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Large quantities of radioactive and chemical wastes resulting from Pu production for nuclear weapons are located in the vadose zone at the USDOE's Hanford Site, north of Richland, WA. The vadose zone here is characterized by often highly stratified glacial-fluvial sediments that give rise to complex subsurface-flow paths that contribute to uncertainty of contaminant fate and transport. Research efforts have focused on answering questions of contaminant transport from the viewpoint of geologic, biologic, geochemical, and hydrologic controls. This special section highlights key research topics concerning vadose zone problems at the Hanford Site. Research indicates that some of the contaminant species (^{137}Cs, ^{60}Co, ^{90}Sr) are retained by Hanford sediments as a result of geochemical reactions, rendering them effectively immobile except under extremely saline or acidic conditions, while other species (^{99}Tc, ^{129}I, ^{3}H) are typically mobile and have moved deep into the vadose zone and subsequently into groundwater. In addition, large quantities of organics, including carbon tetrachloride, have moved in complex ways as both vapor and liquid in the subsurface. Observed transport of mobile species is linked to liquid discharges and to elevated recharge rates that occur primarily at waste sites where land surfaces are void of vegetation and where winter rains have subsequently penetrated the subsurface wastes. A series of papers in this issue documents progress to date in understanding transport rates at Hanford, why anisotropy strongly affects the distribution of subsurface contaminants, why organic contaminants are difficult to find in the deep vadose zone, and what the impacts of hypersaline fluids are on waste form degradation and subsequent transport.

Abbreviations: CT, carbon tetrachloride; DNAPL, dense nonaqueous phase liquid; SVE, soil vapor extraction.

The Hanford Site is 1517 km^2 (586 mi^2) in size, located in a sparsely populated area in the rain shadow of the Cascade Mountains, adjacent to the Columbia River in southeastern Washington State (Fig. 1). The Hanford Site was set aside in the early 1940s by the U.S. Government to perform a top-secret mission, the Manhattan Project, for production of plutonium for atomic weapons used to end World War II and later to support the Cold War with the Soviet Union. Starting in 1943–1945, Hanford employed thousands of workers on the largest construction project in the world at that time. The workers built a series of nuclear reactors along with large monolithic concrete buildings used to reprocess wastes. Plutonium production continued at Hanford for over 40 yr. During the late 1980s, in the aftermath of the Chernobyl accident, plutonium production was suspended at Hanford (Gephart, 2003). Since then, efforts at Hanford have focused on cleaning up the legacy of wastes stemming from years of nuclear-weapons production.

More than 91,000 Mg (100,000 tons) of nuclear fuel were reprocessed between 1944 and 1990. Reprocessing required between 2100 and 16,500 L (550–4360 gal) of water per tonne of fuel. In addition, hundreds of thousand of tonnes of chemicals, including acids, solvents, nitrates, ammonia, and carbon tetrachloride, were used in the reprocessing plants. While some waste streams, containing various suites of radionuclides and chemicals, were put into storage tanks, more than 454 million L (120 million gal) of the waste was directly discharged to the ground (Gephart, 2003). Radioactive wastes from the Hanford Site mission are stored in 177 buried single- and double-shell carbon-steel tanks grouped together in what are called "tank farms" (Zachara et al., 2007). Of the 149 single-shell tanks, 67 are suspected to have leaked more than 3.8 million L (1 million gal) of tank waste to the vadose zone (Gephart, 2003).

Hanford now contains as much as 28,300 m^3 (1 million ft^3) of soil contaminated with radionuclides in liquid wastes released near processing facilities. Figure 2 schematically shows discharges to the vadose zone and the Columbia River from various sources. The total radioactivity discharged to ground is estimated to be nearly 2 million Ci (accounting for decay through the year 2000; Gephart, 2003). This large quantity of radioactivity, combined with 100,000 to 300,000 tonnes of toxic chemicals now residing in the vadose zone (Gephart, 2003), poses an ongoing environmental challenge. All of the contaminants that are not in some way attenuated in the subsurface (or do not decay away soon enough) will potentially be transported to the Columbia River (Gephart,

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2003). For this reason, studies related to understanding the fate of subsurface contamination from these wastes are critical.

