Deep Vadose Zone Contamination Due to Releases from Hanford Site Tanks

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management
Contractor for the U.S. Department of Energy
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ABSTRACT

CH2M HILL Hanford Group, Inc. (the Hanford Tank Farm Operations contractor) and the Department of Energy’s Office of River Protection have just completed the first phase of the Hanford Single-Shell Tank RCRA Corrective Action Program. The focus of this first phase was to characterize the nature and extent of past Hanford single-shell tank releases and to characterize the resulting fate and transport of the released contaminants. Most of these plumes are below 20 meters, with some reaching groundwater (at 60 to 120 meters below ground surface [bgs]).

INTRODUCTION

The Department of Energy’s Hanford Site in south-central Washington State produced much of the special nuclear material used for America’s defense. The most hazardous waste produced (currently just over 200 million liters) is stored in large underground tanks pending treatment. Over 1.5 trillion liters of less hazardous waste were intentionally discharged to the soils. In addition, an estimated 4 million liters were unintentionally released from the tanks. Some of the waste has reached the deep vadose zone (beyond 20 meters bgs).

This paper presents
♦ A background on Hanford Tank Farm history and the history of the program to investigate the releases
♦ The efforts to characterize the nature and extent of the released contaminants as well as to determine parameters important for future transport of the contaminants, and
♦ The efforts to mitigate the future impacts of the contaminants.

BACKGROUND

Tank Farm History

The Hanford Site, a facility in the U.S. Department of Energy (DOE) nuclear waste complex, encompasses ~1517 square kilometers northwest of the city of Richland along the Columbia River in south-central 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.
Over the years, 9 full-size nuclear production reactors were built along the Columbia River and 5 extremely large radiochemical separation plants were built in the Central Plateau. To store the most hazardous waste produced from the production of the special nuclear material, 177 large underground storage tanks were built near the separation plants. Less hazardous waste (over 400 billion liters) was disposed directly to the ground.

Initially, single-shell tanks were built, ranging in size from 200,000 to 4.6 million liters in capacity. These tanks had a single wall of carbon steel, with a concrete shell below the steel bottom, a concrete shell on the outside of the steel wall, and a concrete dome. The 149 single-shell tanks are arranged in tank farms (from 4 to 18 tanks per farm). The 12 farms are arranged into 7 waste management areas (WMA) (WMA A-AX, WMA B-BX-BY, WMA C, WMA S-SX, WMA T, WMA TX-TY, and WMA U) for regulatory purposes. In addition, there are 28 large (~4 million liter capacity) double-shell tanks.

There are no indications that waste contained in the double-shell tanks have leaked into the annulus between the steel walls, much less into the environment. However, it is very clear (from drops in waste levels, external gamma radiation readings, and measurement of contamination levels in outside sediments) that single-shell tanks have released contaminants to the soil (from buckling tank bottoms, from overfilling, and other structural problems). Sixty-seven tanks have leaked or are assumed to have leaked. Estimates of the volumes range up to ~4 million liters and ~1 million curies of activity [1]. All retrievable liquids have been removed from the single-shell tanks. Wastes from 7 of the single-shell tanks (C-103, C-106, C-201, C-202, C-203, C-204, 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 [2].

History of the Tank Farm RCRA Corrective Action Program

In the mid 1990s, Hanford Site regulators and stakeholders became concerned that tank waste constituents (particularly cesium-137) had traveled farther (perhaps to groundwater) than had previously been presumed. The U.S. Government Accounting Office issued a report, Understanding of Waste Migration at Hanford is Inadequate for Key Decisions [3], and an external expert panel provided their views in TWRS Vadose Zone Contamination Issue Expert Panel Status Report [4].

