

## **Deep Vadose Zone Characterization at the Hanford Site: Accomplishments from the Last Ten Years**

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### **ABSTRACT**

The overall goal of the Tank Farm Vadose Zone Project, led by CH2M HILL Hanford Group, Inc., is to define risks from past and future single-shell tank farm activities at Hanford. To meet this goal, CH2M HILL Hanford Group, Inc. tasked scientists from Pacific Northwest National Laboratory (PNNL) to perform detailed analyses on vadose zone sediments collected within/adjacent to the twelve single-shell tank farms contained within Hanford's Central Plateau region. This work has been performed under the Resource Conservation and Recovery Act (RCRA) Corrective Action Program and is associated with the Hanford Federal Facility Agreement and Consent Order. While there are many facets to the laboratory studies employed by PNNL, the four primary objectives of this work are to: identify the type and quantity of contamination present, understand the physical processes that affect the transport of contaminants in the vadose zone sediments, when practical, identify the source(s) of the contamination found in the sediment samples, and when practical, determine if a link can be made between the vadose zone contamination observed and any known groundwater contaminants in the vicinity. Since its inception in 1997, PNNL's Vadose Zone Characterization Project has evolved to better meet these four key objectives. The single-largest adaptation of the Vadose Zone Characterization Project over its ten years of operation was the advent of a tiered sample analysis approach. Use of a tiered approach allows resources to be focused on those samples/tests that provide the largest amount of scientific information to best meet the four key project objectives within the budget available. Another significant, but more recent, adaptation has been the implementation of a rapid turnaround characterization process in which sediment samples are analyzed in near real-time to aid drilling activities within the tank farms. This paper highlights details of the characterization activities performed as well as significant findings from the last ten years.

### **INTRODUCTION**

The Hanford Site, located in southeastern Washington State, was once home to Cold War weapons grade plutonium production. Over the last fifteen years, the Site's function has shifted from production and operation to remediation and decontamination. One legacy still remaining at the Hanford Site are wastes from plutonium processing activities, which are stored in 177 massive underground tanks in eighteen tank farms. Hanford's tank farms are further segregated into twelve older single-shell tank farms (built in the 1940s through the early 1960s) and six newer double-shell tank farms (constructed from 1968 through 1986). The single-shell tank farms have been further grouped into seven waste management areas (WMAs)

Wastes initially disposed to the single-shell tanks were thermally hot because of radioactive decay, and many exhibited extreme chemical character in terms of pH, salinity, and radionuclide concentration. At present, more than 60 of the 149 single-shell tanks are suspected to have released over 1.9 million L of tank waste to the vadose zone, with most leak events occurring between 1950 and 1975 [1]. Tank waste contaminants are present in the groundwater at all seven of the single-shell tank WMAs (i.e., A/AX, B/BX/BY, C, S/SX, T, TX/TY, and U). Boreholes have been placed through the largest known vadose zone plumes so that extensive characterization of the contaminated sediments could be performed. The four primary objectives of these efforts were:

- Identify the type and quantity of contamination present. This is accomplished in part by developing conceptual representations for the lateral location and vertical extent of specific contaminants (e.g., uranium, Tc-99, Cs-137, and Co-60) in the sediments.
- Understand the physical processes that affect the transport of contaminants in the vadose zone sediments. The vadose zone is the area located vertically between the land surface and water table consisting of soil pores that contain a mixture of air and water.
- When practical, identify the source(s) of the contamination found in the sediment samples.
- When practical, determine if a link can be made between the vadose zone contamination observed and any known groundwater contaminants in the vicinity.

These four objectives were established to obtain the data required to assess the long-term environmental impact associated with residual vadose zone contamination. Before an accurate assessment can be made, it is imperative to understand what type of contamination is present as well as how much of it resides in the vadose zone. Also important is knowledge of the location of the contamination within the vadose zone, as different remediation options are available for shallow versus deep contamination. Different physical and chemical processes (e.g., diffusion, sorption, and precipitation) can affect the mobility of contaminants dramatically; therefore, knowledge of the existing state of the contamination can provide valuable insight when estimating its current and future mobility in the vadose zone.