Geologic Studies

The Hanford Site geology has been studied extensively. More than 3000 wells have been logged and their geologic profiles reported (Horton et al., 2005). These reports provide insight into the formation and structure of the sediments beneath the processing areas, where most of the waste currently resides. Sediments underlying the Hanford Site are glacial-fluvial in origin. Great floods swept through the Columbia Basin multiple times during the past 15,000 yr, eroding channels in the landscape and depositing large quantities of poorly sorted sands and gravels. Huge lakes, covering the Hanford Site and southern portion of the Pasco Basin to a depth of several hundred meters, were repeatedly formed and drained. Last et al. (2007) shows a cross-section of the glacial fluvial sediments at Hanford. As the lakes drained, desiccation cracks formed in large polygonal patterns. Differential pore pressures in underlying layered sediments acted to push fine materials upward creating clastic dikes, a prominent feature of the Hanford landscape. Clastic dikes have been found extensively in the waste storage areas as documented by Murray et al. (2007) and illustrated in Fig. 3. It is not known exactly how these clastic dikes act to influence contaminant migration, but it is thought that where the dikes are absent, the natural layering tends to spread contaminant plumes laterally (Ward et al., 2006). Where clastic dikes are present, they can act as cutoff walls. These features may limit lateral spreading, thus concentrating subsurface contaminants and increasing the possibility of gravity drainage to the underlying water table. Because of extensive disturbance of the near surface at disposal sites, geologic mapping of clastic dikes is incomplete. The full impact of these features on waste migration has yet to be fully documented.