In response to these concerns, the Tank Farm RCRA Corrective Action Program was established. With the help of various stakeholders and regulators, the Tank Waste Remediation System Vadose Zone Program Plan [5] was prepared and issued. The activities laid out in this ambitious plan are now being completed. The technical goals outlined in the plan are:

- Provide vadose zone information and impacts to Tank Waste Remediation System decision makers
- Determine the nature and extent of vadose zone contamination in the tank farms through new field studies, laboratory analyses and experiments, and historical data searches
- Validate models used in providing information
- Develop the database needed for tank farm risk models
Perform interim corrective actions that will lessen the impacts of existing tank leak contaminants.

The principles outlined in the program plan are:

- Information generated will be determined by the need of other Tank Waste Remediation System programs.
- Scientific methods and principles will be used.
- Information from other programs will be used in the Vadose Zone Program as appropriate.
- Before new data or tools are generated, current information will be reviewed to develop understanding and identify what, why, where, and how new information will be collected.
- External peer review is important for success.
- Input from the public is important for success.

The first phase has just been completed with the publication of the Hanford Tank Farm RCRA Facility Investigation Report [6]. This report summarizes data, information, and knowledge reported in the Initial Single-Shell Tank System Performance Assessment for the Hanford Site [7] and in the Field Investigation Reports for WMA S/SX [8], WMA B-BX-BY [9], WMAs T and TX-TY [10], WMAs C and A-AX [11], and WMA U [12]. The RCRA Facility Investigation Report also reports new data and information obtained since the issuance of the early Field Investigation Reports as well as synthesizes knowledge across all of the single-shell tank farms.

CHARACTERIZATION

The backbone of the Tank Farm RCRA Corrective Action Program has been characterization. A variety of techniques have been deployed:

- Boreholes: used for collecting samples and geophysical logging
- Hydraulically driven probe holes: used for collecting samples and geophysical logging
- Geophysical logging: gamma radiation and moisture content
- Laboratory analyses of retrieved samples.

Boreholes

Boreholes provide a means of directly interrogating portions of the vadose zone. These boreholes provide soil samples which can be analyzed in the laboratory, access for geophysical sondes (spectral gamma sondes, neutron-moisture sondes, etc.), and for placement of a variety of monitoring sensors. When extended to groundwater, boreholes provide an opportunity to sample the groundwater and can be converted to permanent wells should that be appropriate. Because the Vadose Zone Program initially targeted those places where the greatest insult to the environment was known to have taken place, the number of boreholes was limited due to the inherent dangers. As the efforts have been directed toward areas of lesser insult, the number of boreholes has increased; Table I chronicles the boreholes advanced by the Vadose Zone Program. A major achievement was placing a slant borehole underneath Tank SX-108 (see Fig. 1).
Table I. Distribution of the 11 Boreholes Drilled by the Tank Farm Vadose Zone Program

<table>
<thead>
<tr>
<th>Tank Farm</th>
<th>Boreholes Drilled</th>
<th>Depth (feet)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>SX</td>
<td>3</td>
<td>211&lt;sup&gt;a&lt;/sup&gt;, 144&lt;sup&gt;b&lt;/sup&gt;, 246</td>
<td>[8]</td>
</tr>
<tr>
<td>T</td>
<td>2</td>
<td>130, 127</td>
<td>[10]</td>
</tr>
<tr>
<td>TX</td>
<td>3</td>
<td>115, 116, 117</td>
<td>[10]</td>
</tr>
<tr>
<td>B</td>
<td>1</td>
<td>264</td>
<td>[9]</td>
</tr>
<tr>
<td>BX</td>
<td>1</td>
<td>261</td>
<td>[9]</td>
</tr>
<tr>
<td>C</td>
<td>1</td>
<td>197</td>
<td>[11]</td>
</tr>
</tbody>
</table>

<sup>a</sup> Extended from 131 feet  
<sup>b</sup> Slant borehole: total depth = 171 feet.

