Strategies for Immobilization of Deep Vadose Contaminants at the Hanford Central Plateau - 11503

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ABSTRACT

Deep vadose zone contamination poses some of the most difficult remediation challenges for the protection of groundwater at the Hanford Site in Richland, Washington. This paper describes processes and technologies being developed to use in the ongoing effort to remediate the contamination in the deep vadose zone at the Hanford Site.

INTRODUCTION

During the Hanford Site's plutonium production era, some 1.7 trillion L (450 billion gal) of liquid effluents were discharged into the vadose zone (subsurface) on the Site. Today, the Hanford Site's largest inventory of subsurface contamination lies beneath the Central Plateau, a 200 km^2 (75 mi^2) area containing approximately 800 waste sites and 900 facilities that operated to extract and purify plutonium. The byproducts from of this activity were effluents contaminated in varying degrees with chemicals and radionuclides.

The most dangerous waste is stored in 177 underground tanks. Some of this waste has leaked from these tanks into the vadose zone. Concentrated waste was discharged into engineered surface structures and allowed to percolate through the vadose zone as a means of disposal. This practice resulted in large-scale contamination to the vadose zone and groundwater underlying the Central Plateau. Some of this contamination remains in the vadose zone and has the potential to contaminate groundwater in the future. Liquid releases created large contaminated groundwater plumes covering nearly 300 km^2 (110 mi^2).

The deeper section of the vadose zone, herein termed "the deep vadose zone," poses unique problems for remediation by the very nature of the vadose zone itself. The physical structure, layering of sediments, subsurface emplacement of wastes, geochemical characteristics, and biogeochemical properties of the geologic framework affect subsurface contaminant movement and distribution. A lack of knowledge quantifying key processes affecting contaminant migration challenges the ability of scientists to reasonably predict the location and fate of contaminants under both natural or remediation conditions.

The geohydrologic contrast between sediment types, plus crosscutting and discontinuous geologic features such as stratigraphic facies changes, sediment orientation, fractures, and clastic dikes can impact lateral and/or vertical contaminant movement. The degree of complexity may be pronounced on a local scale, such as near a waste site or beneath a tank farm, but far less influential on a broader field scale.

Additionally, the heterogeneous nature of the Central Plateau vadose zone confounds detailed understanding of the distribution and extent of contamination. Because of the thickness of the vadose zone, thorough characterization using traditional sampling and analysis is extremely costly. Much of the contamination is too deep to apply traditional remediation techniques used for surface waste (removal actions) and alternative remediation approaches are necessary. These issues and others combine to make the deep vadose zone at Hanford one of the most challenging remediation problems within the DOE complex today. In situ treatment and surface barrier technologies offer promise for immobilizing contaminants in place, and in some cases, technology is being proposed that will remove some contaminants from the deep vadose with minimally intrusive techniques.

The treatability testing being performed by the CH2M HILL Plateau Remediation Company (CHPRC) as part of its Soil and Groundwater Remediation Project (S&GRP) demonstrates an innovative first-of-a-kind approach to addressing cleanup challenges within the deep vadose region. The Deep Vadose Zone (DVZ) project team began treatability testing by performing numerical modeling to address uncertainties associated with a technology and employing the technology in the deep vadose zone. Laboratory-scale testing of the technology that would ultimately be used to design the field-scale application was also performed. These studies were performed by the Pacific Northwest National Laboratory (PNNL) under contract to CHPRC, followed by development of a field pilot test for technologies maturing within the laboratory environment and demonstrating the capability to be effectively scaled up to perform field testing. The first of several field-scale tests is currently underway. The pilot test results will be based on performance and data collection for evaluation of the overall success of these technologies, and their capacity for scale up with consideration of cost and schedule should these treatment technologies be chosen for the Record of Decision (ROD) remedial actions.

TECHNOLOGIES CONSIDERED FOR DEEP VADOSE TREATMENTS

The *Deep Vadose Zone Treatability Test Plan* [1] cited previous studies and evaluated technologies for potential field application. Candidate technologies focused on in situ treatment and surface barrier technologies rather than traditional methods such as intrusive removal techniques, which were deemed too costly and burdensome given the extent and depth of the target contaminants being addressed. For this reason, certain candidate technologies were selected due to their maturity in development or considered for future testing based on additional evaluation, and laboratory modeling and testing to provide information sufficient to be able to scale the technology to a field application.

