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# Deploying Technology Advancements for Characterizing the Vadose Zone in Single-Shell Tank Waste Management Areas

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

Contractor for the U.S. Department of Energy  
Office of River Protection under Contract DE-AC27-08RV14800



P.O. Box 850  
Richland, Washington 99352

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S. J. Eberlein  
H. A. Sydnor  
D. A. Myers  
Washington River Protection Solutions

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## **Deploying Technology Advancements for Characterizing the Vadose Zone in Single-Shell Tank Waste Management Areas**

Susan J. Eberlein, Harold A. Sydnor, David A. Myers,  
Washington River Protection Solutions, LLC.

### **ABSTRACT**

As much as one million gallons of waste is believed to have leaked from tanks, pipelines or other equipment in the single-shell tank farm waste management areas (WMAs) within the 200 East and West areas of the U.S. Department of Energy's Hanford Site near Richland, Washington. Although some contamination has reached groundwater, most contamination still resides in the vadose zone. The magnitude of this problem requires new approaches for soil characterization if we are to understand the nature and extent of the contamination and take action to protect the environment. Because of the complexity and expense of drilling traditional boreholes in contaminated soil, direct push characterization using a hydraulic hammer has been extensively employed. Direct push probe holes (<3-inch diameter) have been pushed to a maximum depth of 240 feet below ground surface in 200 East area. Previously gross gamma and moisture logging of these narrow probe holes was performed to identify the location of cesium-137 ( $^{137}\text{Cs}$ ) (which has limited mobility in Hanford soil) and moisture peaks. Recently a bismuth germinate detector has been deployed for detecting and quantifying the spectrum of cobalt-60 ( $^{60}\text{Co}$ ) (a more mobile contaminant), which provides additional information. The direct push system is configured to allow the collection of multiple soil core samples throughout the depth of the probe hole. The direct push unit has been used to place individual electrodes at a variety of depths as the probe hole is being decommissioned. These deep electrodes enable the use of soil resistivity measurement methods between surface and deep electrodes as-well-as between sets of deep electrodes. Initial testing of surface-to-deep electrode resistivity measurements in WMA C demonstrated significant improvement in defining the three dimensional extent of a contamination plume. A multiple-electrode string is presently being developed to further enhance the resolution of resistivity data. The combined use of direct push logging/sampling and soil resistivity measurement allows more extensive characterization of the large tank farm WMAs with less cost and time commitment than required by traditional methods. An additional tool is in the laboratory testing stage to support these investigations. A beta detection tool is being evaluated to determine if it might be deployed with the direct push unit to identify technetium-99 ( $^{99}\text{Tc}$ ) contamination.  $^{99}\text{Tc}$  is a mobile, long-lived contaminant that is the major risk driver from tank waste contamination. A screening tool to locate  $^{99}\text{Tc}$  contamination is anticipated to further increase the cost-effectiveness of vadose zone characterization efforts.

## **INTRODUCTION**

The Hanford "tank farms" are comprised of 177 large underground tanks, used to store the radioactive, chemical waste generated during the production of defense materials at Hanford. The 149 single-shell and 28 double-shell tanks are grouped into farms of 2 to 18 tanks, located in the 200 East and West areas of the U.S. Department of Energy's Hanford Site near Richland, Washington. The double-shell tanks meet current standards, and are receiving waste as it is retrieved from the single-shell tanks. The single-shell tanks are no longer considered fit for use, and the waste they contain is being retrieved and transferred to safer double-shell tank storage in preparation for closure. There are twelve single-shell tank farms, grouped into seven tank farm waste management areas (WMAs). The tanks, ancillary equipment, and associated soil in each WMA will undergo closure as a unit.

As much as one million gallons of waste is believed to have leaked from tanks, pipelines or other equipment in the single-shell tank WMAs. Although some contamination has reached groundwater, most contamination still resides in the vadose zone soil. Characterization of the vadose zone soils in the tank farms is required to perform risk assessments and support decisions on remediation of the soil for tank farm WMA closure.

