Hanford Tank Farm RCRA Corrective Action Program

J.R. Kristofzski, F.M. Mann, F.J. Anderson
CH2M HILL Hanford Group, Inc.
P.O. Box 1500, Richland, Washington 99352, USA

R.W. Lober
U.S. Department of Energy, Office of River Protection
2440 Stevens Dr., Richland, Washington 99354, USA

ABSTRACT

As a consequence of producing special nuclear material for the nation's defense, large amounts of extremely hazardous radioactive waste was created at the U.S. Department of Energy's (DOE) Hanford Site in south central Washington State. A little over 50 million gallons of this waste is now stored in 177 large, underground tanks on Hanford's Central Plateau in tank farms regulated under the Atomic Energy Act and the Resource, Conservation, and Recovery Act (RCRA). Over 60 tanks and associated infrastructure have released or are presumed to have released waste in the vadose zone.

In 1998, DOE's Office of River Protection established the Hanford Tank Farm RCRA Corrective Action Program (RCAP) to:

- Characterize the distribution and extent of the existing vadose zone contamination
- Determine how the contamination will move in the future
- Estimate the impacts of this contamination on groundwater and other media
- Develop and implement mitigative measures
- Develop corrective measures to be implemented as part of the final closure of the tank farm facilities.

Since its creation, RCAP has made major advances in each of these areas, which will be discussed in this paper.

INTRODUCTION

The Defense Nuclear Facilities Safety Board states that "Hanford is engaged in the world's largest environmental cleanup project." [1] A major reason for this description is the work being undertaken to characterize and then remediate past releases from the Hanford tank system. This system was created to store the extremely hazardous radioactive and chemical waste generated during the processes to create the special nuclear material used for the defense of the United States.

The Tank Farm Vadose Zone Project supports the U.S. Department of Energy's Office of River Protection in implementing the Hanford Tank Farm RCRA Corrective Action Program (RCAP). The Tank Farm Vadose Zone Project was created to better understand and deal with contamination in the soil beneath the underground radioactive liquid waste storage tanks. The

contamination is the result of past leaks from tank farm facilities and purposeful discharges from past operations.

The emphasis of this paper will be on the field and laboratory activities.

BACKGROUND

The Hanford Site, a facility in the U.S. Department of Energy (DOE) nuclear waste complex, encompasses approximately 1517 square kilometers northwest of the city of Richland along the Columbia River in southeast Washington State. The federal government acquired the site in 1943 for the production of plutonium. Production of such special nuclear materials continued until the 1980s. Beginning in the 1990s, DOE has focused on cleaning up the Hanford Site. For an extended summary see Michele Gerber's History of Hanford Site Defense Production (Brief) [2]. For more information, read her book On the Home Front: The Cold War Legacy of the Hanford Site [2] or Hanford: A Conversation About Nuclear Waste and Cleanup by Roy Gephart [4].

Once the land was acquired, many facilities were constructed, including 3 full-size nuclear production reactors along the Columbia River and 3 extremely large radiochemical separation plants in the Central Plateau. In addition, 64 large underground, high-level waste storage tanks (B, C, T, and U tank farms) were built near the separation plants in the 200 East and West Areas. Figure 1 shows the current facilities in the 200 East and 200 West Areas, including these early facilities.

In the early 1950s, additional facilities were built in response to world events. Another reactor was built near the Columbia River. A new separations facility and 18 more underground tanks were built in the 200 West Area. During this time, sludges were mined from the underground tanks and re-processed through U Plant to extract uranium for reuse in reactor fuel elements.

The next Hanford facility expansion occurred in the mid 50s. The KE and KW reactors were built along the Columbia River. The PUREX separations plant was built in the 200 East Area. Twenty-one (21) more underground tanks were built.

The peak production years were from 1956 through 1964. During this time, the last production reactor (N) was built. During this time, a number of "isotope campaigns" produced mega-curie quantities of cerium, strontium, cesium, promethium, and other radioisotopes from re-processing the wastes stored in the underground tanks.

A brief production burst occurred in the early and mid-1980s. However, no defense plutonium has been produced at the Hanford Site since 1988. After 1988, the Site turned to cleaning up its wastes.