Technetium-99 Desiccation Testing

Soil desiccation is a potential in situ technology for the arid conditions and the thick vadose zone at the Hanford Site. Historically, 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 of additional water moving into the vadose zone and, thereby, slow contaminant movement. Desiccation could augment surface barriers by removing pore water to further slow the rate of contaminant movement to the groundwater.

Treatability efforts are targeting key technical uncertainties for applying soil desiccation. For instance, evaporative cooling effects of desiccation have been quantified in the laboratory. Although temperature decreases several degrees Celsius at the drying front, the effect on the overall rate of desiccation is small. A benefit of the cooling is that it provides a means to measure the location of the drying front based on temperature changes. Also, because desiccation removes pore water, the concentration of solutes, including contaminants, increases during desiccation. Experiments demonstrated this effect and concluded that the osmotic effects did not impact the desiccation rate. Diffusional movement of solutes due to concentration gradients under drying conditions occurred, but only to a limited extent.

The field test system consists of a nitrogen gas injection well, a soil-gas extraction well, and 25 monitoring locations within the test zone with over 700 in situ monitoring instruments. The six-month test is targeting desiccation of about 300 m^3 within a targeted interval 9 to 15 m below ground surface. [2]

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. 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. When ammonia gas at a concentration of 5 percent flows into vadose zone sediments, it rapidly partitions into the pore water. A portion of the ammonia dissociates and causes the pore water pH to increase to near pH 12. Under these conditions, desorption of ions and dissolution of aluminosilicates occurs. Subsequently, following cessation of ammonia injection, natural buffering processes within the soil and the loss of ammonia cause pore water pH to decline over time and precipitation of the ions in solution occurs. These precipitates coat and bind adsorbed uranium contamination, resulting in a significant reduction of mobile uranium contamination. In this process, uranium is not chemically reduced, so oxidation state does not affect treatment effectiveness. Laboratory experiments have shown this process to be robust in many Hanford sediments. Field testing is planned to further evaluate the process for application in the Hanford vadose zone.

Grouting and Soil Flushing Technologies

Grout injection is the subsurface placement of an encapsulating slurry mixture that, when cured or reacted, stabilizes or isolates the contaminant in a permanent matrix solid. Currently, PNNL is performing laboratory modeling and lab-scale testing for grout injection to evaluate the potential for deep vadose application for the treatment of contaminants, both chemical and radiological. This work will evaluate the injection properties of candidate materials with different viscosity, density, and composition for targeted Hanford Site contaminants.

Soil flushing is an approach that contacts targeted contamination in the vadose zone with a leaching solution that will mobilize contaminants with the intention of recovering them in the groundwater using pump-and-treat technologies. The application and distribution of the leaching solution in an unsaturated zone poses a significant challenge. PNNL also is performing laboratory modeling and lab-scale testing for soil flushing to evaluate the potential for deep vadose application to assess the distribution, location, and stratigraphic factors that control the distribution of vadose zone contaminants and movement of injected fluids.

DESICCATION TESTING (CHARACTERIZATION AND PILOT TESTING)

Desiccation Characterization Test

The overall desiccation field test was divided into two different field applications. First, a characterization test was performed to understand soil characteristics and lithology in the target test zone. Under this characterization test, well 299-E13-65 was drilled to enable the collection, measurement, and analysis of grab and core sediment samples to augment data previously collected from adjacent well 299-E13-62 (Fig. 1). Sediment sampling and laboratory analysis of borehole sediments were conducted to provide the vertical distribution of mobile contaminants, moisture content, and major solutes in the vadose zone pore water to support groundwater remediation related to final site cleanup. The total and water-leachable concentrations of key contaminants of concern (COCs), especially technetium-99 and uranium-238 in the sediments as a function of depth and their distances from inactive disposal facilities, were also collected to augment the conceptual site model for the BC Cribs and Trenches area. Water extracts taken from the sediments were used for electrical conductivity (EC), common cation and anion concentrations, pH, and alkalinity. Pore-water ionic strength was calculated based on the concentrations of major cations and anions. The total and water-leachable concentrations of key contaminants will be used to update contaminant-distribution conceptual models and to provide more data for improving baseline risk predictions and remedial alternative selections.