The magnitude of this problem requires new approaches for soil characterization if we are to understand the nature and extent of the contamination and take action to protect the environment. Each WMA covers multiple acres. The groundwater is at a depth of 200 to 250 feet in these areas. Historic records provide some insights into leaks and releases of waste in the WMAs, but are incomplete. Migration of the contamination through the vadose zone depends on many factors (time of release, additions of water during operations, types of chemicals involved, geologic factors), so the current location of contamination plumes is not predictable by a simple model. The contaminants of particular interest are those which are both mobile and long-lived, since those contaminants will affect the groundwater and will continue to present a risk in the future. One of the greatest risk drivers in the tank farm WMAs is <sup>99</sup>Tc, a mobile, beta-emitting radionuclide.

The Hanford tank operations contractor has worked with multiple companies and national laboratories to identify enhanced characterization methods for the tank farm vadose zone. Three of the approaches being developed or deployed are improved direct push logging/sampling, improved soil resistivity measurements, and an in-situ beta detection probe.

## **ENHANCED CHARACTERIZATION METHODS**

### **Direct Push Logging and Sampling**

Drilling traditional boreholes in contaminated soil is complex, expensive, generates waste, and may lead to spread of contamination. To alleviate these problems, direct push characterization using a hydraulic hammer has been extensively employed for soil logging and sampling in the Hanford tank farms.

Direct push sampling has reached greater depths as technology has improved. Direct push probe holes (<3-inch diameter) have been pushed to 240 feet below ground surface in the 200 East tank farm area of the Hanford Site. In this area, groundwater is approximately 250 feet below ground surface, and the soil is a mixture of silty-sand, sand, and gravel. In the 200 West tank farms, a caliche layer called the Cold Creek Unit is present at a depth of 100-150 feet below ground surface (the depth varies over the several square miles of the 200 West area). Direct push probe holes have been pushed to the top of the caliche layer, but cannot penetrate it.

A disadvantage of the direct push method is that the narrow diameter probe hole does not support logging with the same spectral gamma instruments that may be deployed in a wide diameter borehole. Previously, gross gamma logging of the probe holes was performed using a sodium iodide detector. This detector identifies the location of gamma-emitting radionuclides, but cannot differentiate the energy spectra, and thus does not define which radionuclide is present. The gross gamma response is dominated by  $^{137}\text{Cs}$ , a significant radiological component of tank waste, which has limited mobility in Hanford soil.

A goal of vadose zone characterization is the tracking of mobile contamination through the soil.  $^{60}\text{Co}$  is a gamma emitting tank waste contaminant that has variable mobility in Hanford soils. Methods to detect the  $^{60}\text{Co}$  spectrum in a direct push logging hole were sought, to increase the information about contamination migration. The contractors for the direct push work (Energy Solutions, LLC, Pacific Northwest Geophysics and Three Rivers Scientific) successfully deployed a new spectral gamma logging tool employing a bismuth germinate oxide (BGO) detector to detect and quantify  $^{60}\text{Co}$ .

The BGO detector was first deployed in 241-TY tank farm in 200 West area [1]. The BGO sonde was operated with a counting time of 100 seconds per 0.5 feet. The measurements were processed to determine potassium, uranium and thorium ratios as naturally occurring radionuclides. Based on these ratios, it was possible to quantify  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  at concentrations as low as 1-2 pCi/g. A neutron moisture logging sonde was operated at a counting time of 15 seconds per 0.25 feet. The moisture data was processed and converted to percent moisture for display purposes. Fig. 1 shows the logging results of a direct push probe hole in the 241-TY tank farm. The log analysis shows both moisture and  $^{60}\text{Co}$  increasing at a depth of approximately 105 feet below ground surface. This depth is near the top of the Cold Creek Unit in 241-TY tank farm.

