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Mitigating the Risks of Contamination within Vadose Zone Environments

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Mitigating the Risks of Contamination within Vadose Zone Environments

A SYSTEMS-BASED ASSESSMENT FOR RISK-INFORMED REMEDIATION OF THE HANFORD DEEP VADOSE ZONE

Summary

Vadose zone environments across the U.S. Department of Energy's (DOE's) legacy waste sites represent significant long-term sources of contamination, impacting groundwater and, ultimately, site closure decisions. Cleanup decisions must be correlated to residual inventories of risk-driving contaminants (e.g., ^{99}Tc , uranium ^{239}Pu , chromium, and nitrate) through comprehensive systems-based analyses to provide holistic, integrated approaches to remediation and protection of water resources. The Hanford Site possess the largest fraction of the DOE legacy waste inventory. While some of this inventory has already impacted the groundwater, a substantial inventory remains within the vadose zone and will impact groundwater long into the future. Recently, significant improvements have been made in methods for detecting and characterizing soil moisture and contaminant releases, understanding and decreasing mass-flux, and remediating deep vadose zone and groundwater plumes. However, a consistent, collaborative, and sustained effort including engagement of the DOE Office of Science is necessary to advance progress toward cleanup goals and protect water resources. The following actions are recommended:

1. **Implement the risk-informed, systems-based endpoint framework** to 1) define priorities for cleanup activities; 2) define the technical specifications for cleanup approaches, and 3) provide critical assessments of proposed solutions for the remaining cleanup challenges for the DOE Office of Environmental Management.
2. **Develop a science and technology roadmap to identify the gaps in the Hanford Site baseline** to achieve risk-informed, systems-based endpoints.
3. **Integrate fundamental science and applied research and development to provide risk-informed, systems-based solutions** for remediation of source terms (e.g., waste sites and tanks) and the underlying subsurface environment (i.e., vadose zone), and to protect water resources. Key activities include the following:
 - a. **Develop and maintain mass flux-based conceptual models** for predictions of contaminant behavior and migration to establish risk-based priorities for remedial actions.
 - b. **Implement advanced scientific approaches**, such as monitored natural attenuation for vadose zone contamination, to protect groundwater resources.
 - c. **Decrease mass flux from the vadose zone to groundwater** through treatability testing and implementation of remedies to prevent or mitigate emerging groundwater plumes.
4. **Implement systems-based monitoring programs** to provide an integrated, holistic understanding of contaminant behavior and migration and remediation performance over the entire subsurface profile (land surface to groundwater) as a function of time.

Introduction

The U.S. Department of Energy's (DOE's) legacy waste inventory includes significant long-term sources of contamination within vadose zone environments that will impact groundwater and limit cleanup and closure decisions long into the future. Decisions and approaches to protect and remediate groundwater must be linked to

contaminant “source terms” and key risk-drivers, such as ^{99}Tc , uranium ^{129}I , chromium, and nitrate, within vadose zone environments using holistic, integrated assessments.

In addition, a number of DOE sites within the western United States have deep vadose zone (DVZ) environments, including the Hanford Site, Idaho National Laboratory, Los Alamos National Laboratory, and the Nevada Nuclear Security Site. The DVZ is defined as the region below the zone of practicable excavation and removal, but above the groundwater table. DVZ environments that have been contaminated can continue to be primary sources and pathways for contaminant migration to groundwater and present unique challenges for remediation, management, and monitoring.

The thickness, depth, and intricacies of the vadose zone and DVZ environments, combined with a lack of understanding of key subsurface processes (e.g., biogeochemical and hydrologic processes) affecting contaminant migration, make it difficult to develop and validate conceptual models that sufficiently and accurately represent subsurface flow dynamics and contaminant behavior across multiple scales to enable quantification of contaminant fate and transport. In turn, these factors make it difficult to design and implement sustainable remedial approaches and monitor long-term contaminant behavior and remedial action performance.

Traditional approaches for contaminated sites assume that contamination in DVZ environments is isolated from exposure such that direct contact is not a factor in its risk to human health and the environment (Hoover et al. 2008). However, decades of scientific and technical investigations supported by the DOE Office of Environmental Management, the DOE Office of Science, the U.S. Department of Defense (DOD), and the U.S. Environmental Protection Agency (EPA) have demonstrated that the transport and discharge, or flux, of contaminants from vadose source terms to groundwater, creates the potential for exposure and risk to receptors using groundwater as a resource or by discharge to surface water resources. Therefore, while DVZ environments are not considered a resource requiring restoration, limiting flux from this environment and understanding and quantifying the movement (or lack thereof) of contaminants through or within this unit is critical to protecting water resources and determining the risks to human health and the environment, particularly over the long-term as conditions, actions, and land uses change or are altered.