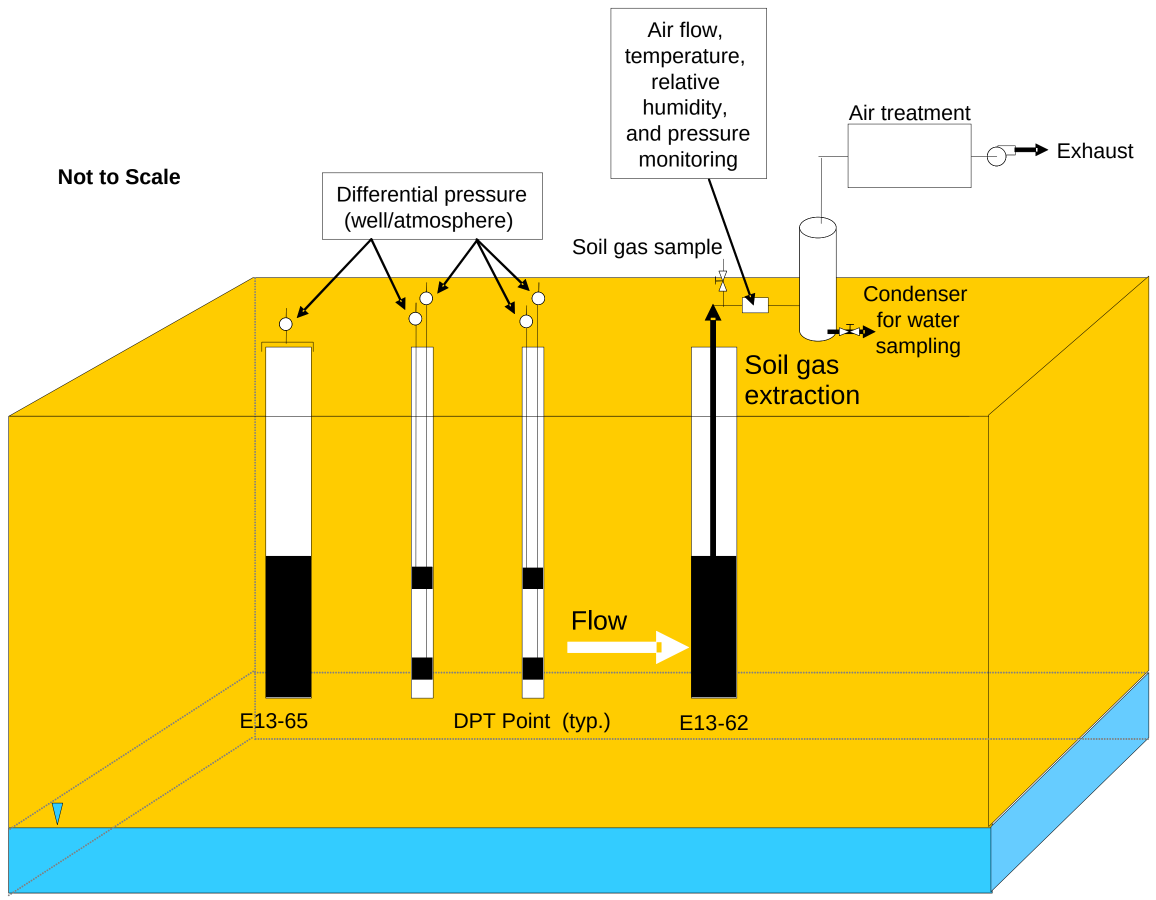


Fig. 1. Schematic of characterization phase test (Phase 1: Complete) configuration

The characterization test for this project was used to gather data on soil moisture content, temperature in the desiccation region during operation, and sediment air permeability. Additionally, in-ground sensors were evaluated to determine their applicability for the pilot test as well as other future deep vadose activities. Many of these sensors were installed for the pilot test per the recommendations of an independent expert review panel that was contracted to overview the project plan, monitoring and equipment operation and provide the project team with guidance based on their collective expertise. The expert panel was contracted by the DVZ project as an outgrowth of the 2004 request by the regulators to

DOE to identify a path forward to identify tools to address technetium-99 contamination in the deep vadose zone. Panel members provided expertise in vadose zone transport, infiltration control, hydrology, geochemistry, instrumentation, and geology, and came from academia, government laboratories, industry, and consulting.

