A broad spectrum of geophysical methods was incorporated into the vadose zone transport field studies conducted by Integration Project researchers. Electrical geophysical methods were refined for subsurface characterization and monitoring of both the vadose zone and groundwater. An advanced high-performance geophysical imaging code was developed that reconstructs subsurface images using electrical resistance tomography (ERT) to "see" subsurface contaminant plumes in three dimensions and in unprecedented resolution. ERT provides a critical capability for integrating site characterization data and support implementation of remediation strategies.

Building on these accomplishments and contributions of the Hanford Groundwater/Vadose Zone Integration Project, PNNL is currently stewarding initiatives that will provide the technical basis for developing and enhancing site conceptual models. Specifically, we are providing the technical basis to support evaluation of remediation approaches of ¹²⁹I in the 200-UP-1 Operable Unit and enable an effective assessment of the natural attenuation element of remediation. Additionally, PNNL is providing the technical understanding of the interactions between co-contaminants in the 200-BP-5 and 200-UP-1 Operable Units to support development of remediation approaches for co-mingled contaminants and evaluate the impact on the geochemistry of the system. Finally, PNNL will provide a technical approach for assimilating time-lapse geophysical monitoring data into predictive simulators to assess current and future DVZ contaminant distributions and flux to groundwater with reduced uncertainty. These efforts will address data gaps in conceptual models and further characterize the distribution, fate, and transport of contaminants in the subsurface, which will provide a technical framework for remedial design and action.

REMEDIAL DESIGN

For more than 50 years PNNL has developed and maintained a robust capability in subsurface science. By integrating this capability with our core strength in systems engineering, PNNL has been designing, developing, and demonstrating novel remediation technologies from bench- to pilot-scale that today are the basis for remedial actions at Hanford. Our core approach is to understand the natural system and the features, events, and processes that control the behavior, transport, and fate of contaminants and use this understanding to develop effective remediation technologies and approaches, including incorporation of natural attenuation in remedy designs.

In 2010, EM headquarters, DOE RL, and the site contractors signed a memorandum of understanding that established the Deep Vadose Zone - Applied Field Research Initiative (DVZ-AFRI) at PNNL. The DVZ-AFRI is addressing the long-term protection of water resources at EM sites by developing and applying effective solutions for DOE's DVZ challenges in characterization, prediction, remediation, and monitoring. This is the cornerstone providing a technical basis to quantify, predict, and monitor natural and post-remediation contaminant discharge from the vadose zone to the groundwater and to facilitate in situ solutions that limit this discharge and protect water resources. The DVZ-AFRI provides critical scientific and technical underpinnings to link vadose zone processes, and contaminant nature and extent with remedial treatment processes to conduct treatability tests, quantify how technologies change subsurface and contaminant conditions, and evaluate the performance of remediation options. Results are published in peer-reviewed journals, including a special issue of the Vadose Zone Journal (Volume 11, Number 4, November 2012). Impacts of the DVZ-AFRI are reflected in the following major outcomes:



331 Building

The 331 Building is a radiological facility equipped to support many subsurface science and engineering initiatives at PNNL. Capabilities include characterization of contaminated sediment, metals and radionuclide research, sediment column flow experiments, remediation technology development, and systems-based monitoring.

- Identified and characterized mechanisms and processes that influence transport of vadose zone contaminants such as ⁹⁹Tc, which enabled predictions and guided site characterization. Simulations were performed with the Subsurface Transport Over Multiple Phases (STOMP) code developed at PNNL and the Advanced Simulation Capability for Environmental Management (ASCEM) developed collaboratively by DOE national laboratories. The characterization and simulation work supported implementation of a treatability test for soil desiccation at BC Cribs.
- Evaluated a range of waste disposal chemistries to provide scientific and technical support for remedial decisions. Conditions during waste disposal at the Hanford Site ranged from alkaline to acidic, which impacts the long-term geochemical behavior of contaminants and remedial design.