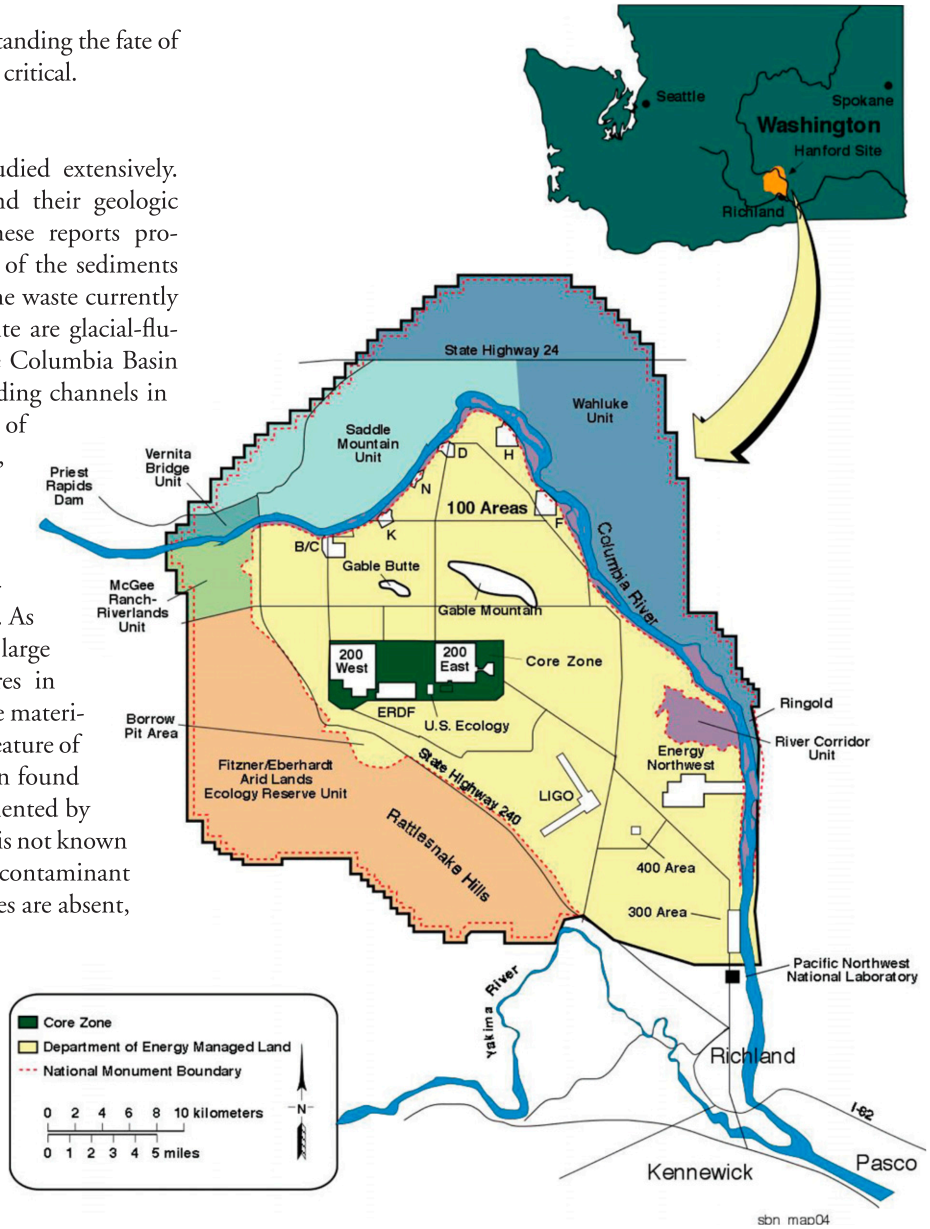

Fig. 1. Map of Washington State and location of the Hanford Site.

Soil Studies

Early studies of land disposal of waste at Hanford focused on assessing soils and sediments in and near the processing areas and evaluating their capacity to act as retention zones for radionuclides, where species such as ^{137}Cs (which dominates the radioactivity load of Hanford's waste) would be selectively retained in the subsurface sediments and subsequently decay away in time. Based largely on laboratory tests, which demonstrated that Hanford sediments had a large capacity for retaining ^{137}Cs, wastes were discharged directly to the ground in massive quantities. However, as noted by one early researcher, "The disposal of liquid waste to the ground in the process areas . . . was considered originally a temporary measure to be discontinued as soon as process development permitted. Then, as now, waste disposal techniques were an attempt to secure maximum economic benefit from waste disposal to the ground consistent with the welfare of human and other life" (McHenry, 1954, p. 4). The early studies treated the deep vadose zone sediments that lay under the waste sites as a large "ion exchange" column, where the radionuclides of interest (primarily ^{137}Cs and ^{90}Sr) would be retained by the sediments as long as their retention capacity was not exceeded.

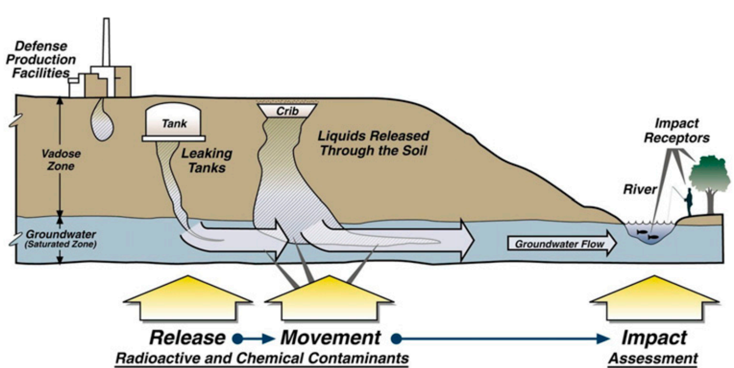

Fig. 2. Schematic diagram of waste discharges to the Hanford Site vadose zone and the Columbia River.