After completion of direct push logging, the logs are evaluated to determine where physical samples should be taken. If samples are warranted, a second probe hole is pushed adjacent to the logging hole. The direct push system is configured to allow the collection of multiple soil core samples throughout the depth of the probe hole. The logging results are used to guide depth selection for the samples. Since mobile contamination is of interest, locations of increased moisture are often selected. Spectral data indicating the presence of  $^{60}\text{Co}$  was also used to select sampling sites. Samples are analyzed for a variety of contaminants. Key indicators of tank waste are nitrate and  $^{99}\text{Tc}$ .  $^{99}\text{Tc}$  is a primary risk driver, so its location and concentration is of particular interest.

# C6915 Spectra & Moisture

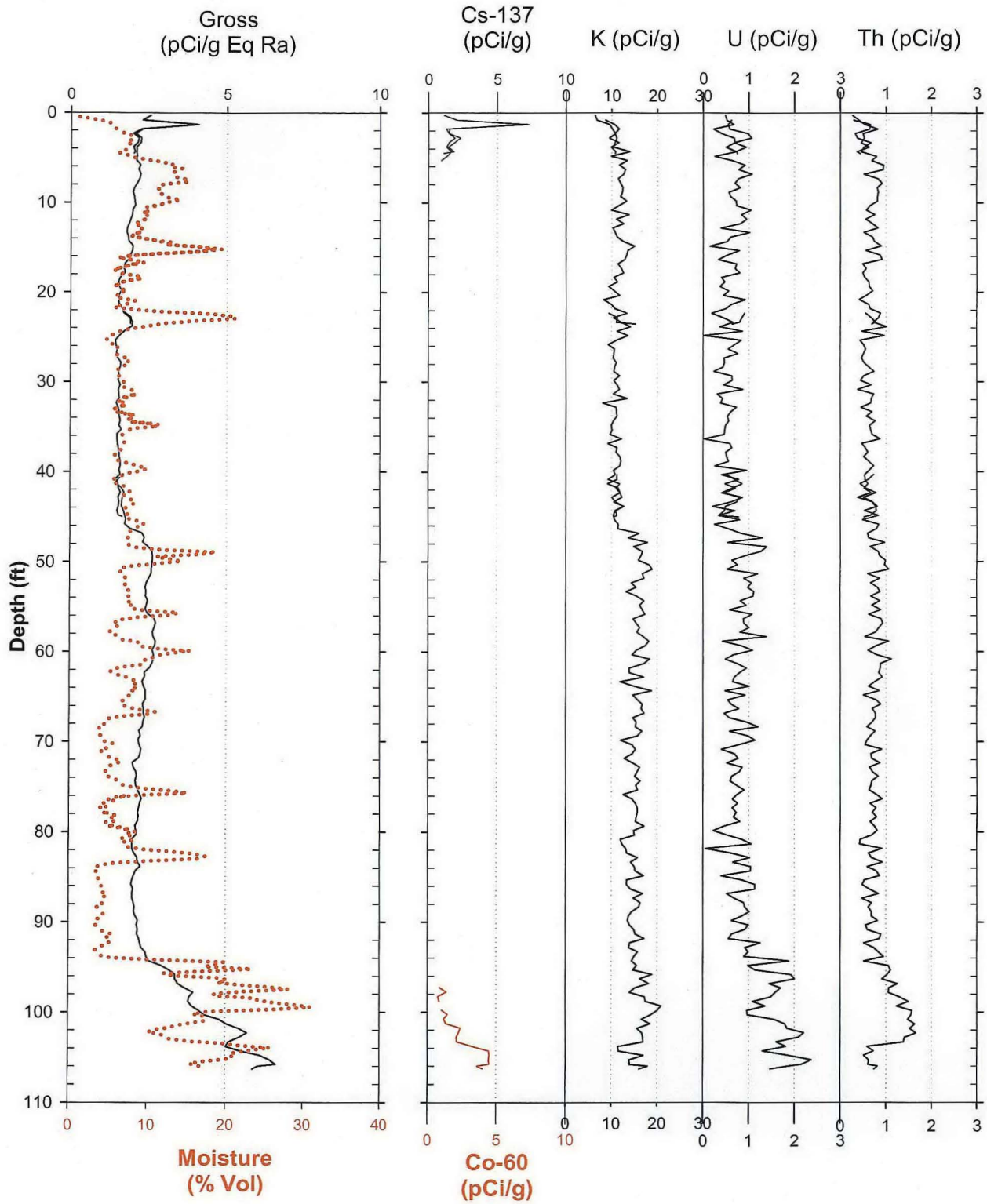


Fig. 1. Small diameter spectral gamma (bismuth germinate oxide detector) and moisture survey for 241-TY tank farm location C6195 (from RPP-RPT-41100).

For the sampling campaign in 241-TY tank farm, 12 probe holes were sampled following logging. The gamma logs of three of these 12 locations showed  $^{60}\text{Co}$  at depth. In all three cases the soil samples taken from the same depth contained more than 500 pCi/g  $^{99}\text{Tc}$ . Six of the remaining probe holes showed no detectable  $^{60}\text{Co}$  and no  $^{99}\text{Tc}$  in the soil samples (i.e. <1 pCi/g). The remaining three locations did not show detectable  $^{60}\text{Co}$ , but samples contained up to 200 pCi/g of  $^{99}\text{Tc}$ .

These initial observations suggest that the use of BGO detector will provide additional information that could support selection of sampling sites to look for  $^{99}\text{Tc}$ , but will be a useful tool only in some cases.  $^{60}\text{Co}$  concentration in tank waste streams varies, and may not be present at high enough concentrations to be detected. The half-life of  $^{60}\text{Co}$  is short, limiting the time window in which the approach will be useful.  $^{60}\text{Co}$  may also form chemical compounds that are less mobile, and therefore will not always migrate with the  $^{99}\text{Tc}$ . Even with these limitations, the addition of a tool for soil characterization is valuable.