The DVZ project team has found the panel recommendations to be invaluable. The most significant comment ultimately changed the basic design of the pilot test. This adaptation was to operate a single extraction well associated with the injection well (a dipole system) and resulted in reducing the number of monitoring locations required, eliminating three of the four extraction wells originally planned. This also simplified the test data evaluation. The characterization test was performed during June and July of 2009 and yielded information on soil gas transmissivity and soil permeability that helped optimize the overall project design and monitoring for the Desiccation Pilot Test.

Desiccation Pilot Test

A number of instruments were emplaced in situ for monitoring the desiccation test: (1) thermistors, (2) heat dissipation units (HDU), (3) thermocouple psychrometers (TCP), (4) humidity probes, and (5) dual-probe heat pulse (DPHP) sensors. These instrument clusters were installed in 10 boreholes, resulting in excess of 700 underground instruments to monitor the Desiccation Pilot Test. The signals from these instruments will be related to the moisture content of the sediment, most particularly for the HDU, TCP, and DPHP instruments. In addition to these instruments, ground-penetrating radar -- cross-hole radar (GPR), neutron moisture logging, and electrical resistance tomography (ERT) are being used to monitor sediment moisture content change during desiccation. Prior to startup of the field test, baseline measurements using GPR, logging, and ERT techniques were tested. For ERT, this field information was coupled with laboratory data that evaluates the change in conductance versus moisture content as the sediment is dried. Soil gas and condensate samples are being gathered during the performance period of the Desiccation Pilot Test in accordance with the sample analysis plan (SAP) [3].

The pilot phase of desiccation will operate continuously for a period of nominally six months. Following the six-month performance period, a periodic monitoring plan will be established for nominally five years to examine horizontal and vertical rebound of moisture in the desiccated region.

This project uses standard commercial air movers and nitrogen gas delivery systems. The nitrogen gas injection well and the soil gas exhaust/removal well are monitored for temperature, pressure, flow and moisture (see Fig. 2). A series of boreholes were established as part of baseline preparation for operation of the Desiccation Pilot Test and these were also monitored for temperature, moisture, and pressure differential between monitoring locations. The exhaust train also monitors temperature, humidity, and flow rate. All the in-ground related sensors and instruments and above-ground equipment/systems monitoring are monitored via data loggers in the field and downloaded to a computer in the operations/instrument trailer.

The design injection rate for the Desiccation Pilot Test is nominally 300 standard cubic feet per minute (scfm) of nitrogen gas. The nitrogen gas injection unit injects conditioned nitrogen gas into well 299-E13-62 at a temperature of 68 degrees Fahrenheit plus or minus 2 degrees. The extraction well is located approximately 40 ft away from the injection well and operates at 100 scfm.