- Developed the scientific and technical basis for a mass-flux-based design and performance assessment approach to remediation in vadose zone environments and published a guidance document for use at DOE and Department of Defense sites. This approach provided the technical justification that performance objectives had been met so that Hanford's active soil vapor extraction system for carbon tetrachloride could be shut down.
- Extended the mass-flux-based framework to inorganic contaminants and radionuclides, providing decision support for monitored natural attenuation (MNA) in vadose zone environments. The mass-flux framework is being applied to problematic vadose zone contaminants including ⁹⁹Tc, ¹²⁹I, uranium, plutonium, and americium in the Central Plateau.
- Applied scientific and technical understanding of fate and transport including biogeochemistry and hydrogeology to define an approach for risk-informed endpoints for remediation.
- Developed a technical approach to manipulate vadose zone geochemistry with ammonia. Delivered in the gas phase, ammonia interacts with pore water to raise the pH and dissolve native minerals. This process is temporary, and as the native minerals re-precipitate, the contaminants are immobilized by being bound in or coated by these precipitates.

We are continuing to support DOE RL and site contractor efforts to address the most challenging problems within the Hanford Central Plateau. PNNL is currently assessing and quantifying the biogeochemical impacts on ⁹⁹Tc, uranium, and ¹²⁹I and biogeochemical interactions of co-mingled plumes to 1) enable resolution of critical data gaps that prevent selection of effective remedies; 2) enable scientific and technical support for leave-in-place strategies for these contaminants through evaluation and demonstration of biological immobilization/degradation; and 3) provide the scientific and technical understanding necessary to develop remediation approaches for co-mingled contaminant plumes. Furthermore, we are providing a scientific basis to develop and assess the performance of remedial alternatives for the vadose zone. These efforts in remedial design provide the basis and impetus for remedial action that will protect human health and the environment.

REMEDIAL ACTION

The key to successful transition of technologies from conception to implementation is a collaborative relationship between the technology developers and implementers. We are committed to supporting this collaborative effort with DOE site contractors to achieve mission objectives. Since the late 1980s, PNNL has worked closely with the site contractors to translate fundamental principles underpinning remediation technology performance to full-scale operations. PNNL's knowledge of the fundamental scientific and engineering principles allows us to effectively transition remediation technologies to the field scale and is central to our role in technology analysis and maturation. Our support enables the site contractor to implement new technologies within the site baseline. The following examples demonstrate our support for technology maturation and implementation:

- Quantified mechanisms for chromium sequestration, resulting in implementation of the In Situ Redox Manipulation Barrier to protect critical salmon spawning habitat in the Columbia River. Concepts for the redox barrier were developed during basic science studies. The Hanford Site and DOE Headquarters funded technology maturation and received an R&D 100 Award for the project. Currently, the redox barrier is part of a record of decision for the 100-D Area chromium plume.
- Developed phosphate remediation technologies for in situ remediation of contaminants. Phosphate remediation allows contaminants to be sequestered in place. Phosphate technology is currently being implemented in the 300 Area to sequester uranium in the periodically rewetted zone. This effort leveraged results from the DOE Office of Science 300 Area Integrated Field Research Challenge Project. Phosphate injection also has been tested for ⁹⁰Sr in the 100-N Area.
- Developed concepts for soil desiccation to reduce the driving force for vadose zone contaminants and reducing flux to groundwater. The concepts were evaluated



Vadose Zone Field Studies

Vadose zone field studies provide insight into the important remedial/transport processes, developing mass flux-based site conceptual models, and systems-based approaches. Studies conducted at vadose zone field sites helped to inform and support recharge measurement and characterization, surface barrier design, waste form degradation performance assessments, simulated tank leaks, subsurface heterogeneity characterization, remediation treatability development and demonstration, and geophysical characterization. The contributions of these field studies provide defensible criteria/data for defining risk-informed endpoints for cleanup, supporting remediation decisions, and developing and implementing systems-based remediation approaches and systems-based monitoring strategies.

in the laboratory and implemented in a field treatability test at BC Cribs. Removing the driving force, water, from the vadose zone delays contaminant transport and reduces mass flux to groundwater, protecting underlying water resources. Along with soil desiccation, proof-ofprinciple field tests were conducted for extraction of pore water as a means of remediating vadose zone contamination that is difficult to access.