### Soil Resistivity Measurements

In its natural state, the dry Hanford soil is not very electrically conductive. Tank wastes are concentrated sodium nitrate solutions; addition of tank waste to soil increases conductivity. Measurement of conductivity/resistivity through the soil can be used to identify areas of resistivity anomalies that could represent soil contamination. Previous applications of electrical resistivity measurements in tank farms have deployed multiple surface electrodes [2], or have used the metal casings of existing drywells [3]. These efforts have met with varying degrees of success. Limitations of the method come from sub-surface objects, such as pipelines, that offer alternative electrical conductance pathways.

The resistivity information can be improved by including measurements between deeper soil locations and the surface, or among deeper soil points. The direct push unit has been used to place individual electrodes at a variety of depths as a probe hole was being decommissioned.

Initial testing of surface-to-deep electrode resistivity measurements was performed in WMA C [4,5]. One pair of discrete depth electrodes was placed in each of two direct push probe holes near a historic contamination site called UPR-200-E- 81 (UPR designates an unplanned release). The historic leak occurred in the vicinity of a sub-surface diversion box, near two other diversion boxes and a sub-surface vault. These sub-surface structures are concrete, with associated metal piping and infrastructure. Their presence limits the capability of surface-to-surface resistivity measurements.

The discrete depth electrodes were placed at 50 feet and 150 feet below ground surface in one probe hole, and at 50 feet and 200 feet below ground surface in the other probe hole. Resistivity data was collected in multiple surface-to-depth combinations and evaluated. Fig. 2 shows the results of analysis of resistivity measurements using the depth electrodes and surface electrodes (Model 19) compared to the analysis using only the surface electrodes (Model 20). Incorporation of data from the depth electrodes demonstrated significant improvement in defining the three dimensional extent of a contamination plume. The analyses also evaluated how "a-priori" information about the site (that is, information about the physical location, size



and composition of structures such as the sub-surface diversion boxes and vaults) could be incorporated to improve the data analysis process. The greatest improvement was observed when the depth electrode information was combined with a-priori information regarding the configuration of the sub-surface structures (diversion boxes, vault and pipelines) (Fig. 2, Model 27 compared to Model 56).