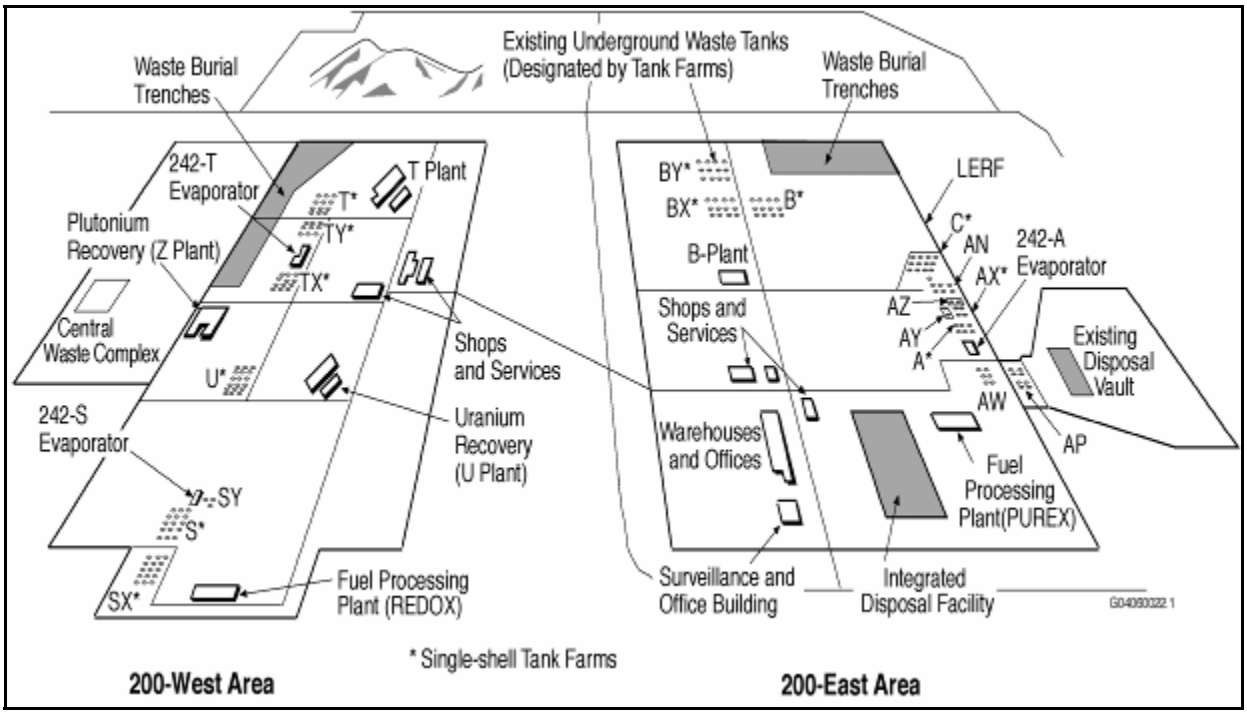

Fig. 1. Facilities in the 200 East Area and 200 West Area of Hanford's Central Plateau. (Letters followed by * are single-shell tank farms)

Hanford operations have released contaminants to the environment. In the early years of Hanford's plutonium production, airborne releases from the stacks of the separations plants constituted the largest waste releases to the environment. During the years of peak production, the cooling waters for the reactors near the Columbia River became contaminated and then contaminated the river as they were discharged to it. Hanford operations even purposefully discharged waste to the soil.

Hanford policy allowed untreated radioactive waste discharges to the soil from 1943 to 1995. Over 400 billion gallons of waste (predominately contaminated cooling water) was discharged to the soil, producing large mounds in the groundwater system. Even tank waste fluids (on the order of 100 million gallons) were intentionally discharged to the ground after cascading through three or more tanks to allow settling of the more radioactive sludges.