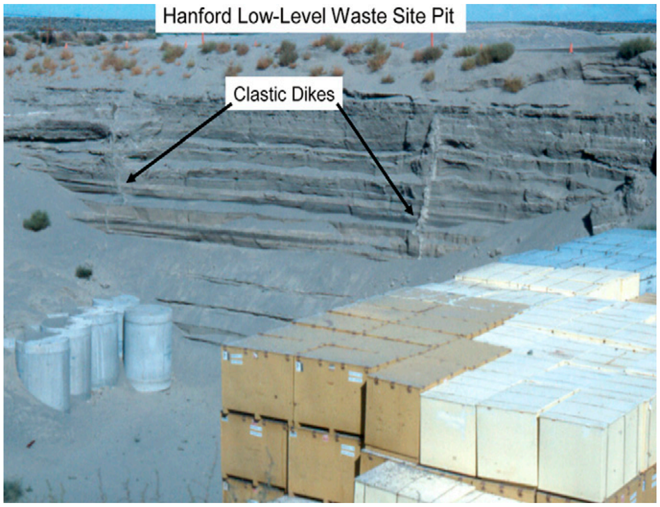

FIG. 3. Waste trench at the Hanford Site showing exposed clastic dikes (vertical features) in otherwise horizontally layered sediments.

Vadose Zone Flow Studies

Consideration was given early on to quantifying flow processes such as discharges from an unlined ditch to underlying ground water. Nelson (1962) and Reisenauer (1963) were among the first to attempt to quantify flow in Hanford's vadose zone. They developed numerical schemes to solve liquid discharge problems, using a nonlinear Richards'-type equation for flow in unsaturated porous media. This required an assessment of the unsaturated hydraulic conductivity, which they recognized as a highly nonlinear function of water content and water potential. Others used similar approaches to evaluate the flow of contaminants from tank leaks by assuming that the contaminants either moved with the water or that their movement was controlled by linear sorption mechanisms (Smoot and Sagar, 1990; Ward et al., 1997). Nonisothermal flow, including thermally enhanced vapor transport, was also considered in the early analyses (Enfield et al., 1973; Reisenauer et al., 1975; Finlayson et al., 1978). Since those early days, considerable effort has been spent in determining hydraulic properties of Hanford sediments for the purpose of using numerical models to solve the flow equation and to assess travel times from discharge points to the underlying ground water. A summary of such efforts has been documented recently by Last et al. (2004) and illustrated by Khaleel et al. (2007).

Another key parameter determined through vadose zone studies and modeling is recharge. Hanford Site researchers, starting with Hsieh et al. (1973) and continuing with Gee (1987) and Gee et al. (1992, 1994, 2005a,b), reasoned that modeling and other methods for estimating recharge have limitations at Hanford but that flow in the vadose zone can be measured directly using lysimetry. The common approach of estimating recharge (drainage below the root zone) using water balance parameters is difficult because of the desertlike conditions at Hanford. It is often assumed that measuring evaporation rates, precipitation, and runoff provides sufficient data to calculate the water balance and allow estimates of recharge. However, in semiarid climates like Hanford, standard methods for calculating the water balance require subtraction of two large, but uncertain, measurements (e.g., precipitation and evaporation), which can result in a recharge estimate that can be in error by as much as 1000% or more (Gee and Hillel, 1988). The use of lysimeters, which directly measure drainage, has provided estimates of recharge, over a wide range of soil and plant cover conditions at Hanford. Large lysimeters were constructed at several locations on the Hanford Site, and measurements were made over three decades to document recharge rates. The magnitude of observed drainage and resulting recharge from coarse sediments at Hanford has been surprisingly large for an area that has a cool-desert–type climate with mild winters, hot, dry summers, and an annual rainfall averaging less than 180 mm yr^{-1} ($\sim$7 in yr^{-1}).

Figure 4 shows the surface condition of a typical tank farm at the Hanford Site. Figure 5 illustrates the range of expected recharge rates due to changes in vegetative surface conditions under the typical Hanford climatic regime. Recharge averaging more than 60 mm per year (about one-third of the annual precipitation) has been measured at one of the Hanford lysimeter sites for the past 25 yr (Gee et al., 2005a,b). Such recharge appears to be typical of the Hanford Site when surfaces are stripped of vegetation and natural sediment layering is disrupted.