## **CHARACTERIZATION APPROACH**

### **Tiered Analyses**

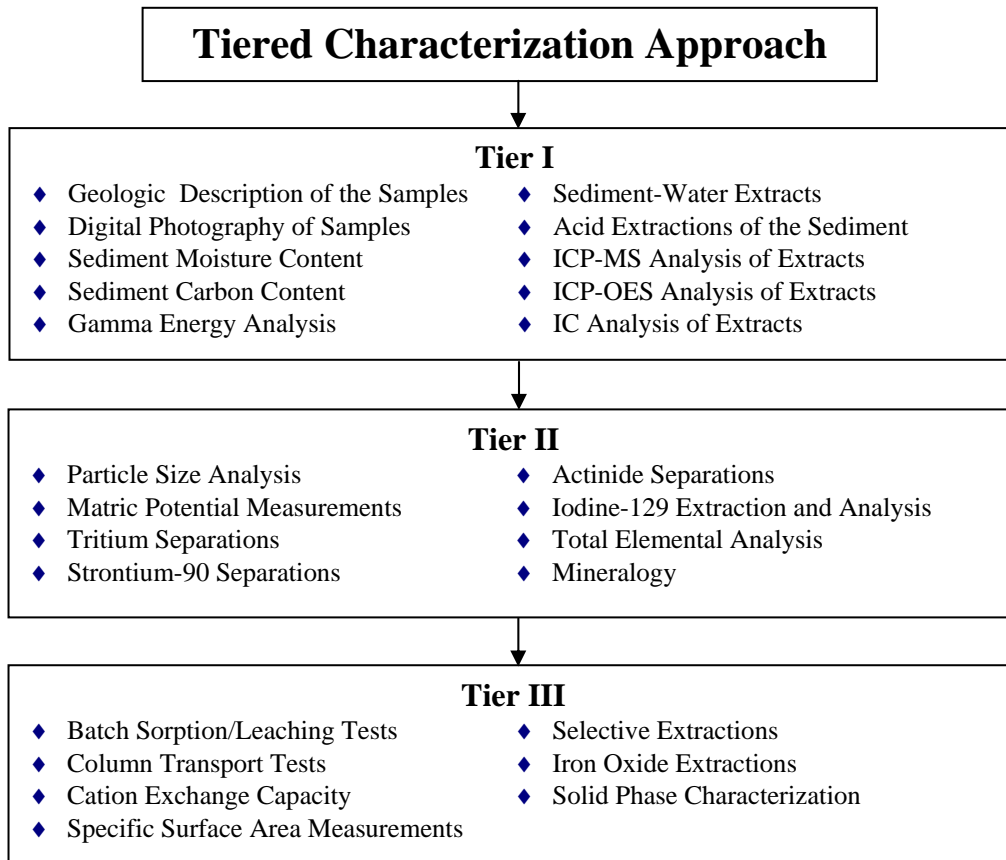
Since its inception in 1998, Pacific Northwest National Laboratory's Vadose Zone Characterization Project has evolved to better meet its four key research objectives. The single largest adaptation of the project during that period was the advent of a three-tiered approach to sample analysis. The basic premise of a tiered approach is to acquire the largest amount of scientific information, through an incremental process, that can be used to best meet project objectives while conserving resources and limiting risk to workers. Another factor that must be considered when working with sediment samples is that sample mass often is limited as a function of the field techniques used to acquire the sediment from beneath the tank farms. Depending upon the drilling or coring technique [2,3], sediment samples range in mass from a few hundred grams to several kilograms. When only a few hundred grams of sample material are available, it is increasingly important to judiciously select the order in which the analyses will be performed.

In the tiered analysis approach (Figure 1), the first tier of tests (Tier I) includes those analyses that:

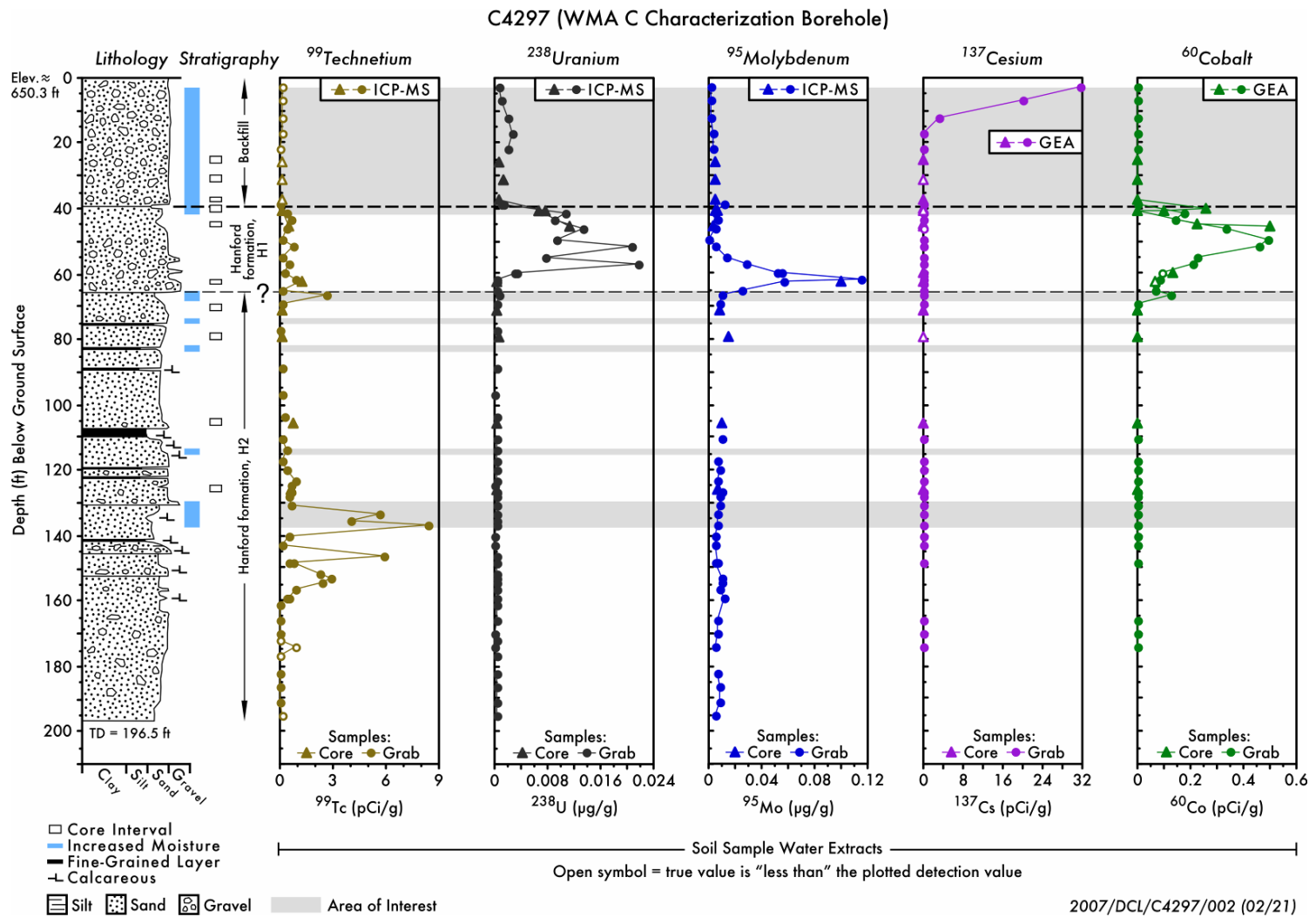
- Provide information paramount to identifying contaminants present in the samples.
- Help determine the basic chemical properties of the sediments.

Additionally, several of the Tier I analyses are nondestructive (i.e., they do not alter the properties of the samples), so these sediments can be used again in other tests. The concentration profiles of contaminants measured in the sediment as a function of sample depth and sediment geology are one of the most important results from Tier I testing. These concentration profiles provide a measure of the extent of migration and relative mobility of each detected contaminant along the borehole length. As an example, Figure 2 shows the concentration profiles of selected contaminants measured in sediment–water extracts from sediments from borehole C4297, which was emplaced adjacent to tank 241-C-105.

Upon completion of the Tier I tests, the data are evaluated; the Tank Farm Vadose Zone Program then decides whether there is a need to extend the studies to include Tier II and III testing. Tier II testing is focused on better resolving the type and extent of contamination present in the samples. Tests employed during Tier II characterization activities include specialized sediment chemical-dissolution (i.e., extraction) and analytical techniques, in addition to tests focused on identifying differences in the physical properties of the sediments. The final tier of testing, Tier III, is reserved for those analyses and experiments that will provide detailed information on the type of contamination present (e.g., its oxidation state or its physical/chemical state within the sample) as well as information on the transport properties of the contaminants. More detailed information about the specific tests performed during each tier of analysis is included in several reports [2-13] and reference procedures [14-20].