Double-shell tanks were built starting in the late 1960s to supplement and replace the older single-shell tank (SST) design. Many of the first generation SSTs have leaked, with current estimates being about 500,000 gallons being lost to the soil [5]. Retrievable liquids have been removed from most of the SSTs and wastes from 6 of the SSTs (C-103, C-106, C-201, C-202, C-203, and S-112) have been removed such that residual wastes in these tanks meet the removal goals of the Hanford Federal Facility Agreement and Consent Order (also known as the Tri-Party Agreement) (Ecology 1998).

The current plan is to retrieve most of the 53 million gallons of waste in the tanks, leaving about 0.5 million gallons as residual in the tanks. The retrieved waste will be processed, creating three waste forms:

- Vitrified (glass) high-level waste, destined for a federally licensed geologic repository
- Vitrified low-activity waste (that is, waste processed to remove most of the radioactivity) destined for disposal at Hanford's Integrated Disposal Facility
- Grouted secondary waste (that is waste produced from making the glass products), also destined for the Integrated Disposal Facility.

ACTIVITIES

Overview

The range of activities in RCAP is large, ranging from planning (and then implementing) characterization efforts to implementing mitigative measures to lessen impacts. The emphasis of this paper is on the field and laboratory activities.

Sediment Collection

A major effort of the RCAP has been to determine the nature and extent of the past releases, particularly of the largest past releases. A significant subsurface sediment collection effort has been and is currently underway. Table 1 displays these RCAP sediment collection activities.

Two major advances have been the use of slant borehole drilling to obtain samples under a tank known to have leaked and the optimization of direct push technology inside tank farms.

The largest leaks from the single-shell tanks are thought to come from the bottoms of the tanks. Yet the most difficult locations to investigate are underneath the tanks, as the last event that is wanted is to drill into and through a tank so that more contamination is released. Efforts early in the project life looked at directional drilling (that is, drilling in which the direction of the bore could changed during drilling). However, the industry techniques for such drilling would introduce significant amounts of water into the subsurface, potentially accelerating the movement of already released contaminants. A safer technique is to drill a borehole at an angle passing near the tank and penetrating the sediment beneath the tank bottom. In the late 1990s, many believed that such a slant drilling could not be done or would not be done in a tank farm. However, after many demonstrations outside of tank farms, the slant borehole drilling under tank SX-108 (the source of one of the largest subsurface plumes) was approved.

The intent was not only to drill underneath the tank, but also to retrieve samples. The samples were expected to contain 10^9 picoCuries of ^{137}Cs per gram of sediment. An unshielded Coke-can sized sediment sample with this level of contaminant would be lethal. So additional planning occurred to protect the workers who would be gathering and handling of samples.

The planning was successful. The borehole was drilled, passing within 9 ft of the base of the tank. Seventeen split spoon samples each containing two two-inch diameter by 6 in long cores were gathered from beneath the tank and then analyzed. All of these data were reported in the Field Investigation Report for Waste Management Area S-SX [6]. Figure 2 shows a picture and diagram of the SX-108 Slant Borehole operation.

Table I. RCAP Sediment Collection Activities

Tank Farm	Activity
B	Drill, sampled, and decommissioned a borehole near Tank B-110, collecting 135 samples.
	Placed 8 vertical direct pushes around diversion boxes B-151, -152, and -153, collecting 8 samples
BX	Drill, sampled, and converted into a groundwater well a vertical borehole 75 feet to the northeast of Tank BX-102, collecting 118 samples.
	Placed 8 vertical direct pushes in the BX-102 uranium plume, collecting 3 samples.
	Placed 3 vertical direct pushes near Tank BX-110.
C	Drill and sampled borehole near Tank C-105, collecting 118 samples
	Placed 20 vertical direct pushes near the pipeline leak, collecting 20 samples
	Placed 6 slant direct pushes near the pipeline leak, collecting 15 samples
S	Placed vertical direct pushes down to 40 feet around diversion box near S-104, retrieving samples
SX	Extended, sampled, and decommissioned borehole near Tank SX
	Drilled, sampled, and decommissioned 30 degree slant borehole under Tank SX-108 (Total length=170 feet), collecting 17 samples.
	Drill, sampled, and converted into a groundwater well a vertical borehole south of Tank SX-115, collecting 43 samples.
T	Drilled, sampled, and decommissioned two (2) boreholes near Tank T-106, collecting 44 samples
	Placed 6 vertical direct pushes near Tank T-101, collecting 2 samples
TX	Drilled, sampled, and decommissioned three (3) boreholes near Tanks TX-104, -105, and -107, collecting 56 samples
TY	Placed 24 vertical direct pushes near Tanks TY-103, -105, -107, collecting 5 samples