Fig. 1. Diesel hammer drill rig in SX Tank Farm

Each of these boreholes was extensively sampled (every 1 to 2 meters). Collection of sediment samples for laboratory analyses of the major contaminants of concern is the single most important reason for advancing a borehole through the tank farm vadose zone. These samples provide the basis of our knowledge concerning how contaminants interact with the sediments and ultimately how they move through the vadose zone. Sample collection consists of two basic approaches:  
- Split-spoon sampler driven into the formation and retrieved intact for laboratory analysis; and  
- Grab-sample collected from the drill cuttings that are returned to the surface.
Most drilling and sampling activities use both techniques. The split-spoon samples provide an exact position. The samples are maintained so that fine-grained lenses can be identified and separated if desired. The geologic fabric is maintained so that the interaction of the contaminants of concern with the geologic environment can be seen and interpreted. Grab samples are readily taken, without interfering, or slowing, the drilling process. Positions of these samples are generally less precisely known and interpretation of the geology is more difficult to make.

Besides being geophysically logged (see below), the boreholes were often used to place in-situ instruments or resistivity electrodes (see below under Surface Geophysical Logging).

**Hydraulically Driven Probe Holes**

Direct push techniques are used to rapidly and cost-effectively characterize the upper portions (typically upper 30 to 40 meters) of the vadose zone. Specific horizons in the vadose zone sediments are targeted for sampling by first pushing a hole to about 30 meters. This hole is then interrogated for moisture and/or gamma emitting radionuclides. The moisture and gamma logs are then used to identify specific zones to be sampled. Subsequent holes are advanced to sample the selected zones. These holes are then decommissioned by pulling back the casing and filling the resultant hole with bentonite clay. No drilling waste is returned to the surface using this approach, thus the generation of waste and the probability that workers would be contaminated are minimized.

Sediment samples are collected from targeted zones that were identified by the geophysical logging. To collect a sample, a special drive tip is used. When the target depth is reached, the tip is released and the drill string driven ahead. A sample, up to 0.7 meters long, is then captured inside the drill string. This sampler is then pulled back to the surface and sent to the laboratory for analysis. Samplers that can collect sediments at various depths have been developed and deployed in the tank farms.

Hydraulically driven probe holes have been placed in B, BX, C, T, TY, and U Tank Farms. Over 125 samples have been gathered and analyzed. Deep resistivity electrodes have also been placed in many of the decommissioned probe holes.

**Geophysical Logging**

New boreholes and probe holes are routinely logged (passive gamma radiation and neutron scattering to determine moisture content) immediately upon completion. In addition, there has been and continues to be a program to re-gamma log existing holes.

Gamma radiation logging has long been used at Hanford. In its early days, gross gamma logging of drywells provided a means of detecting releases from the single-shell tanks. Measurements were taken on fixed schedules that ranged from weekly to annually. Gamma logging remains an essential part of characterizing the impact of Hanford operations on the vadose zone. The emphasis has changed from detecting leaks to monitoring the stability of radiological plumes present in the vadose zone. Gamma logs of drywells are run prior to the onset of retrieval, as
needed during retrieval, and following completion of retrieval. The data derived from these logs are analyzed to assess whether or not a release has taken place during retrieval.

Two tank farms, the A Farm and SX Farm, have near horizontal tubes (laterals) that extend beneath many of the tanks in those farms (all six in A Farm and nine in the SX Farm). Routine logging of these laterals stopped in about 1984. The Vadose Zone Program revisited those laterals to reassess the distribution of gamma emitting radio-contaminants beneath the two farms. The effort is being used to provide additional evidence on the leak status of those tanks that were logged.

Neutron logging refers to the tool used to estimate the volumetric water content of soils; it is also used as a qualitative tool to identify regions of relatively high moisture content. Moisture content is an important parameter because water is the means by which contaminants are carried through the vadose zone, ultimately reaching the water table and entering the groundwater. Moisture content typically ranges from 2 to 30% by volume with 30% being near saturation for Hanford sandy sediments.

**Surface Geophysical Logging**

Surface geophysical exploration is the name that has been given to the suite of geophysical tools that generally do not use deep penetration of the ground. Surface geophysical exploration tools that have been applied in the tank farms include:

- Ground-penetrating radar,
- Electromagnetic induction,
- Differential magnetometry and
- Electrical resistivity.