- Implemented a treatability test for the Hanford Prototype Surface Barrier, from initial concepts and design to implementation. Surface barriers reduce recharge controlling mass flux of contaminants in the vadose zone. Combined with other remediation approaches such as soil desiccation, surface barriers will protect underlying water resources. Vadose zone monitoring support was also provided for interim surface covers implemented over past tank leaks at several waste management areas, reducing downward migration of radionuclides.
- Provided technical support and modeling for removal of high-uranium concentrations from perched water beneath the B Complex, reducing mass flux to underlying groundwater and protecting underlying water resources.

PNNL is currently developing and evaluating new approaches and technologies for full-scale remedial action implementation, including evaluation of pump-and-treat technologies, development of a technical evaluation plan for ¹²⁹I, and the development of DVZ enhanced implementation control and performance monitoring. Specifically, we are providing the technical evaluation of the 200 West pump-and-treat system fluidized bed reactor (FBBR) that contains a microbial community to treat carbon tetrachloride and nitrate. Biofilm materials were forming in the FBBR, decreasing efficiency. This effort optimized the FBBR component of the pumpand-treat system, resulting in uninterrupted operation and more efficient contaminant removal. We are also evaluating novel materials for ⁹⁹Tc, ¹²⁹I, and U removal by pump-andtreat. Results demonstrate the applicability of advanced inorganic composites for removal of ¹²⁹I and other contaminants from Hanford groundwater, and future work is focused on scale-up and engineering appropriate forms of the composites for deployment in the pump-and-treat system. PNNL is also developing and supporting implementation of sustainable groundwater remediation technologies consisting of solar-powered groundwater extraction systems. Utilizing solar-powered technology will ultimately

help minimize the environmental footprint of remediation efforts and reduce remediation and long-term monitoring costs. Demonstrations of the applicability of a pump-andtreat exit strategy focusing on systems installed in the Hanford 100 Area are also being conducted.

Within the 200-UP-1 Operable Unit, PNNL is developing a technical evaluation plan for remediation of ¹²⁹I. Iodine-129 is present in the groundwater and is one of the primary risk drivers for the site. Hydraulic containment is the currently selected remedy for ¹²⁹I in the groundwater as outlined in the 200-UP-1 Record of Decision and Risk Assessment Work Plan, but there is currently no remedy selected for controlling migration of ¹²⁹I from the vadose zone to the groundwater. Research is underway to develop a site conceptual model to decrease the uncertainty related to the inventory, spatial distribution, and transport properties, which will lead to appropriate treatment strategies.

Finally, PNNL is developing an enhanced control and monitoring system in parallel with planned vadose zone treatability testing (i.e., DVZ Treatability Test Program and 200-DV-1 treatability test efforts) to provide options for real-time control of remediation systems linked with performance monitoring. Integration and interpretation of monitoring data at a system-level related to performance goals is needed to facilitate effective treatment implementation that will also transition readily to longer-term performance monitoring of the vadose zone remedy.

END STATES

PNNL scientists and engineers are leaders in risk assessment and associated research to evaluate and mitigate potential nuclear safety issues. We currently perform this work for DOE, the Nuclear Regulatory Commission, industry, and international entities. However, this capability is deeply rooted in the Hanford mission. As the Hanford Site transitioned from plutonium production, operations, and waste management to cleanup, PNNL led the waste site evaluation that resulted in Hanford's listing on the National Priorities List. In 1998, PNNL conducted the first sitewide composite analysis to investigate cumulative impacts from existing and future wastes, to support performance assessments of individual waste management actions. We have conducted and provided technical underpinnings for numerous performance assessments and led environmental impact statements.

Our experience in risk assessments and remediation led to development of a framework for evaluating risk-based remediation endpoints, ultimately leading to final end states for the Hanford Site. Many of the remaining challenges at Hanford are complex, requiring a framework that provides a structured, systems-based approach to regulatory processes established for remediation under CERCLA and the Resource Conservation and Recovery Act. The framework establishes a technical approach intended to facilitate remedy decisions and implementation at complex sites where restoration may be uncertain, require long timeframes, or involve adaptive management approaches.

Recent and current accomplishments in implementing end states include the following:

Led a probabilistic seismic hazard analysis (PSHA) of the Hanford Site for DOE and Energy Northwest. The analysis provides a detailed characterization of the vibratory ground motion hazard at the Hanford Site from potential future earthquakes. The PSHA is just the first step in reassessing the seismic safety of facilities located on the Hanford Site and establishing design requirements for new facilities.