The second major advancement is in the area of direct push technology. Boreholes are very expensive to install in the tank farms. Although costs have been reduced from many millions of dollars per borehole down to less than a few hundred thousand dollars, many sampling events are needed, particularly for samples near the surface. Direct push technology (where a rod is pushed through the sediment) was investigated, but found wanting as the rods could not penetrate the compacted sediments at the bottom of the tank farms. A new technique (using a hydraulic hammer) was developed that is now lowering costs and shortening the field schedule. It has been used in B, BX, C, T, TY, and U Farms.

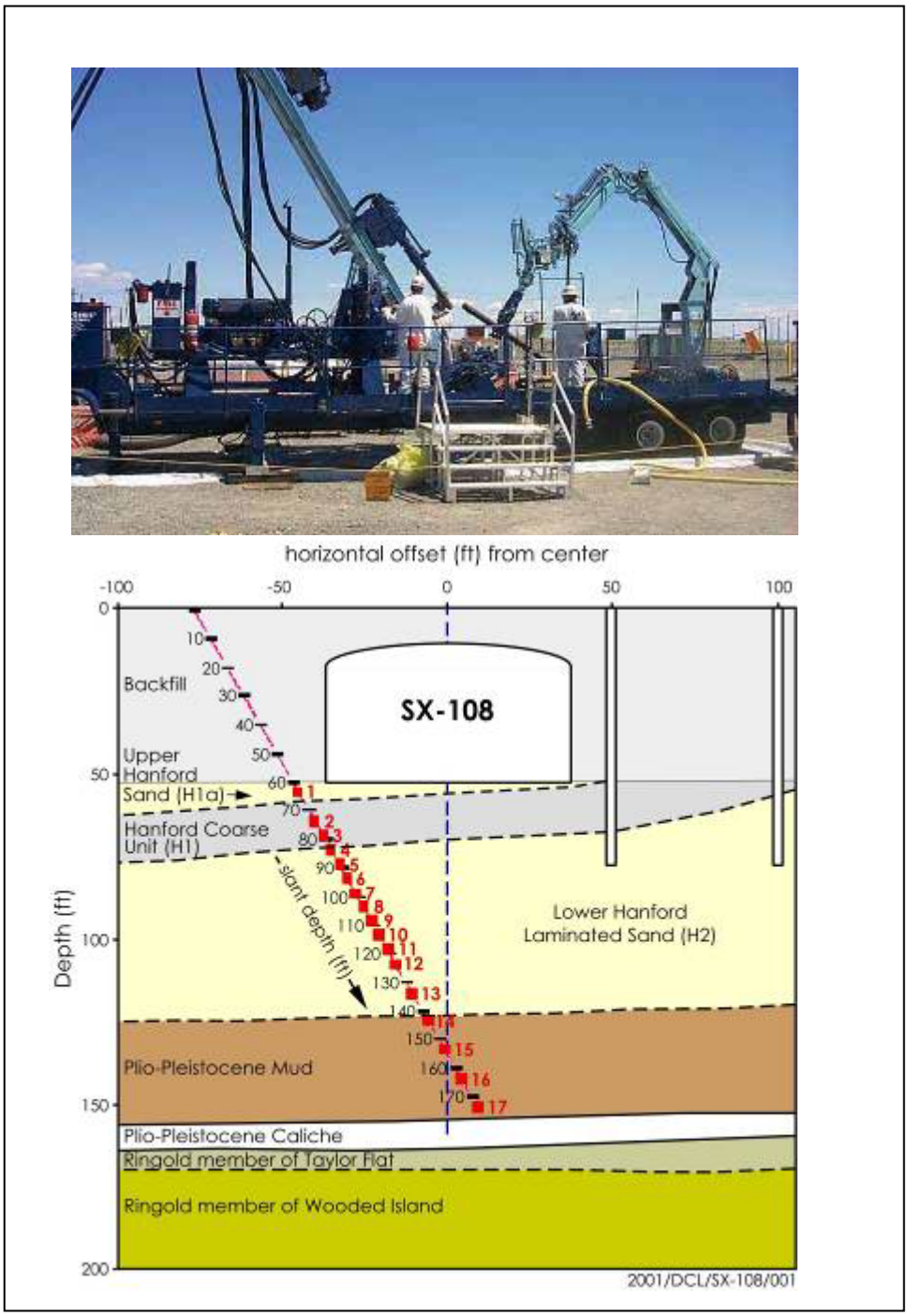

Fig. 2. Photo and diagram of SX-108 slant borehole drilling.

Sediment Analysis

Once sediment samples are obtained, they undergo laboratory analysis not only to determine contaminant concentrations, but also (and usually more importantly) the processes that control the movement of contaminants [6-18]. A relatively recent extension of the Vadose Zone project has been the laboratory analyses of sludges and salt cake from pre- and post-retrieved single

shell tanks in support of determining what processes are important in releasing contaminants from closed tanks into the environment. Some laboratory highlights from the 10 years are:

- Interactions of Highly Caustic Tank Waste with Sediments
- Cesium Mobility
- Uranium Mobility
- Fate of Technetium Through the Bismuth Phosphate Precipitation Process.

Tank waste is kept extremely basic (pH > 14) to minimize corrosion of the single-shell carbon steel tanks. In addition, some of the tanks had temperatures above 200 ^{o}F because of the radioactive decay of the large concentrations of short-lived radionuclides. At the birth of the Tank Farm Vadose Project, many believed that the waste would greatly alter the physical and chemical properties of the sediments that it encountered right outside the leak location. Some changes in chemical and mineralogical properties of near-tank sediments have been seen. Besides the presence of the contamination, the pH of the soil has been increased from its natural value of about ~7.5 to 8 to as high as ~9.5 to 10. In addition, water soluble magnesium and calcium generally present on the sediment cation exchange sites have been displaced downward having been replaced by sodium present in very high concentrations in the tank liquids that leaked.