**Figure 1. Tiered approach to sample analysis and characterization.**



**Figure 2. Tier I characterization data from borehole C4297 emplaced adjacent to tank 241-C-105 (from Brown et al. 2006).**

### **Rapid Turnaround Analysis**

Another significant but more recent adaptation of the Vadose Zone Characterization Project has been the implementation of a rapid turnaround characterization process, in which sediment samples are analyzed in near real time to aid drilling activities within the tank farms. One of the primary objectives of a borehole characterization effort is to define the vertical extent of contamination at a specific location. The ability to accomplish this can be complicated by several factors. For example, the subsurface could become impenetrable by the drill rig—i.e., cause the drill rig to *meet refusal*. A few sediment layers at the Hanford Site are very difficult to penetrate via push techniques; these include the Cold Creek Unit (Figure 3) and basalts from the Columbia River Basalt Group [21]. Although it may be difficult for a drill rig to penetrate these layers, these sediments do not necessarily prevent the downward migration of contaminants. In other rare situations, the drilling activity could be halted before the maximum depth of contamination is reached. To ensure capture of the maximum depth of contamination within specific drilling activities, the boreholes often are emplaced much deeper than necessary. While this practice is necessary to most accurately define the contaminant plume(s) at specific locations, it leads to additional safety risks and chemical/radiological exposures to tank farm workers due to the additional time spent in the farms retrieving samples.

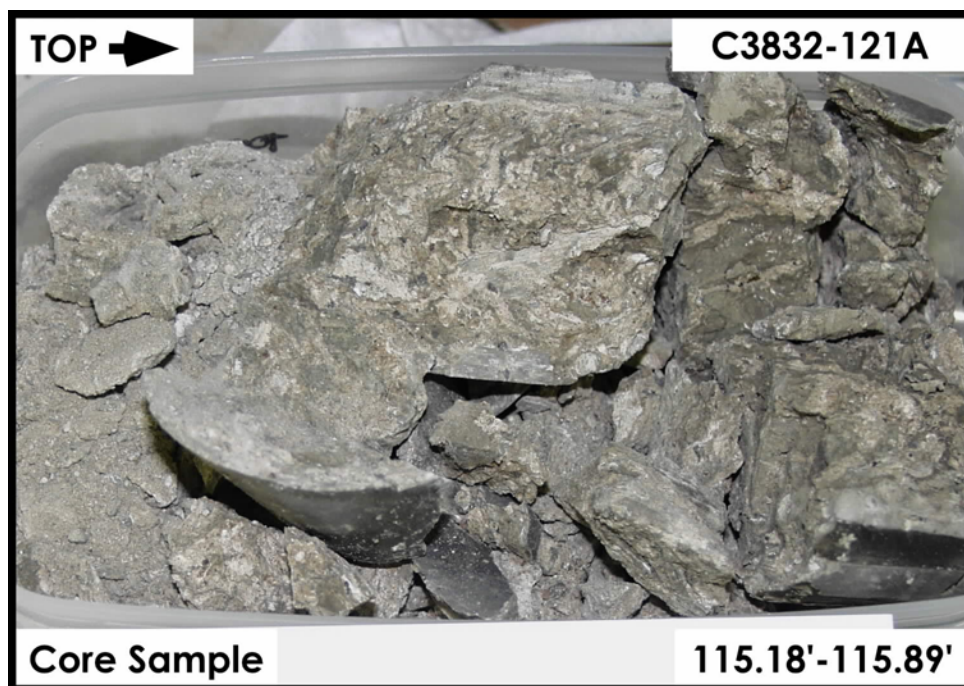
To minimize worker exposure while still capturing the maximum extent of contamination, the PNNL Vadose Zone Characterization Project has developed a process in which vadose zone sediment samples can be processed for key contaminants of concern in near real time. Previous research has shown that two contaminants, nitrate (NO<sub>3</sub>) and Tc-99, are typically found at the leading edge of waste plumes [6-8]. Beginning with samples that have been collected from approximately 150 feet below ground surface, samples every 10 feet are processed via water-dissolution extraction and the resulting leachates are rapidly analyzed for nitrate and Tc-99. Results from these extractions are made available to the Tank Farm Vadose Project contractor within 24 hours of receipt of the sediment by the laboratory. If either (or both) of the contaminants are measured at quantifiable concentrations in the extracts, borehole drilling continues. Once neither contaminant is detected in the sediment, another 10 to 20 feet of borehole will be drilled; the sediment will be extracted, and leachates from the water-dissolution extractions will be analyzed for nitrate and Tc-99. Should subsequent analyses of these samples fail to detect either of these two contaminants, drilling activities can be halted. This staggered drilling/characterization approach was used for the characterization effort conducted at the 241-C Tank Farm [2] and now serves as a model for future borehole drilling campaigns.