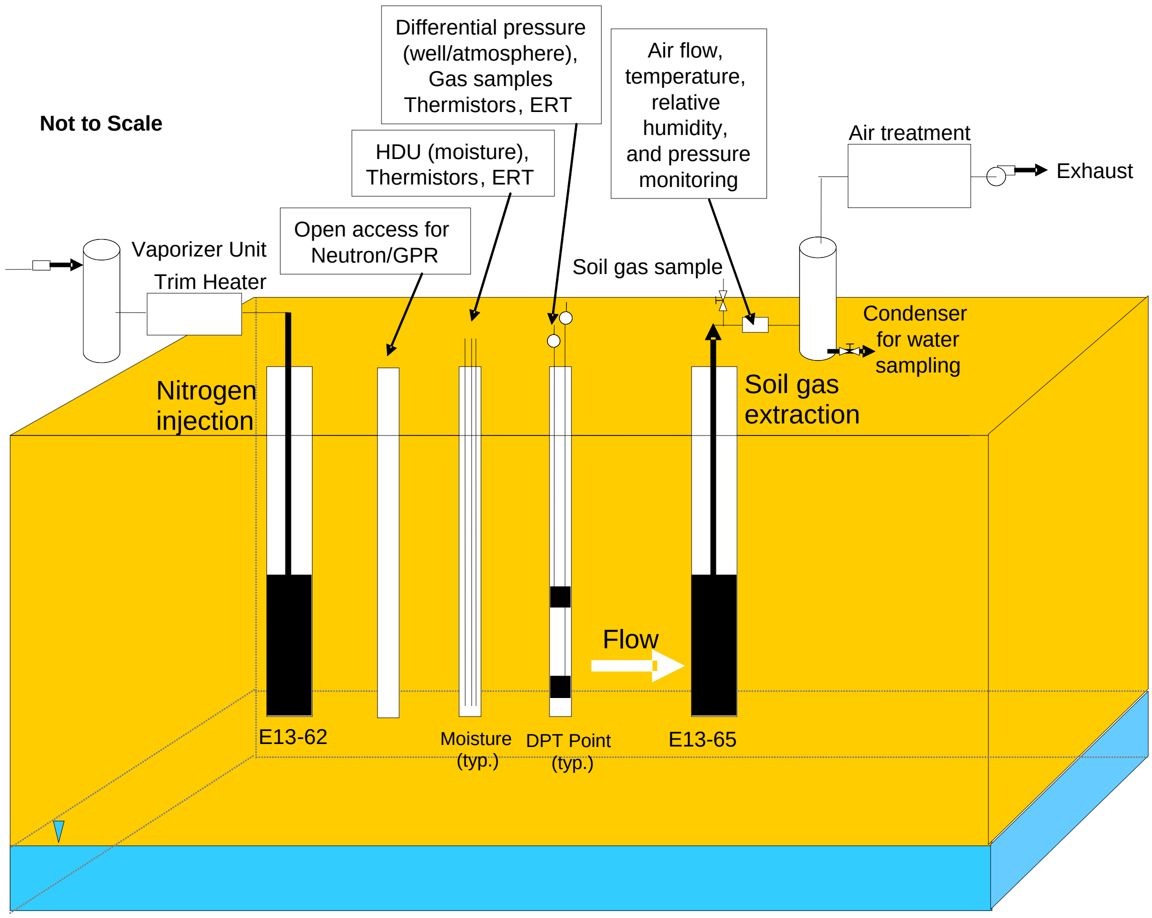


Fig. 1. Schematic of pilot test configuration

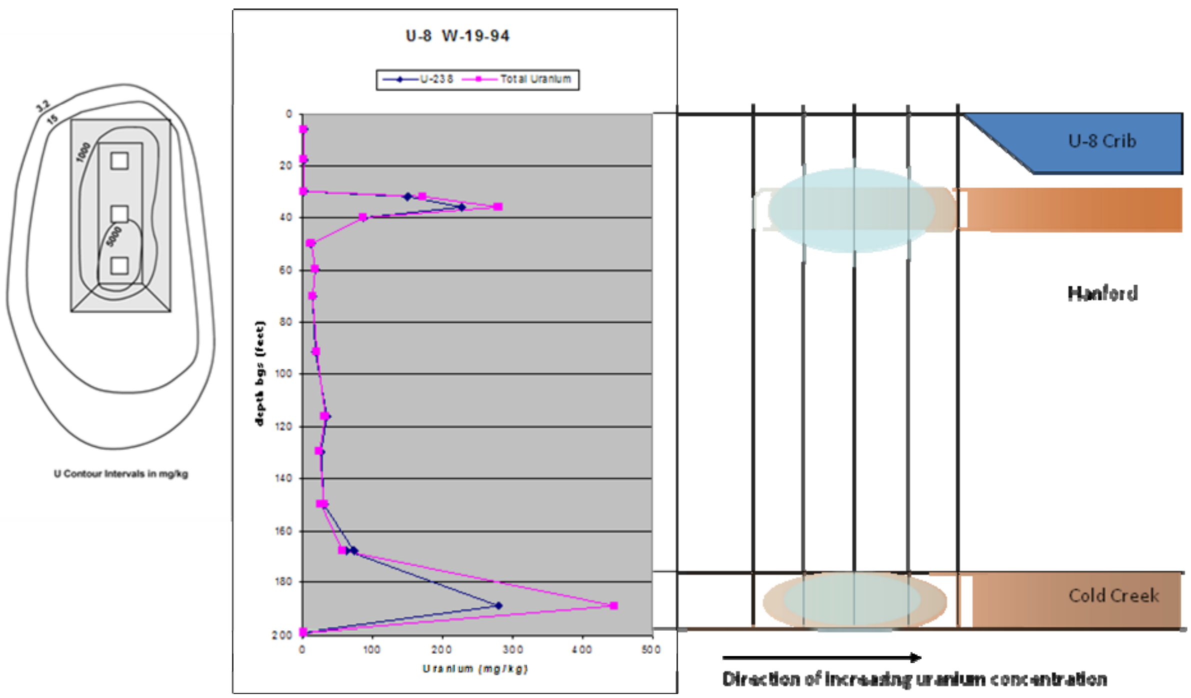
Both the extraction and injection wells are screened from -30 to -50 ft bgs to establish air flow between these two wells. The exhaust system also uses a HEPA filter and record sampler to monitor the system as part of the air emissions plan.