One of the main technical reasons that the Tank Farm Vadose Zone Project was created was because radioactive cesium was found much further away from tanks in the SX tank farm than then-current theories could explain. Based on extensive laboratory analyses, John Zachara and his team from the Pacific Northwest National Laboratory studied the highly contaminated sediments from below the SX tank farm and developed a more accurate and mechanistically based cesium adsorption model that explained the processes involved and determined the parameters needed to quantify cesium migration through sediments (see Appendix D of the Field Investigation Report for Waste Management Area S-SX [19] and primary references by Zachara et al. [20], Liu et al. [21-23], and McKinley et al. [24-25]). These key findings that were not appreciated in earlier cesium adsorption models recognized that the large amount of sodium that is in tank waste was concentrated by self boiling to concentrations up to 4 times higher than previously thought and that the fission process created four isotopes of cesium that when also concentrated by the self boiling resulted in high enough mass concentrations to saturate available sorption sites in Hanford sediments.. As sodium is very similar in its chemical properties to cesium, the large quantities of sodium significantly competed for the available sorption sites usually preferred by cesium and the larger than previously considered mass concentrations of cesium in the leaked fluids also overwhelmed (saturated) the low quantity of preferred sorption sites in the sediments closest to the tank leak. Thus, instead of being relatively easily adsorbed in a small volume of sediment, cesium present in fluids that leaked from self boiling tanks migrates further than previously predicted until the cesium atoms find sorption sites not already saturated with cesium or sodium. Hence cesium from the self boiling tanks that leaked, a subset of all of Hanford's single shell tanks, moves with the water larger distances than previously predicted. As the leaked fluid migrates away from the leak location, the sodium in the leaked fluids is diluted by direct ion exchange removal onto the native sediments and by mixing with the sediment's natural moisture. The cesium concentrations also diminish by sorption onto both cesium preferred sites and general cation exchange sites on the sediments, and by the same

dilution with the sediment's natural moisture content. Once the sodium and cesium concentrations in the pore fluids drop to lower concentrations, the normal sediment interactions adequately modeled in the past take place with the result that cesium's migration is significantly retarded in relation to sodium and most other cationic species. The parameters needed to model the behavior of cesium in the liquids that leaked from self boiling tanks have now been determined and current cesium migration modeling predictions agree well with the field observations (both spectral gamma logs and distributions measured in samples from contaminated boreholes).