At tank farms and many other waste areas, vegetation is undesirable because of the potential hazard of uptake of radionuclides by plants (Gee et al., 1992). Issues related to Hanford's radioactive tumbleweeds and other biotic vectors have been openly discussed in *Science* magazine (Marshall, 1987) and have been a continuing concern for site managers. Reducing the risk of such problems prompted the site to initiate what has been called "controlled, clean and stable" actions at tank farms, which means that the surfaces of the tank farms are covered with gravel-dominated surfaces that have remained barren through the use of herbicides (Gee et al., 1992).

These actions serve to maximize the potential for recharge from Hanford waste sites. Extensive studies of landfill cover designs have indicated that covering waste sites with fine soils can prevent recharge under both normal and elevated precipitation conditions at Hanford (Ward and Gee, 1997; Fayer and Gee, 2006). However, for above-grade barriers that are armored by gravel or rock for wind and water erosion protection, there is a risk that the lack of vegetation on the side slopes can increase the potential for recharge in deep vadose zone areas surrounding waste locations. Therefore, landfill cover designs must be

FIG. 4. Bare surfaces at a Hanford tank farm.

holistic and consider the impact of the both the covered surface and the adjoining sideslopes and the interaction between surface and sideslope (Last et al., 2004). Three-dimensional modeling of water flow both in and around above–grade covers likely will be required to fully evaluate recharge control at Hanford landfills. In assessing the future of Hanford waste sites, reduced recharge from properly designed surface barriers (typically consisting of vegetated, fine soils) will be key to controlling contaminant migration rates.

Transport of Organic Liquids

Organic liquid wastes were disposed in large quantities into cribs, tile fields, and French drains at the Hanford Site, and monitoring has confirmed that organic contaminants are now present in groundwater beneath a number of waste-processing areas. The largest quantities of discharged organic wastes consisted of carbon tetrachloride (CT) mixed with lard oil, tributyl phosphate, and dibutyl butyl phosphonate. The major disposal facilities received a total of about 13.4 ML of liquid waste containing 363,000 to 580,000 L of CT. Assuming a maximum solubility of 800 mg L^{-1} and a fluid density of 1.59 g cm^{-3}, the 13.4 ML of liquid waste would contain approximately only 6700 L of CT dissolved in the liquid waste. This indicates that the majority of the CT entered the subsurface as an organic liquid (i.e., pure product). An estimate of the discharge inventory by Last and Rohay (1993) suggests that 21% of the CT has been lost to the atmosphere, 12% is retained in the vadose zone (dissolved in water, sorbed to the solid phase, and as a component of the gas phase), and 2% is dissolved in the saturated zone. The remaining 65% has not been accounted for and may have been initially present as residual liquid saturation in the vadose zone.

In recent years, two major remediation technologies have been applied to remove CT from the vadose zone and groundwater at Hanford. Since 1991 about 79,000 kg of CT was removed using a soil vapor extraction (SVE) system in the vadose zone. In addition, a pump-and-treat system for the unconfined aquifer removed close to 10,000 kg of CT from groundwater since 1994. Because neither remediation scheme has been effective in removing all of the CT from the subsurface, scientific issues have been raised and research initiated to improve remediation strategies for CT and other organic contaminants (Freshley et al., 2002). The science related to CT focuses on theory development, experimental verification, and numerical implementation into the STOMP (Subsurface Transport Over Multiple Phases) simulator (White and Oostrom, 2006). For instance, a recent theory describing the formation of residual CT in the vadose zone (Lenhard et al., 2004) has been experimentally tested in intermediate-scale flow cells (Oostrom et al., 2003; Oostrom and Lenhard, 2003) and has subsequently been incorporated into the STOMP simulator (White et al., 2004).