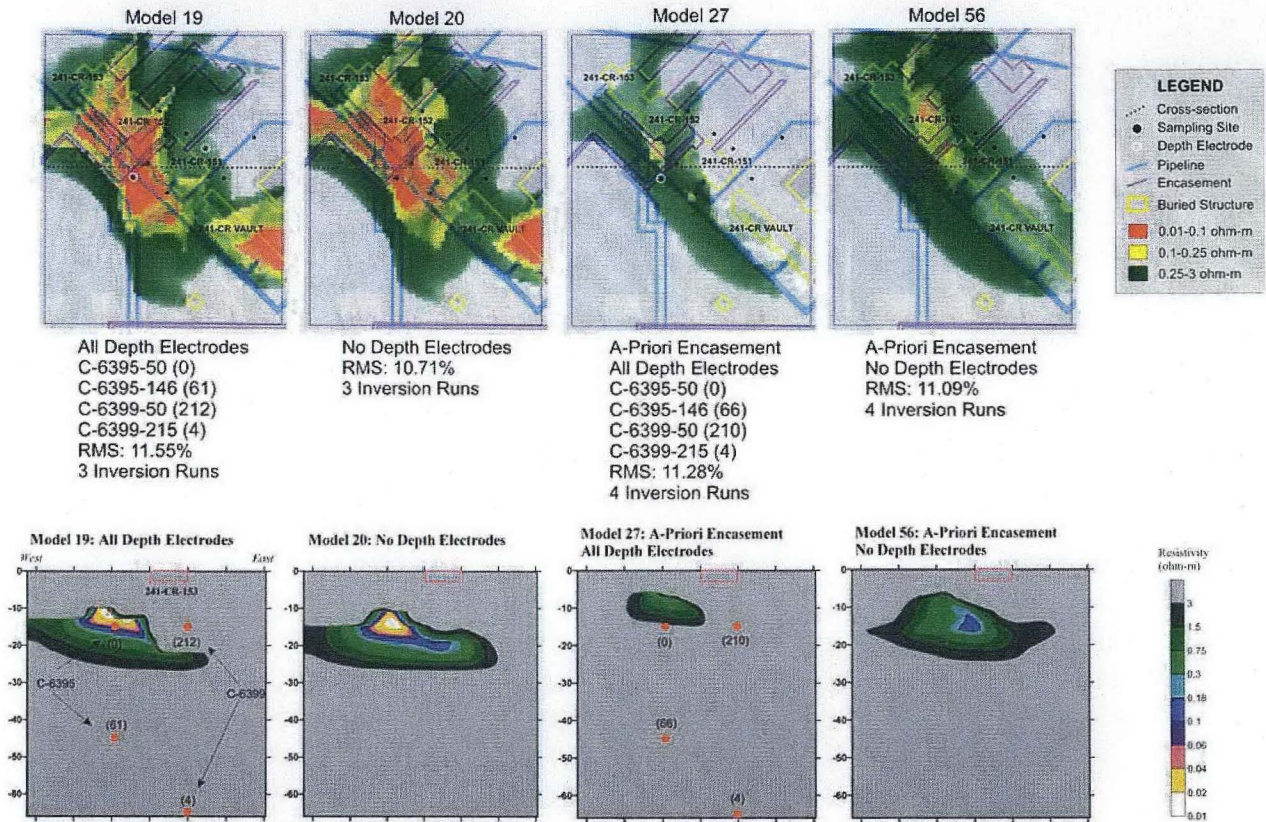


Fig. 2. Comparison of inverse model results for varying depth electrode combinations (from RPP-RPT-41236). Depth electrode locations are shown in white for plan view sections (above) and in red for cross sections (below). The quantity of associated data points are listed next to the specific depth electrode.