The current methods for assessing the threat to groundwater and surface water receptors from vadose zone source terms are based on establishing remedial action goals using fate and transport modeling (Hoover et al. 2008). However, the conceptual and numerical models used to establish these cleanup goals may not quantitatively account for critical processes controlling contaminant fate, transport, and flux to groundwater. Another key challenge for reaching regulatory decisions results from an approach of making separate remedy decisions for vadose zone source term, DVZ, and groundwater operable units. Remediation of DVZ environments must be linked to cleanup goals for groundwater in order to protect human health and environmental resources. Within this construct, remediation efforts within the vadose zone are solely intended to mitigate sources of contamination and reduce future groundwater contamination, rather than meeting a specific concentration limit for contaminated sediments. This approach underscores the need for a systems-based approach to define and achieve risk-informed cleanup priorities, develop scientifically defensible remediation approaches, and achieve long-term protection of groundwater resources.

Systems-Based Assessment for Remediation Decision Support

It is critical to quantify and predict contaminant mass flux to groundwater in order to quantify the risk posed by vadose zone contaminants and select and implement appropriate, cost-effective remedies. Assessing risk and defining and achieving endpoints requires a sound technical basis that includes the following:

- A foundational framework that is cognizant of the risks and challenges of remediation and cleanup activities, while appropriately acknowledging technological limitations to achieving regulatory goals.
- Comprehensive understanding of the nature and magnitude of problems and associated assessment of which risks are most critical to remedy first based on:
 - spatial distribution of contaminants in source terms (i.e., stabilized/entombed facilities and tank residuals) and the underlying subsurface environment (e.g., vadose zone)
 - accounting for the hydrogeologic and biogeochemical controlling processes and opportunities to retard contaminant release from source terms to the point of compliance
- Defining mechanisms to address inherent uncertainties of environmental cleanup and respond to new information.
- Providing credible predictive models that depict natural subsurface dynamics, contaminant behavior, and remedial performance at spatial and temporal scales of importance and under the range of waste, geochemical, and hydrological conditions prominent to natural attenuation or engineered remediation necessary to make defensible remedial decisions and achieve cleanup goals.
- Monitoring systems that efficiently and effectively assess the long-term performance of remediation systems and behavior of residual contamination.

DOE is working with DOD and EPA to develop **a systems-based endpoint framework** to understand the risks associated with subsurface contamination and to develop and implement cost-effective and technically defensible remediation approaches (Lee et al. 2013). The endpoint framework consists of the following:

1. Defining the technical basis for remedial action
2. Performing a systems-based assessment of the remediation approach
3. Performing systems-based monitoring of the remediation approach (Bunn et al. 2012) and predicting long-term impacts of actions

The endpoint framework provides a structured approach to progress through data collection, remedy assessment, and remedy implementation phases to be consistent with the processes described by the Comprehensive Environmental Response, Compensation, and Liability Act and the Resources Conservation and Recovery Act. In addition to providing a technical foundation for addressing vadose zone contamination and achieving groundwater protection, the endpoint framework also enables effective communication with stakeholders and regulators, throughout the process, to garner support for cleanup and closure decisions.

Case Study: Hanford Site Central Plateau

Significant current and future impacts to water resources remain at Hanford from more than 800 waste disposal sites (e.g., ponds, ditches, cribs, trenches, reverse wells, and landfills) containing radioactive and other hazardous contaminants on the Hanford Central Plateau (DOE 2010). Past liquid discharges to the soil on the Central Plateau included large volume intentional discharges (~450 billion gallons) and relatively smaller volume, but higher concentration, unintentional releases from single-shell tank (SST) system components (~1 million gallons). These past releases have created multiple plumes of contaminated groundwater. The largest plumes from Central Plateau operations are tritium (95 km²) and ¹²⁹I (53 km²) from the final operating campaigns of the PUREX facility in the 200 East Area (DOE 2012). There is a 13 km² plume of carbon tetrachloride in the 200 West Area resulting from discharges from plutonium processing operations. Smaller groundwater plumes of ⁹⁹Tc, uranium, chromium, nitrate, and other contaminants also exist on the Central Plateau. However, a significant fraction of this inventory

remains within the vadose zone and may continue to contaminate groundwater. Understanding and managing the nature and extent of contaminant flux from the DVZ to groundwater is critical for Hanford's overall groundwater protection strategy.