The improved simulator has been used to complete detailed three-dimensional simulations for the three major dense, nonaqueous phase liquid (DNAPL) waste sites in support of the development of a conceptual flow and transport model (Oostrom et al., 2004, 2006a,b, 2007). The simulations, including a detailed sensitivity analysis, consider the infiltration and redistribution of fluid waste (aqueous phase and DNAPL containing CT) as well as remediation using SVE.

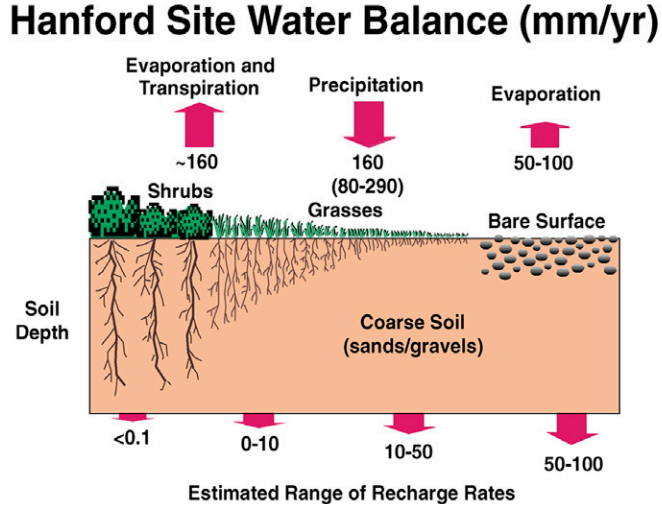

FIG. 5. Schematic diagram of Hanford Site water balance, including precipitation, evaporation, transpiration, and recharge rates, expressed in mm yr^{-1}, as related to surface conditions ranging from deep-rooted vegetation to bare surfaces.

Simulation results show that DNAPL behavior is strongly related to source, porous media, and fluid properties. Results indicate that the DNAPL at one disposal site with a small footprint but large waste stream penetrated deeper than other sites with larger footprints and smaller waste streams. Simulations indicate that some DNAPL at the smaller footprint site may have reached the water table while at adjacent sites with larger footprints, the discharges are still in the vadose zone. Carbon tetrachloride fluxes related to these simulations are consistent with estimates of dissolved CT in the groundwater (Murray et al., 2006). Although the simulations, conditioned on laboratory evidence, show that the majority of the DNAPL emanating from the sources remains below the footprint of the disposal sites, the CT vapors can move rapidly in the lateral and vertical directions. As a result, the vapor plumes below the sites are far more extensive than the DNAPL plume. Through phase partitioning, the CT vapors have the ability to contaminate large amounts of groundwater. Figure 6 shows the present conceptual model of CT transport at a Hanford waste site. Current experimental research at Hanford is directed toward an improved understanding of nonequilibrium CT volatilization and aqueous dissolution, as well as porous medium wettability effects. Such studies, combined with efforts to upscale the detailed laboratory studies, will assist site managers in reducing uncertainties in locating organic contaminant zones and implementing workable remediation strategies to reduce the liquid organic contamination problem at Hanford.

Geochemical Considerations

The concept of specific retention of radionuclides, such as ^{137}Cs and ^{90}Sr, was used in the early days of Hanford to justify discharge of tank wastes directly to the ground (McHenry, 1954). In most instances, the Hanford sediments have held strongly reactive contaminants at or near the discharge points. This has been determined by monitoring of waste sites with gamma scintillation probes that detect ^{60}Co and ^{137}Cs among other radionuclides. The waste sites that received intentional discharges of waste are undergoing characterization and remediation (Gephart, 2003).

The tank farms have been the focus of much of the geochemical and hydrologic characterization work at Hanford. Boreholes have been placed inside these tank farms and adjacent to the tanks to characterize the subsurface contaminants. Vadose zone monitoring has been performed for years, consisting of borehole geophysical logging to detect gamma-emitting radionuclides (Henwood and McCain, 2006). Because only gamma-emitting radionuclides can be detected with this method, the distribution of radionuclides such as ^{99}Tc and other nongamma or nonradioactive sources must be determined through drilling and sample characterization.