Moisture (water) is the only constituent requiring removal and is the focus of this project. Although there is no requirement for the removal of co-contaminants, monitoring and sampling is required in accordance with the SAP [3] and was also established in the air emissions plan [4], both of which were regulatory documents approved by the U.S. Environmental Protection Agency (EPA). The Desiccation Pilot Test was officially started on November 9, 2010. The desiccation test site is shown in Fig. 3.

Fig. 3. Desiccation test site

URANIUM SEQUESTRATION TESTING

Mobile uranium contamination in the deep vadose is also a unique challenge. Differing from desiccation where the overall objective is to alter the water flux to slow or halt the advance of contaminants before they migrate to the groundwater, uranium sequestration techniques are aimed at immobilizing mobile species of uranium by locking them up with a precipitated matrix resistant to resolubilization. In 2009, PNNL was tasked with evaluating differing in situ gaseous reduction compounds in addition to compounds needed to achieve multi-step geochemical manipulation. The results of the 2009 study [5] revealed that ammonia gas injection provided the most significant decrease in changing mobile uranium to non-mobile species based on sequential extraction techniques that were applied to all the test agents. Based on this study, ammonia injection was been selected as the reactive gas to perform the field test. The injection well for gas delivery to the target zone and the associated monitoring boreholes are scheduled to be drilled during spring of 2011. Mobilization of equipment and monitoring systems will follow, and



when complete, a field sequestration pilot test will follow in 2012 (Fig. 4).

Fig. 4. Schematic of the uranium sequestration test location and proposed test regions

Initially, five boreholes will be drilled and sampled. Based on evaluation of sediment analyses, one of these five will serve as the injection well and the others will serve as monitoring boreholes. Up to six additional monitoring boreholes will be drilled following selection of the injection test well. In parallel, the soil samples removed will be tested by PNNL using the same ammonia sequestration techniques to be deployed in the field to assess and confirm adequacy and feasibility of sequestration of the test bed material. Additionally, during the laboratory testing, instrumentation will be imbedded in the test specimens to evaluate effectiveness of instruments used to monitor the test. Final instrumentation selection for placement of instrument cluster down the instrumentation boreholes at the test site will be selected based on the outcome of the laboratory analysis.

Once the test is initiated, an injection of 5 percent ammonia gas and 95 percent nitrogen gas will be injected into the target locations as depicted in Fig.s 1, 2, and 4. At the screened intervals in the injection well, the ammonia will partition into the sediment pore water and establish a high pH front (nominally a pH of 12) as it advances. This high pH front will continue to advance as long as the injection of this gas mixture continues. As this high pH front advances, it will dissolve some of the aluminosilicates within these soil regions. Sensors in the instrumented boreholes will monitor and evaluate passage of these reaction fronts. Evaluation of data will allow management to determine when the areas targeted for reaction have been treated. Once this happens, the injection of the ammonia will be shut down and the project team will wait for approximately three months to allow for buffering processes within the test region to return to a near neutral pH. At that time, the dissolved aluminosilicates will have precipitated to form a rind over much of the previously mobile uranium, thus rendering it immobile.