Central Plateau: Principle DVZ Areas of Contamination

Figure 1 shows the principal DVZ regions of interest on the Hanford Central Plateau. These areas are known to retain large inventories of mobile contaminants of concern in the vadose zone, typically ^{99}Tc , uranium, and ^{129}I . Some of these waste sites have known contaminant flux impacting groundwater, other areas such as the BC Cribs and Trenches received a significant inventory of contaminants (in this case more than 400 Ci of ^{99}Tc) that has not yet reached groundwater. This is a key example of where critical investments from the Office of Soil and Groundwater Remediation, Richland Operation and the Office of Science have been integrated through the Deep Vadose Zone-Applied Field Research Initiative to provide critical new knowledge demonstrating the existence of technetium in multiple recalcitrant phases under the oxidative conditions. These findings are critical to understanding and defining the associated risks and relevant remediation endpoints for technetium in the vadose zone. Table 1 provides a summary of the inventories of key risk driving contaminants from the tank farms and other waste site. Table 2 presents delineates the principal DVZ areas of interest, the estimated inventory of key risk driving contaminants that were discharged, and recent observations of groundwater impacts in these regions (Corbin et al. 2005; Serne et al. 2009, 2010; DOE 2012). The groundwater impacts in the last column are an indication of risk for the contaminants of concern. Understanding the hydrogeologic, geochemical, and microbial factors that control the flux of contaminants at these waste sites will be essential to determining what, if any, action is needed to protect groundwater.

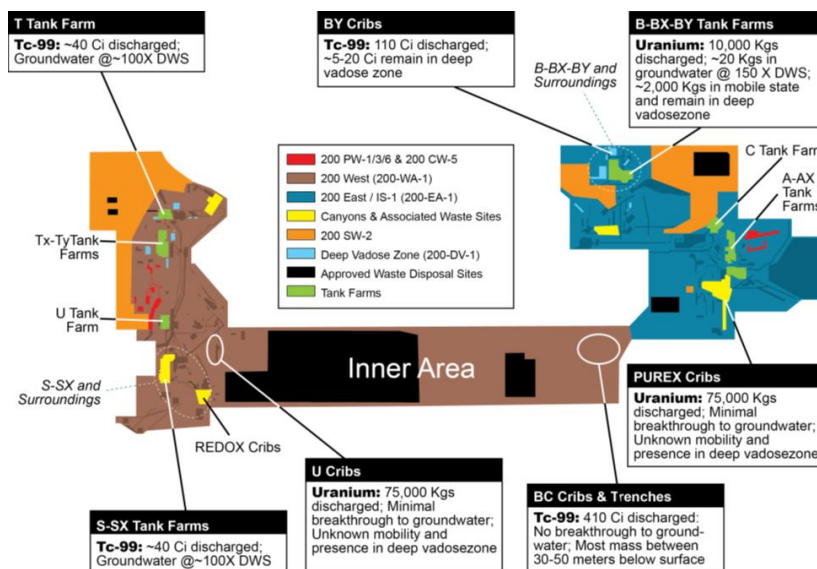


Figure 1. Inner Portion of Hanford's Central Plateau Showing Principal Deep Vadose Zone Regions of Interest

Table 1. Summary of Key Risk-Driving Contaminant Inventory Estimates for the Central Plateau Deep Vadose Zone Areas of Interest

DVZ Interest Area Category	Key Contaminant Inventory or Contaminant Mass		
	^{99}Tc	U	^{129}I
Tank Farm Waste Management Areas	101.4 Ci	10.2 MT	0.5 Ci
Soil Waste Sites, Cribs, and Trenches	571.3 Ci	58.7 MT	4.2 Ci

Table 2. Estimated Inventory of Key Risk-Driving Contaminants and Recent Observed Groundwater Impact for Deep Vadose Areas of Interest