## **SIGNIFICANT FINDINGS FROM THE CHARACTERIZATION ACTIVITIES**

### **Influence of Fine-Grained Lenses**

Results from the Vadose Zone Characterization Project have revealed that mobile contaminants, such as Tc-99 and nitrate, migrate much differently in the subsurface than was previously believed. Conventional thinking was that once mobile contaminants entered the subsurface, they migrated in a nearly vertical path through the soil column. However, the extensive amount of work performed by the Vadose Zone Characterization Project has shown that the geologic layering of the vadose zone sediments has an important impact on the direction and rate of migration of the waste liquids and dissolved contaminants. The detailed sampling and

characterization of both contaminated and uncontaminated vadose zone sediments has shown the importance of thin lenses (often a few millimeters to a few centimeters in thickness; Figure 4) of fine-grained sediments in the various Hanford formation sediment types. These fine-grained lenses have been shown to cause significant horizontal spreading of leaked fluids within the vadose zone. For example, characterization work performed on sediments collected from two boreholes emplaced in the 241-T Tank Farm has shown that a large amount of horizontal spreading (in excess of 25 meters) occurred when the tank waste solution intercepted the Cold Creek unit [11]. As a result of findings such as these, characterization campaigns now target fine-grained lenses as optimal sampling points to locate contaminants.