Much work has been done to understand the fate of uranium in subsurface systems below the single-shell tanks in the Central Plateau and the fuels fabrication disposal facilities in the 300 Areas. However, at the present time a universal and simple uranium fate conceptual model is not available to quantitatively describe uranium migration. Work continues; predictions and mechanistic understanding are improving.

Although Hanford Site operations were well planned and documented in the early years, sometimes assumptions were made on, what were then, less important items. However, sometimes more recent information reveals that a technical issue is important to making clean up decisions or for understanding the fate of key contaminants. One good example is determining for the Bismuth Phosphate process into which waste stream technetium was partitioned. This process was the first one used to separate plutonium from irradiated reactor fuels at Hanford. The volumes of waste created by the bismuth phosphate processing at Hanford constitute 80% of the total volume of fuel reprocessing wastes generated. At the time, technetium was thought to be unimportant because of its long half-life and lack of penetrating radiation. We now know that technetium usually drives long-term groundwater risks. During bismuth phosphate operations, it was assumed that 90% of the technetium went with the metal waste stream and 10% with the first and second cycle product wash waste streams based on gross beta analyses of batches of the various waste streams. The Tank Farm Vadose Zone Project funded the Pacific Northwest National Laboratory (PNNL) to simulate the bismuth phosphate process. The results of four replications of the bismuth phosphate process found that >97% of the technetium would be found in the metals waste solution that was disposed to single-shell tanks and that 1% or less would be found in the waste streams that were sent directly to cribs and trenches. This new information provided better agreement with tank waste measurements as well as technetium measurements below the trenches just west of the BX Tank Farm.

Geophysical Measurements

Geophysical measurements have long been used in the Hanford tank farms to infer the nature and extent of released contamination. In the 1940's continuing into the 1990's, gross gamma radiation measurements were done in the hundreds of boreholes that exist in the tank farms. The measurements from the 1970s on (when the measurements were stored electronically rather than just on paper) were reanalyzed and published [26-37]. Starting in the late 1990's, the boreholes were remeasured with spectral gamma radiation detectors [38-49]. Combining the analyses of the historical and the modern spectral gamma logs allowed the Tank Farm Vadose Zone Project to identify the most impacted areas in the single-shell tank farms. It was through these assessments of the gross and spectral gamma historical information that the project determined

where to place new boreholes to obtain samples for laboratory analyses. In addition, moisture measurements are now routinely made with around tanks being retrieved.

Other geophysical techniques have been tried at the Hanford Tank Farms, but have been found to have limited applicability. However, recent use of high-resolution resisitivity has shown great promise. This technique, known as surface geophysical exploration (SGE) measures the electrical resistance between many different points (most on the surface, but some subsurface). These resistance values can be inverted (as is done in computer assisted tomography –or CAT-scans) to show the electrical resistance of a particular volume of sediment. Knowing that the key species that influence the vadose zone resistivity or its reciprocal (conductivity) in the moist sediment are dissolved salts, usually sodium and nitrate from the neutralization of highly concentrated nitric acid wastes with sodium hydroxide, the actual concentration can then be inferred. Figure 3 shows the "resistivity" plumes under S Farm. Recent reports [50-53] give more information on the use of high-resolution resistivity or SGE at the Hanford Tank Farms.