PORE WATER EXTRACTION

The Pore Water Extraction (PWX) phenomenon was encountered at the BC Cribs waste site by CHPRC personnel during characterization testing that was performed as part of the Deep Vadose Desiccation Characterization Test. During the performance of the Desiccation Characterization Test, extraction flows rates were significantly high to understand the soil gas transmissivity characteristics. Rates were obtained out of the extraction well in excess of 500 scfm. Corresponding vacuum pressures in excess of 80 in. of water column were achieved. The project had expected water removal from the deep vadose via an evaporative model that left the contaminants behind; however, due to these substantially high flow rates and pressures, pore water was actually stripped out of the soil through the screened section of the extraction well, and the speed of the airstream being extracted (calculated to be around 65 mph) had the effect of entraining these pore water droplets and arriving in our collection tank for sampling. This resulted in very high levels of technetium-99 being removed from the vadose. The attractiveness of this phenomenon was that instead of leaving the contaminants locked in place using technologies such as desiccation, we could remove significant quantities of contaminants with minimal intrusion into the subsurface. In effect, this was closer to a remediation technique rather than a “lock it up and leave it” approach.

Subsequently, PNNL was requested by DOE to perform modeling to determine conditions required and the area of influence that could be achieved with a vacuum applied for the purpose of removing contaminants from the deep vadose zone. Based on these modeling results provided by PNNL, it became evident that PWX can occur in the Hanford formation (a sandy/silt loam region that can be a few hundred feet thick) under specific conditions, but that application of the PWX process in high moisture content regions of the Cold Creek unit (a high carbonate tight silt lens region underneath the Hanford formation and ranges in thickness from several feet to tens of feet) would produce the highest yields.

Using the results of the modeling study, the PWX field test site selection was broken into two independent tests: One for the Cold Creek unit to test the optimal conditions for PWX and the other for the Hanford formation to validate PWX occurrence and potential application under sub-optimal conditions. For this reason, the site selection process reviewed sites that were previously well characterized to ensure the soil characteristics within each of these regions were suitable to meet the necessary conditions to achieve the pore water extraction, inclusive of well design and finishing. The selection process reviewed both the positive and negative issues associated with each site reviewed by the selection team. The resulting conclusions from this site selection process was to recommend a field test adjacent the B tank farms in the Cold Creek unit (well 299-E33-344) within the perched water region to remove mobile uranium and technetium-99 present in the pore water. The BC Cribs and Trenches site within the cribs discharge portion is recommended as the field test site in the Hanford formation to evaluate pore water and technetium-99 removal from high moisture content Hanford formation silt lenses. This site is adjacent to the currently operating Desiccation Pilot Test. The PWX test will use the same infrastructure (power, data collection systems, and extraction and monitoring equipment) currently operating for the Desiccation Pilot Test after it concludes in the spring of 2011.

SURFACE BARRIERS

Surface barriers are one of the remediation options available for isolating deep vadose zone contaminants to protect groundwater. The effectiveness of surface barriers is highly dependent on the complex interaction of site conditions, surface barrier design and performance features, and vadose zone contaminant conditions. PNNL was contracted by CHPRC to evaluate surface barriers and provide a strategy for evaluating the effectiveness of surface barriers for site-specific deep vadose zone remediation. This strategy provides a technically defensible approach to determine the effective depth to which a surface barrier can effectively isolate contaminants in the vadose at a specific site as a function of subsurface properties, contaminant distribution, barrier design, and infiltration control performance. The strategy also provides an assessment of additional data and information needs with respect to surface barrier performance for deep vadose zone applications. The strategy addresses the linkage between surface barriers and deep vadose zone in situ remediation activities, monitoring issues, and emerging science, technology, and regulatory objectives.

The strategy is focused on deep vadose zone contamination and the methods needed to determine the impact to groundwater from vadose zone contaminants. Utilizing a surface barrier strategy would be most likely be performed in tandem a deep vadose treatment technology such as desiccation or uranium sequestration as a defense in depth approach to protect groundwater from deep vadose contaminants.

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