DVZ Interest Area	Interest Area Description	Key Contaminant Inventory ^(a) or Contaminant Mass		Observed Groundwater Impact
				Max Times DWS ^(b)
200 East Area				
WMA A-AX	10 SSTs with 5 assumed leakers. ⁹⁹ Tc contaminant of concern.	⁹⁹ Tc	7.0 Ci	15
WMA B-BX-BY	40 SSTs with 20 assumed leakers. 10,000 kg uranium from BX-102 overfill event is the largest groundwater threat. Perched water 20 ft above aquifer contains uranium at 2,000 times the drinking water standard.	⁹⁹ Tc ¹²⁹ I U	7.2 Ci 0.01 Ci 10 MT	39 9.1 185
200-DV-1 B Complex	Includes 23 waste sites near the tank farms. BY Cribs discharged 110 Ci of ⁹⁹ Tc; conceptual model shows 5-20 Ci of ⁹⁹ Tc remaining in vadose zone.	⁹⁹ Tc ¹²⁹ I U	137 Ci 0.2 Ci 2 MT	111 9.1 185
WMA C	16 SSTs with 7 assumed leakers. ⁹⁹ Tc is the principal contaminant of concern for groundwater. Uncertain release inventory (e.g., 6-60 Ci ⁹⁹ Tc)	⁹⁹ Tc	5.5 Ci	29
200-EA-1, PUREX cribs	PUREX cribs and related waste sites (e.g., 216-A-4, 216-A-9, 219-A-19) received significant discharges of uranium containing waste. However, significant breakthrough to groundwater has not been detected.	⁹⁹ Tc ¹²⁹ I U	2.3 2.8 Ci 13 MT	<1 11 7
BC Cribs and Trenches (200-BC-1)	410 Ci of ⁹⁹ Tc discharged to the subsurface. More than half of total ⁹⁹ Tc discharge to soil in the Central Plateau. No breakthrough to groundwater.	⁹⁹ Tc ¹²⁹ I U	411 Ci 0.6 Ci 3.7 MT	0.0 0.0 0.1
200 West Area				
WMA S-SX	Principal threat to groundwater is from ⁹⁹ Tc. More than 30 Ci of ⁹⁹ Tc released from past tank leaks; 11 of 27 SSTs are assumed leakers.	⁹⁹ Tc ¹²⁹ I	32.0 Ci 0.5 Ci	209 9.5
S Complex (200-DV-1 waste sites)	Includes 4 waste sites within the general vicinity of the S and SX tank farms. Principal threats to groundwater are from ⁹⁹ Tc, nitrate, ¹²⁹ I, and chromium.	⁹⁹ Tc ¹²⁹ I	5.7 Ci 0.5 Ci	12 3
WMA T, TX-TY	Principal threat to groundwater is from ⁹⁹ Tc. Nearly 40 Ci of ⁹⁹ Tc released from past tank leaks from T farm; 20 of 40 SSTs are assumed leakers.	⁹⁹ Tc	46.3 Ci	206
200-DV-1 T Complex	Includes 17 waste sites within general vicinity of T, TX, and TY tank farms. Principal threats to groundwater are from ⁹⁹ Tc, nitrate, ¹²⁹ I, and chromium	⁹⁹ Tc ¹²⁹ I U	4.6 Ci 0.05 Ci 2.5 MT	206 71.7
WMA U	Principal threat to groundwater is from ⁹⁹ Tc. 4 of 16 tanks are assumed leakers. Inventory estimate for is very uncertain with possible large uranium inventory.	⁹⁹ Tc U	3.4 Ci 0.2 MT	3.8 0.01
200-WA-1 (U cribs and other waste sites)	Includes 129 waste sites in this consolidated geographic operable unit. Principal threats to groundwater are from U, ⁹⁹ Tc, nitrate, and chromium.	⁹⁹ Tc U	10.7 Ci 37 MT	3.5 42.7
Notes and Assumptions:				
^(a) Inventory estimates are from the <i>Hanford Soil Inventory Model, Rev. 1</i> (Corbin et al. 2005). Soil Inventory Model 2005 site groupings do not always match current operable unit scope definitions. Inventory estimates should be considered a general indication of relative inventories across the DVZ interest areas.				
^(b) Recent groundwater impacts report the maximum monitored value since January 2000. The source of these results is direct queries of the Hanford Environmental Information System database. In general, groundwater contaminant concentrations cannot be attributed to individual waste sites or tank releases. Reported groundwater concentrations reflect the aggregate impact from sources in an interest area				
Red denotes high impact/risk; blue denotes medium impact/risk; black denotes low impact/risk.				