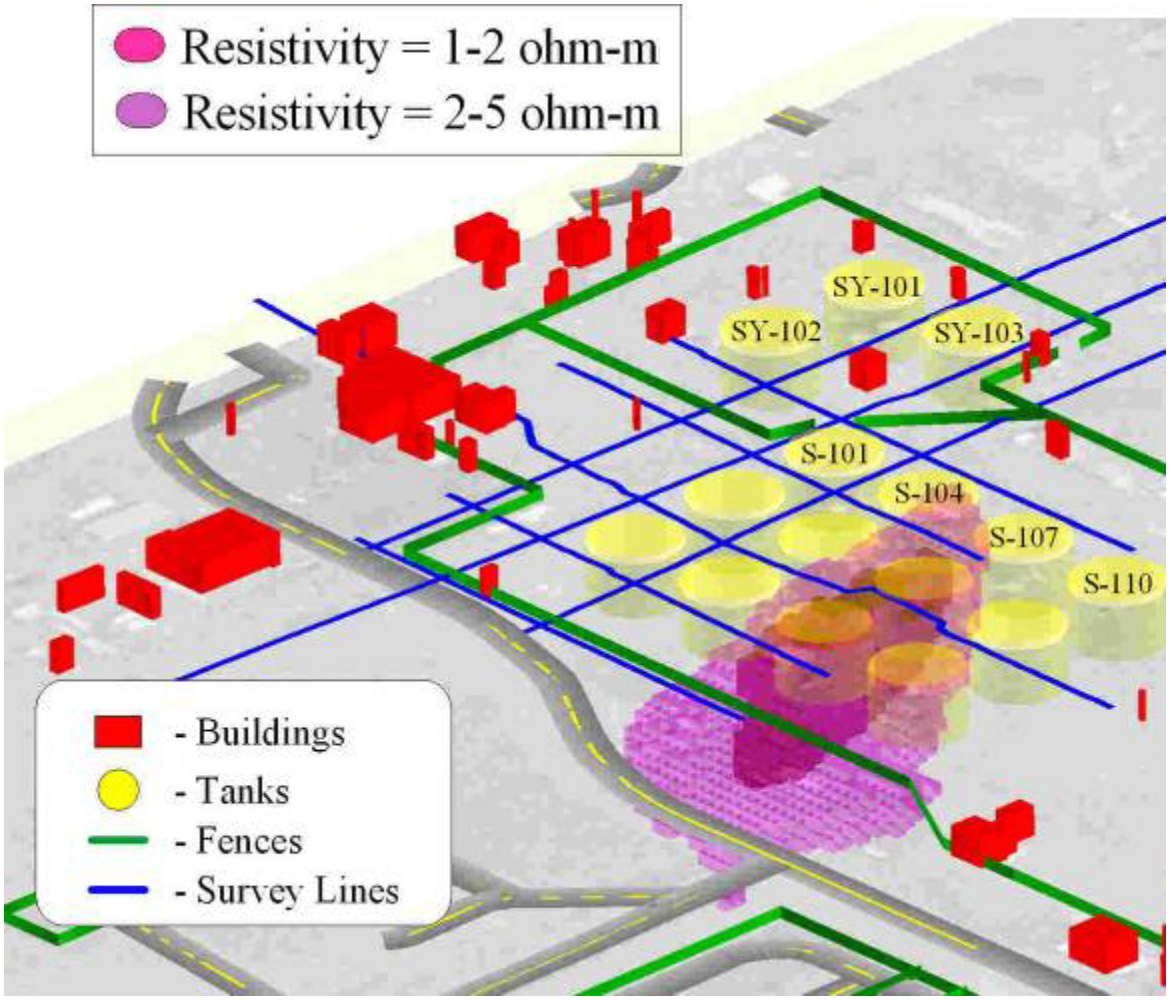

Fig. 3. Plumes under T Farm as determined by surface geophysics exploration.

Analysis

However, data are not just gathered, they are analyzed and their implications to understanding vadose zone properties and processes are determined. Before any field investigations were

performed, all available technical and geotechnical data were collected in Subsurface Conditions Descriptions Reports [54-58]. As data were collected and analyzed, the future impacts from such past releases were estimated in Field Investigation Reports [19, 59, 60]. Finally, estimates of the impacts from past releases and from residues left in the tanks and other farm infrastructure was estimated in the Initial Single-Shell Tank Farm System Performance Assessment for the Hanford Site [61] (and a paper at this conference).

Interim Measures

The tank farms will undergo final closure (including remediation of the soil and structures) many years in the future. However, mitigative measures (known as interim measures) have already been implemented and more are planned. These interim measures are designed to reduce the flow of moisture into the farm subsurface.