The observed deep movement of ^{137}Cs at several of the tank farms led to intensive investigation of the geochemical controls on migration of ^{137}Cs, ^{90}Sr, and several other contaminants. Zachara et al. (2007) address the geochemical processes that have controlled the migration of tank waste in the vadose zone. These studies have consisted of drilling characterization boreholes and collecting and characterizing contaminants in the vadose zone. Laboratory studies have shown that a wide range of geochemical processes, including ion exchange, precipitation and dissolution, and surface complexation reactions, have occurred involving tank waste and vadose zone sediments, moderating their chemical character and retarding the migration of select contaminants.

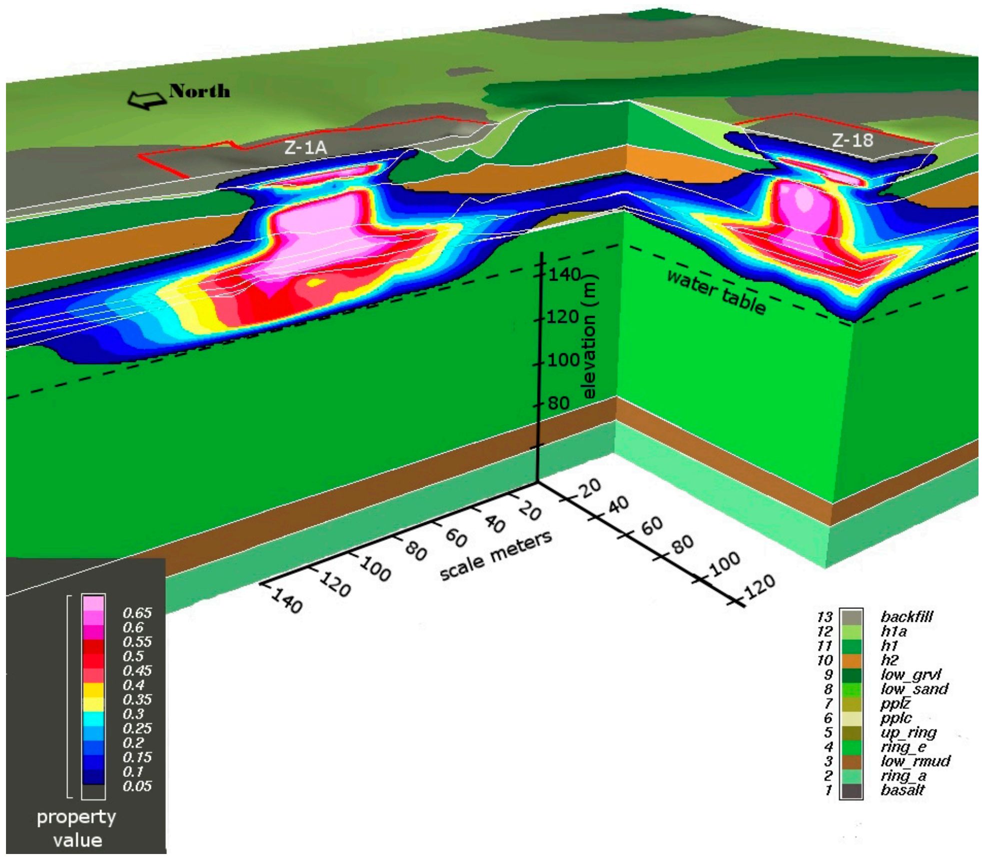

Fig. 6. Simulated carbon tetrachloride (CT) vapor plumes emanating from dense nonaqueous phase liquid disposed at the 216-Z-1A tile field and the 216-Z18 crib at Hanford. The "property value" legend reflects gaseous CT concentrations (g L^{-1}) in the vadose zone. The stratigraphic units denote Backfill, Hanford 1a (h1a), Hanford 1 (h1), Hanford 2 (h2), Lower Gravel (low_grvl), Lower Sand (low_sand), Cold Creek silt (pplz), Cold Creek carbonate (pplc), upper Ringold (up_ring), Ringold E (ring_e), Ringold Lower Mud (low_rmud), Ringold A (ring_a), and Basalt.