**Figure 3. Core sample from lower cold creek unit.**

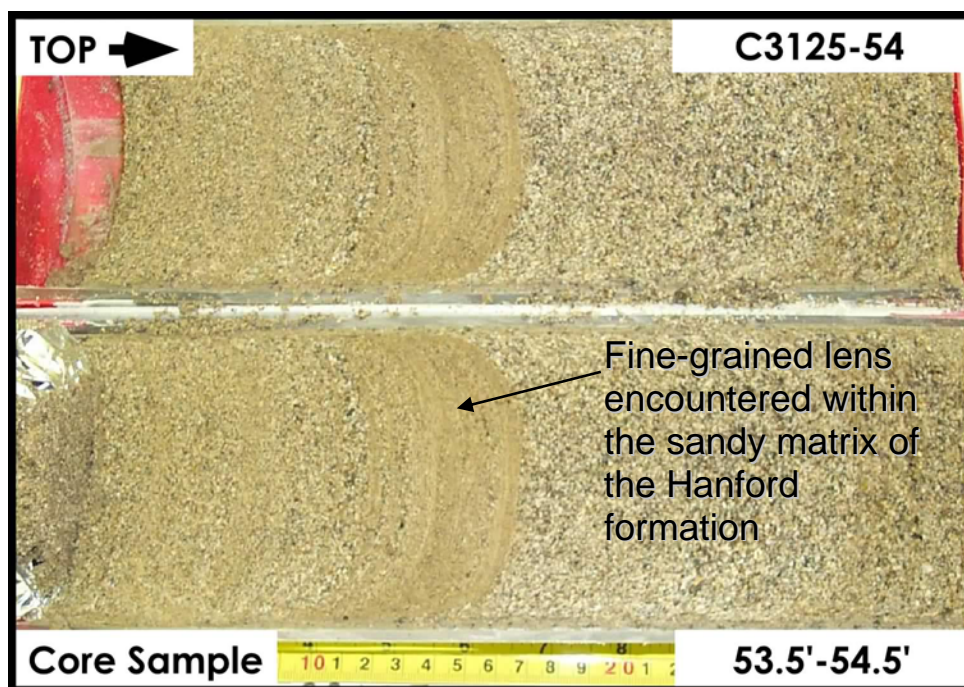
### **Effect of High Solution pH**

Another discovery from the laboratory studies has been the observation that the zone of caustic attack, due to the high pH tank waste, can be determined by measuring the pH of the soil. Given that tank waste is generally considered caustic (in excess of 1M free hydroxide), it is quite common to find elevated soil pHs (between 8.5 and 10) in the vadose zone adjacent to the point of waste release. However, natural minerals present in the sediment act to buffer the elevated pH tank waste; as such, the total area exhibiting elevated soil pH is always considerably smaller than the footprint that has been contaminated by more mobile tank waste constituents, such as nitrate or Tc-99. Because of this, soil pH has become one of the key parameters measured when looking for waste discharge locations in the vadose zone.

In addition to increasing the soil pH near the point of waste discharge, numerous laboratory studies and a few field samples have shown that highly caustic tank liquors dissolve some of the



native minerals in the Hanford sediments. This facilitates the precipitation of secondary minerals (zeolites, feldspathoids, and clays) that in general, have greater capacities and tendencies to sequester trace contaminants, such as the radioactive fission products and actinides present in tank liquors, than the majority of the native minerals. The secondary minerals also tend to be more voluminous (take up more space on a mole basis than the native primary minerals), such that bulk porosity is reduced in the zone of active reprecipitation. Both the higher capacity for sequestering contaminants and the changes in porosity have led to changes in our conceptual models of the fate of fluid flow and contaminant migration. The caustic tank fluids also dissolve ferrous iron bearing minerals, which leads to the release of the reducing agent ferrous iron. The released ferrous iron can reduce redox sensitive contaminants, such as Cr(VI), pertechnetate ( $\text{TcO}_4^-$ ), and U(VI). All three of these contaminants are less mobile in their reduced states than in their oxidized states. However, field evidence below single-shell tanks that have leaked in the past show that only Cr(VI) migration at the SX Tank Farm has been measurably reduced in the near field sediments below tanks SX-108 and SX-109. Either the impacted zones are quite small or the reduction of other key contaminants has been quickly reversed by oxygen entering through diffusion through the partially saturated pore space. Unlike Tc-99 and uranium, once chromium has been reduced to Cr(III), it is not readily re-oxidized by atmospheric oxygen.



**Figure 4. Split sediment core taken from the Hanford formation containing a thin (approximately 3 centimeters wide) lens of fine-grained sediment.**



### **Ion Exchange**

Based upon a large number of sediment characterization studies of borehole samples collected within the single-shell tank farms, adsorption to mineral surfaces is typically the single-most important geochemical process impacting the transport of contaminants in the vadose zone at Hanford. Adsorption occurs as atoms, ions, and complexes (multi-atom molecules or ions) exert forces on each other at the solid-water interface. Calcium and magnesium are the two dominant cations present in Hanford sediments. When waste solutions containing high concentrations of dissolved sodium (in excess of 4M) contact the sediment, the sodium exchanges with the calcium and magnesium, creating an ion exchange front. In this scenario, a front or wall of naturally present calcium and magnesium that have been removed from the sediment exchange sites precede the tank waste plume containing the elevated sodium. Although the ion exchange front will not necessarily define the total vertical impact of tank waste contamination, it can be used to target the most appropriate depths to look for mobile contaminants.

Additionally, this ion exchange front can significantly impact the mobility of Cs-137 in vadose zone sediments. Generally, Cs-137 is quite immobile in the vadose zone; typical partition coefficients, or  $K_{ds}$ , for Cs-137 are in excess of 1000 ml/g [22]. However, when solutions containing greater than 10M dissolved sodium are present,  $K_{ds}$  of less than 10 ml/g for Cs-137 are not uncommon. This dramatic increase in contaminant mobility is directly proportional to the concentration of dissolved ions in the porewater. The large amount of sodium leaked from the tanks displaces the cesium from the sediment sites that normally would retard cesium movement. Given the large amounts of sodium, the relatively small amount of cesium is thus free to move with the porewater and travel significant distances. However, as the concentration of sodium is reduced in the subsurface by ion exchange retention on the natural sediment cation exchange sites, the exchange sites in the sediments farther away from the high sodium pulse become available and cesium once again binds tightly with the sediments.