Already, all of the existing nearly 800 boreholes have been recapped, eliminating a potential preferential pathway through the vadose zone. In addition, all water lines going into and in the tank farms have been tested. All water lines that failed were cut and capped outside of the farms as were water lines that were no longer needed. As the tank farms are in local depressions (to aid the gravity feed of waste from processing plants), berms and gutters were constructed to divert rain water, snow melt, and accidental water discharges away from the Tank Farms. Figure 4 shows some of the interim measures already installed.

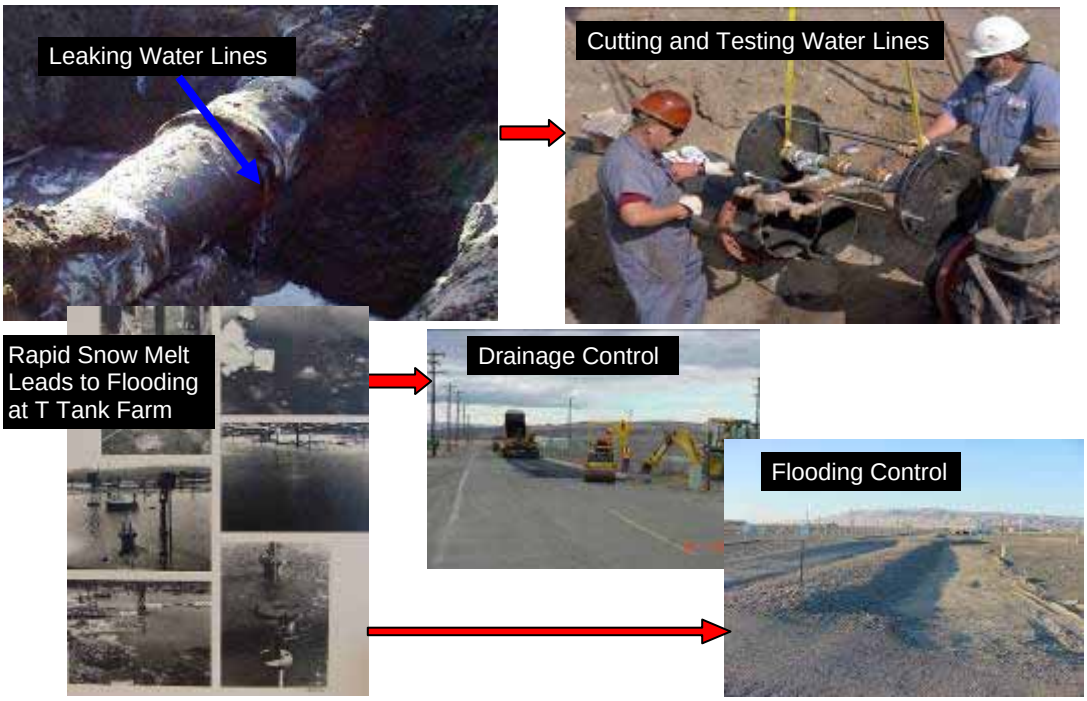

Fig. 4. Examples of interim measures installed at Tank Farms.

The next interim measure is the installation of temporary barriers over the tanks that have leaked and are expected to produce most of the groundwater impact. This barrier, made of polyurea and polyurethane, is similar to the protective pads put into truck beds. Already, moisture measuring equipment has been installed. Additional moisture measuring equipment and the barrier itself

will be installed over Tank T-106 and its neighboring tanks to cover the biggest (in terms of leak volume and technetium-99 release) plume in the tank farms.

PATH FORWARD

The Tank Farm RCRA Corrective Action Program is finishing up the first phase of its activities (the characterization of the largest tank farm releases). It has begun Phase 2 (characterization and analysis sufficient to determine the corrective measures needed for final soil and infrastructure remediation). The program will continue to seek new methods and technologies. The program's goal, as that of CH2M HILL Hanford Group, Inc and DOE's Office of River Protection, is to reach final tank farm closure (including the remediation of the soil and infrastructure).

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