The studies demonstrated that mass action and osmotic effects from extremely high Na^{+} concentrations in the leaked wastes and enthalpy effects from high subsurface temperatures were responsible for anomalous deep migration of ^{137}Cs beneath tank SX-108 in the SX tank farm. Ion exchange was determined to be the dominant attenuation mechanism for ^{90}Sr associated with leaked tank wastes, but enhanced migration beneath the waste tanks was the result of a complex process of isotopic exchange between the contaminant ^{90}Sr and the native stable Sr pool within the interstices of basaltic lithic fragments in coarse-textured Hanford sediments. The subsurface migration of uranium is slowed in certain cases by surface complexation adsorption reactions, but studying the reactions in detail is problematic because of the difficulty of working with coarse-textured Hanford sediments, which are extremely heterogeneous.

Processes that were thought to facilitate migration of normally immobile species, including stable aqueous complex formation and mobilization of colloids, were found to be potential causes for transport but unlikely to occur in the field, with the exception of cyanide-facilitated migration of ^{60}Co. As a divalent cation, ^{60}Co is adsorbed strongly by Hanford sediments, but it has exhibited unretarded migration through the vadose zone beneath a waste site that received ferrocyanide wastes. Fission products including the oxyanions Tc, Mo, and Se are the most mobile tank waste constituents because their adsorption is suppressed by large concentrations of waste anions, the negative surface charge of the vadose zone clay fraction, and by the fact that their reduce forms are unstable in oxidizing environments.

Current Studies

In the following papers, a number of current studies at Hanford are presented that cover a wide range of investigations of Hanford's vadose zone. Last et al. (2007) describe use of borehole geologic data to provide a geologic framework for vadose zone flow and transport simulations. Ward and Zhang (2007) describe methods to estimate effective hydraulic properties for anisotropic unsaturated flow. In a companion paper, Zhang et al. (2007) describe the use of a pore connectivity–tortuosity concept to model saturation-dependent anisotropy. Khaleel et al. (2007) present an impact assessment of tank farm vadose zone contamination, and Rucker and Fink (2007) show how geophysical characterization can be used in some cases to delineate contaminant plumes and to assist with vadose zone characterization. Murray et al. (2007) present the influden of clastic dikes on contaminant migration. Oostrom et al. (2007) summarize CT flow and transport in the vadose zone at a waste site responsible for most of the subsurface accumulation of this important DNAPL. Zachara et al. (2007) summarize an extensive body of work evaluating the reactive chemistry of tank wastes discharged to the Hanford sediments. The geochemical speciation of uranium is described by McKinley et al. (2007). Christensen et al. (2007), Conrad et al. (2007) and Evans et al. (2007) illustrate that isotope geochemistry can be used to track waste plumes in the vadose zone. Finally, Thornton et al. (2007) present an analytical assessment of a vadose zone remediation technology involving reactive gases.

The papers in this special section represent a small fraction of the extensive studies focused on fate and transport of contaminants in the Hanford vadose zone. Wastes at the site have been transported through the vadose zone and resulted in nitrate, chromium, tritium, uranium, ^{99}Tc, ^{90}Sr, and carbon tetrachloride groundwater plumes. As cleanup continues at the Hanford Site, additional vadose zone studies will be performed to characterize the extent of the contaminant plumes, determine their rates of migration, and evaluate remediation solutions. These solutions may include strategies such as in situ treatment to immobilize wastes (e.g., Thornton et al., 2007). They may also include the use of effective landfill covers over sites where waste removal may be impractical and cause excessive risk to workers (Gephart, 2003; Fayer and Gee, 2006). Understanding the hydrologic controls and geochemical interactions that govern vadose zone migration rates will be key factors in ultimately dealing with the legacy from nuclear waste production at the Hanford Site.

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