Other tools have been applied outside the farms that include

- Seismic studies,
- Cross-borehole radar and seismic analyses, and
- Induced potential approaches.

Tank farm use of surface geophysical exploration has provided insight into where potential contaminants reside within the vadose zone. Surface geophysical exploration analyses are being used to direct more conventional sampling. Refined analysis efforts will hopefully lead to fuller understanding of the nature and extent of contamination originating in the tank farms.

Surface geophysical exploration has been used in WMAs T [12], U [13], C [14], S [15], and B-BX-BY [16]. Fig. 2 shows some results, including the large plume from the Tank S-104 release, which has reached groundwater.

For many of the tank farms, subsurface infrastructure (such as pipelines) restricts the depth of investigation. However, using existing boreholes, a two-dimensional representation of resistivity can be obtained. With the placement of deep resistivity electrodes, improved three-dimensional representations have been obtained even in the presence of pipelines.
Laboratory Analysis of Sediments

The laboratory analyses of collected sediments involve a tiered approach. The basic premise of the three-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, 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 (see Fig. 3), the first tier of tests (Tier I) includes those analyses that

- Provide information paramount to identifying contaminants present in the samples and
- 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 1 testing. These concentration profiles provide a measure of the extent of migration and relative mobility of each detected contaminant along the borehole length.
Tiered Characterization Approach

Tier I
- Geologic Description of the Samples
- Digital Photography of Samples
- Sediment Moisture Content
- Sediment Carbon Content
- Gamma Energy Analysis
- Sediment-Water Extracts
- Acid Extractions of the Sediment
- ICP-MS Analysis of Extracts
- ICP-OES Analysis of Extracts
- IC Analysis of Extracts

Tier II
- Particle Size Analysis
- Matric Potential Measurements
- Tritium Separations
- Strontium-90 Separations
- Actinide Separations
- Iodine-129 Extraction and Analysis
- Total Elemental Analysis
- Mineralogy

Tier III
- Batch Sorption/Leaching Tests
- Column Transport Tests
- Cation Exchange Capacity
- Specific Surface Area Measurements
- Selective Extractions
- Iron Oxide Extractions
- Solid Phase Characterization

Fig. 3. Tiered approach to sample analysis and characterization.

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. 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 [17-29].
CORRECTIVE MEASURES

The Initial Single-Shell Tank System Performance Assessment for the Hanford Site [7] estimates that future impacts from the past tank waste releases will exceed groundwater protection standards. Therefore, the Tank Farm RCRA Corrective Action Program has taken a number of steps to slow down future migration. Along with others at the Hanford Site, deep vadose zone treatability studies are being planned.

Implemented Interim Measures

Water is the major driving force to carry contaminants down into groundwater. As tank farms are located in depressions (to allow gravity to carry waste from the chemical separation plants to the tank farms), water has often collected over the tank farms in the past (particularly from melting snow). Berms and gutters have been installed to divert water away from the farms. In addition, water lines have been tested for fitness for use and those not needed have been capped outside of the farms. A major interim measure now being installed is of an interim barrier of polyurea over the subsurface plume from Tank T-106. The plume extends to 40 meters below the ground surface. This barrier, designed to last for 30 years, will allow tank waste retrievals to occur while preventing moisture from entering the vadose zone over the plume. Fig. 4 shows the footprint of the interim barrier.

Fig. 4. Interim barrier being constructed in T Tank Farm
Deep Vadose Zone Treatability

The best treatment is at the location where the contamination currently resides. Along with Fluor Hanford, Inc. and the DOE field offices at Hanford, a deep vadose zone treatability plan [30] has just been released.

CONCLUSION

Phase 1 of Tank Farm RCRA Corrective Action Program has just been completed. This effort had a strong deep vadose zone component. Phase 2 will continue the efforts at characterizing, analyzing, and remediating contamination that has been released from Hanford Single-Shell Tank Farms.

REFERENCES