Although resistivity methods are not likely to provide quantitative methods for measuring soil contamination, they have already proven to provide guidance on locations for additional sampling or logging. Enhancements to the resistivity measurements will improve this capability.

### In-Situ Beta Detection Tool

An additional tool is in the laboratory testing stage to support these investigations. A beta detection tool is being evaluated to determine if it might be deployed with the direct push unit to identify areas contaminated with  $^{99}\text{Tc}$ , which is a beta emitter, but not a gamma emitter. As noted above, spectral gamma logging of direct push probe holes can provide information about the location of  $^{60}\text{Co}$ , which coincides with  $^{99}\text{Tc}$  in some cases. However, this capability is

limited. Direct in-situ measurement of the beta spectrum is the preferred means of locating  $^{99}\text{Tc}$  contamination.

Beta radiation has a short path length in air, and is readily shielded by many materials. A beta detector cannot be deployed from inside a steel encased probe hole because the steel would effectively shield it from the beta radiation. During the process of decommissioning a direct push probe hole, the steel casing is withdrawn, section by section. There is a short period of time after withdrawing a section of casing before the surrounding soil falls back into the probe hole. The deployment concept for an in-situ beta detector is to provide a probe that could be deployed into the direct push probe hole after withdrawing a section of casing. The probe would be nearly in contact with the soil, with only a small air gap.

A prototype in-situ beta detection probe was developed by North Wind, Inc. and Idaho National Laboratory (INL), based on a detector system produced by St. Gobain Crystals, to perform proof-of principle testing (Fig. 3). The beta detector is composed of two cylindrically coupled scintillators with different decay time constants. By discriminating on the pulse shape, one can distinguish interactions in either scintillator with a common photomultiplier tube. The system showed that it could readily detect the beta particles from a 100.8 nanoCurie (nCi) source of  $^{99}\text{Tc}$ . However, because of high background radiation (partially from the laboratory room used on the INL Site and partially from the natural potassium-40 ( $^{40}\text{K}$ ) in the Hanford soil), the system had difficulties measuring Hanford soil containing about 140 nCi of  $^{99}\text{Tc}$  (at a concentration in the soil of 500 picoCi/g). It is believed that slightly more complex, but readily available, electronics can be employed to significantly reduce the background so that much lower concentrations of  $^{99}\text{Tc}$  can be detected.