Status of Central Plateau Remediation

Significant steps have been taken to characterize vadose zone contamination in the Central Plateau and implement actions to reduce the risk to groundwater, but much remains to be done. The Groundwater/Vadose Zone Integration Project roadmap (DOE 2000; Freshley et al. 2002) was used to engage the DOE national laboratories and universities to conduct science to improve the conceptual models and predictions of subsurface fate and transport to ensure that cleanup actions and decisions at Hanford protect the Columbia River. The strong integration between the DOE science community and the Hanford Site resulted in significant impact, including 1) a significant step change in the state of knowledge about the distribution of contaminants in the subsurface beneath Hanford's Central Plateau, including information on past tank leaks and an improved understanding of the distribution and mobility of contaminants in the subsurface and specifically the vadose zone; 2) new discoveries of vadose zone contaminant plumes (^{99}Tc at BC Cribs and Trenches); and 3) detailed information on the mobility of ^{137}Cs beneath the SX Tank leak. This knowledge has led to the understanding that significant contaminant inventories remain in the vadose zone and that these inventories pose long-term risk of contaminant flux to the underlying groundwater. However, sustained and concerted support for this effort has not continued at the level necessary to ensure continued progress and protection of regional and national (Columbia River) water resources. Moreover, recent discovery of new tank leaks raises further questions about environmental and human health risks from vadose zone source terms.

While some remediation decisions have been made, many sites are still early in the remedy decision process. Recent actions include the following:

- Interim measures have been implemented at the Hanford SST farms that divert drainage and surface water flow away from gravel-covered tank farms to reduce infiltration. Clean water discharges have been eliminated by cutting and capping leaking water lines (Kristofszski et al. 2007). Interim surface covers were installed at the T and TX tank farms.
- A Phase I investigation of the largest contamination releases from tank farms was completed in 2008 (DOE 2008a). The Phase II investigation is underway for C Tank Farm to support tank farm closure. Additional interim actions to mitigate the effects from previous tank leaks are being investigated for additional tank farms. A pore water extraction technology (derived from the BC Cribs desiccation test) will undergo a proof-of-principle field test at the SX Tank Farm in 2014.
- Vadose zone source reduction has not been applied other than for carbon tetrachloride via soil vapor extraction (SVE), which has diminished the vadose zone source to levels where termination of the SVE system is anticipated (Carroll et al. 2012). Perched water removal of a high-concentration uranium plume from the 241-BX-102 tank overflow event in 1951 has been continuing since fall 2010 (Truex et al. 2013a).
- Surface barriers and interim covers have been deployed at a few sites (DOE 1999; Field et al. 2010; Ward et al. 2011; Zhang et al. 2012) as interim actions with a projected reduction of contaminant transport from the vadose zone to the groundwater.
- A treatability test for soil desiccation has been performed at the BC Cribs and Trenches site (Truex et al. 2011; 2012a, b; 2013b). Laboratory and modeling evaluations of other treatment technologies have also been completed (Truex et al. 2010a, b; Szecsody et al. 2010a, b; Fayer et al. 2010) as elements of the DVZ treatability test plan (DOE 2008b).
- The feasibility of pore-water extraction to remove vadose zone contaminants is being evaluated based on laboratory and modeling investigations of this process (Truex et al. 2012c; Oostrom et al. 2011).

The complex array of waste sites, tank farms, former processing facilities, and burial grounds presents challenges for regulatory decisions that are generally made for subsets of waste sites or specific plumes of contaminated groundwater. The variety of commingled discharges that remain in the vadose zone makes it essential that remedy decisions for these contaminants be made with an understanding of their potential impact on groundwater, and remedy decisions need to be part of an integrated systems-approach for remediation and groundwater protection. A consistent, collaborative, and sustained effort that is based on a top-down science and technology roadmap and integrated into the baseline Hanford site efforts is necessary to advance progress toward cleanup goals for the remaining source terms and DVZ contamination and to protect regional and national water resources. The effort must be fundamentally based on a comprehensive, mass flux-based assessment applied to refine understanding of the coupled groundwater and vadose zone system and provide the technical basis for predictive assessment and remedial design and decision support based on the risks to human health and the environment. This collaborative approach will provide the scientific foundation for making defensible risk-informed remediation decisions that are acceptable to regulators, cost-effective, advance progress toward cleanup goals, and protect water resources.

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