Ion exchange also serves as the dominant attenuation mechanism for Sr-90 in Hanford sediments. However, unlike Cs-137, in which the contaminant is pushed in a high dissolved salt waste plume, Sr-90 mobility is controlled by the exchange of contaminant strontium (Sr-90) with stable strontium present in the native sediments. Further, ion exchange occurring within basaltic lithic fragment interiors has been found to strongly limit the desorption and further migration of contaminant strontium leaked beneath single-shell tank 241-B-110 [6] in an otherwise extremely coarse-textured Hanford sediment.

### **Isotope Ratio Analysis**

The Vadose Zone Characterization Project continues to evolve to better meet the needs of the Tank Farm Vadose Zone Program. While a significant amount of work has been performed to characterize contamination found in the vadose zone within and around the single-shell tank farms, little success has been achieved at linking contamination found in the groundwater with that found in the vadose zone due to the complex flow paths for pore water and multiple sources for contamination. As mentioned previously, fluid releases from single-shell tanks and their supporting infrastructure are not the only potential sources of contamination in Hanford's Central Plateau region. Because the single-shell tank farms are sited close to waste disposal sites (i.e., cribs and/or trenches), it is imperative to determine the source of the contamination in the groundwater. Historically, this has been difficult because the characteristics of the waste change

significantly as it migrates through the sediments. However, innovative approaches, such as analyses of certain stable and radioactive isotope concentrations and signatures now are being used. These studies employ advanced analytical techniques and instrumentation to enable better “fingerprinting” of contaminant plumes, making quantitative identification of contaminant source terms plausible [23].

Use of ruthenium isotopic ratio analysis of sediment and groundwater samples collected in the vicinity of WMA T showed that vadose zone sediments from borehole C4104 (emplaced adjacent to tank 241-T-106) were contaminated by a single leak event. Further evaluation of groundwater samples collected from WMA T indicated that multiple sources (at least two) of contamination were present in the aquifer to the east of the T Tank Farm. The shallow groundwater samples had ruthenium isotopic ratios consistent with those measured in vadose zone samples from borehole C4104. Analysis of ruthenium isotopic ratios in depth-discrete groundwater samples collected from wells adjacent to (299-W11-25B) and east of (299-W11-45) the T Tank Farm, respectively, resulted in two distinct sets of isotopic ratio data. These results have led to the inference that a yet unidentified source, distinct from the T-106 tank leak in 1973, is responsible for the high Tc-99 concentrations observed with depth in the aquifer underlying Waste Management Area T. While the use of “fingerprinting” at the Hanford Site still is relatively new, it has the potential to provide significant support in defining the current and future risks associated with contamination currently residing in the vadose zone.

## CONCLUSIONS

As a result of continuing research by the scientific community engaged in Hanford Site issues, the understanding of processes controlling water flow and contaminant transport in the vadose zone have advanced significantly. The importance of small-scale variations in sediment texture on lateral flow of water and contaminants is now understood. Tank waste contaminants released to the vadose zone exhibit a wide range of mobility behaviors. Many tank waste contaminants have been strongly retarded by adsorption and precipitation reactions (e.g., cesium, plutonium, americium, and europium), while others have remained mobile (e.g., technetium, nitrate, and selenium). Still others show variable, waste-specific behaviors (e.g., cesium [where sodium concentrations are very high], strontium, chromium, and uranium) that are closely tied to evolving composition of porewater in the sediments, and for chromium, a change in oxidation state. Heat exchange with the subsurface sediment lowers the temperature of tank waste released to the vadose zone, and reactions with sediment minerals and secondary mineral precipitation have neutralized the high concentrations of base (OH<sup>-</sup>). Major geochemical features that may potentially affect the mobility of key contaminants of interest in the vadose zone include oxidation state, aqueous speciation, solubility, and adsorption reactions.

Laboratory studies conducted through PNNL’s Vadose Zone Characterization Project and basic-science geochemical studies are expected to continue in partnership to provide information to further refine the conceptual models used for risk assessment modeling. Results of these studies can be used to help ascertain uncertainties associated with the refinement of the conceptual models for contaminant migration in the vadose zone beneath the single-shell tanks. Results can also provide improved parameters, such as distribution coefficient ( $K_d$ ) values, and provide more

realistic models to predict contaminant movement under Hanford conditions. Ongoing research is focused on uranium and Tc-99.

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