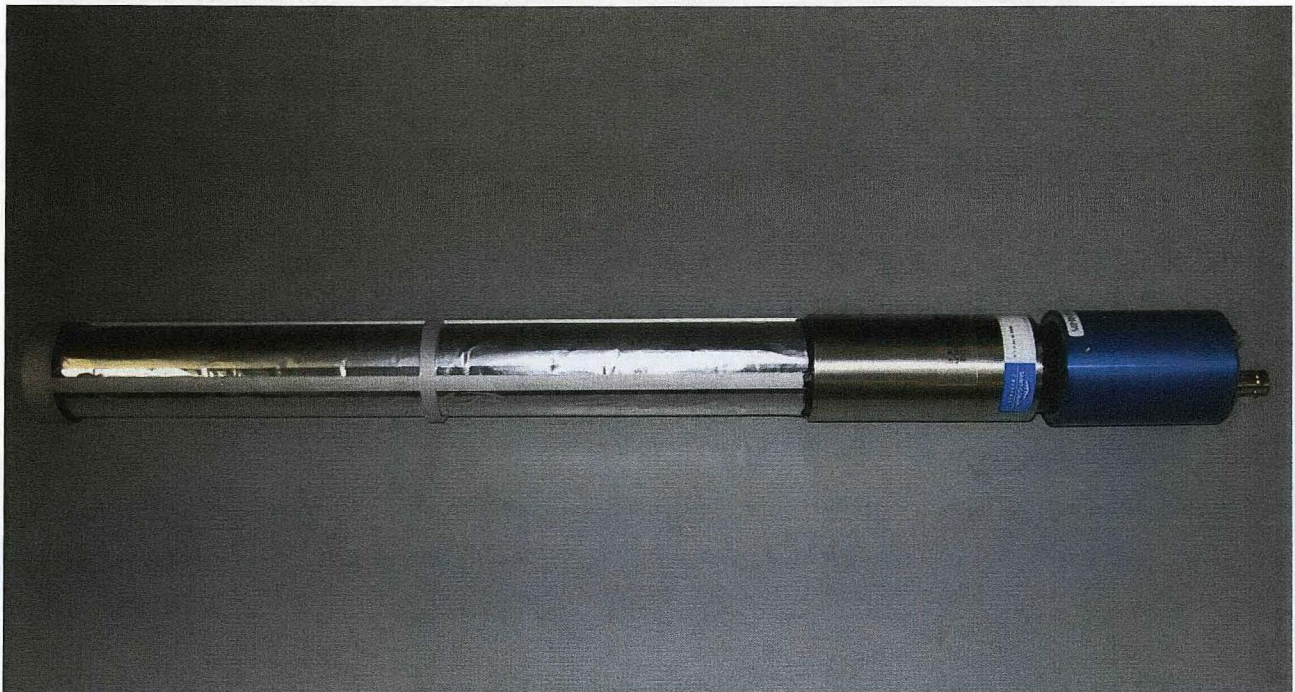


Fig. 3. Prototype in-situ beta detection probe.

## CONCLUSIONS

No single characterization method is sufficient to address the complex problem of characterizing the contamination in the tank farm vadose zone. A continued effort to add new tools and technologies, and to combine characterization methods, is showing promise. The combined use of direct push logging/sampling and soil resistivity measurement allows more extensive characterization of the large tank farm WMAs with less cost and time commitment than required by traditional methods. Increasing the types of detection capabilities that can be deployed with the direct push unit will enhance capability.

**Direct Push Logging and Sampling:** The use of a direct push sampling unit has allowed collection of soil samples in more locations at lower cost and with less waste generation than traditional borehole drilling methods. Although spectral gamma logging capabilities are limited in the narrower direct push holes, the use of the BGO detector has added some spectral gamma logging capability.

**Soil Resistivity Measurements:** Soil resistivity measurements are providing valuable qualitative information. Combining the measurements with existing information about sub-surface structures serves to improve the interpretation of the measurements. Combining the measurements with quantitative results (such as soil sample analysis) may allow a semi-quantitative interpretation of the resistivity results. Use of both surface and deep electrodes for collecting resistivity data will improve the depth differentiation of soil anomalies.

**Beta Detection Tool:** More development is needed to construct a field-deployable beta detection tool, but initial testing indicates that the concept has merit.

## RECOMMENDATIONS

Recommendations for future work include the following.

### Direct Push Logging and Sampling:

- Continue deployment of the BGO spectral gamma detector for direct push logging in areas where  $^{60}\text{Co}$  may be present.
- Continue to review gamma logs and moisture profiles as sampling locations are determined; perform systematic comparisons of sample results to the logs and profiles to refine future sample selection locations.

### Soil Resistivity Measurements:

- Place deep electrodes during direct push probe hole decommissioning, to enable future resistivity measurements. Continue to evaluate the contribution of deep electrodes to the overall quality of the resistivity evaluations.
- Develop and deploy a multiple-electrode string to further enhance the resolution of resistivity data.
- Integrate existing information on sub-surface structures to improve resistivity data evaluation.

Beta Detection Tool:

- Continue laboratory testing and development of a beta detection tool to locate <sup>99</sup>Tc.
- Develop an enhanced probe configuration to increase the signal to noise ratio when deployed in soil containing <sup>40</sup>K.
- Configure the system for deployment with the direct push unit and perform a field test.

Implementation of these recommendations is anticipated to further increase the cost-effectiveness of vadose zone characterization efforts.

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