Institutional Computing

The Institutional Computing program at PNNL is focused on Laboratory needs and DOE missions. One critical component of the program is Olympus, a 162-Teraflop peak supercomputer that can compute as fast as about 20,000 typical personal computers combined. Parallel application software such as eSTOMP and ASCEM are implemented on supercomputers such as Olympus to provide detailed calculations in simulations that address complex issues such as reactive transport and challenging unsaturated vadose zone problems such as very dry conditions in soil desiccation.

- Supported implementation of MNA as part of the 100-F Area Operable Unit Record of Decision. PNNL quantified site-specific chromium attenuation data for the Hanford 100 Area to support the attenuation remedy. Natural attenuation is part of any remedy, but often the technical basis has not been established.
- Expanded the template for MNA to contaminants in the Central Plateau, consistent with existing guidance that incorporates information and analysis approaches based on recent efforts including the vadose zone flux framework, waste chemistry assessment, conceptual model efforts, and studies related to ¹²⁹I, ⁹⁹Tc, and uranium. Ultimately, MNA will be a significant component of remediation of Central Plateau waste sites for the 200-DV-1 Operable Unit.

Achieving end states also requires that long-term stewardship and monitoring be implemented. Recent PNNL efforts have focused on developing monitoring technologies to enable cost-effective long-term stewardship:

- Provided the scientific underpinnings and technical framework for advancing from point-source to integrated systems-based monitoring at DOE sites; the approach is beginning to be implemented at the Hanford Site with assistance by PNNL.
- Developed a web-based application, the PNNL Hanford Online Environmental Information eXchange (PHOENIX), including a version for the annual groundwater report. PHOENIX provides analysis tools and access to groundwater monitoring results, to present groundwater monitoring and remediation treatment information on a geographic information system (GIS) framework. PHOENIX significantly reduced the costs of reporting monitoring data and made it much more accessible and useable for stakeholders and regulatory agencies.

PNNL is developing and implementing risk-informed approaches for endpoint selection and the establishment of systems-based monitoring programs. Specifically, in the technical evaluation plan for the selection of a remediation endpoint for ¹²⁹I, PNNL is building on previous efforts to include an evaluation of ARAR (applicable or relevant and appropriate) waivers. We are also working with the site contractor to develop a structured approach to assessing pump-and-treat performance to support a decision to optimize, transition, or close a pump-and-treat remedy. The approach focuses on identifying data collection and analysis that will support decision makers in evaluating a pumpand-treat remedy endpoint for systems with diminishing returns. Metrics for evaluating appropriate endpoints include assessment of the contaminant mass discharge and the attenuation capacity within the aquifer. This approach provides DOE RL with a strategy for transitioning or terminating pump-and-treat operations once they reach a point of diminishing return. Finally, PNNL is providing the technical basis for using sensors and sensor system technology to reduce costs and improve the efficiency and quality of vadose zone and groundwater monitoring.

Despite significant progress, Hanford still faces some of the most technically difficult challenges ever encountered in environmental remediation. Advances in science and engineering and transformational approaches to cleanup are required to address long-term cleanup challenges. Going forward, a technically defensible strategy for remediating deep vadose zone contamination and protecting underlying groundwater resources requires a sustained investment and increased resources to accelerate the reduction of impacts from legacy waste. The following elements are key to this strategy.

- Complete development of the Technical Basis for Remedial Action through site characterization necessary to develop improved site conceptual models depicting information about contaminant source terms, the distribution of contaminants in the environment, the hydrological properties of the site, and the physical setting with respect to potential impacts to human health and the environment. This will allow contamination issues to be defined for input into baseline risk assessments and definition of effective remedial approaches.
- Use Systems-Based Assessment to determine the remediation approach, including 1) resource use strategy, 2) refining the conceptual model, 3) remedial investigation, and 4) assessing risk and appropriate endpoints for the site.
- Develop and adaptively apply a Systems-Based Monitoring strategy, integrated into conceptual models, to identify appropriate lines of evidence (monitored parameters) that can be used to verify that contaminant behavior over time is within expected limits, will meet site remediation goals, and maintain compliance.



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