SMART 3-D CHARACTERIZATION OF SUBSURFACE CONTAMINATION AROUND THE BELOW GRADE DUCTS AT THE BROOKHAVEN GRAPHITE RESEARCH REACTOR

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ABSTRACT

During operation of the Brookhaven Graphite Research Reactor (BGRR), the Below Grade Ducts provided passage for cooling air flowing through the graphite reactor pile to the exhaust stack. Following shutdown of the reactor in 1968, the ducts and fuel transfer canal collected water and were a potential source of contamination to the soils beneath the facility. Thus, thorough subsurface soil characterization is required to determine the location and extent of potential contamination to facilitate appropriate remedial action planning for the below grade facilities and surrounding soils. The characterization approach used in this project integrates several innovative technologies to provide improved characterization and data interpretation. The innovative technologies include the use of a small footprint Geoprobe to install sampling ports in the soil and beneath the buildings, perfluorocarbon gas tracers (PFTs) to define the leak pathways, three-dimensional (3-D) visualization tools to investigate the data, the In Situ Object Counting System (ISOCS), for rapid field gamma surveys of soil samples, and BetaScint, for beta surveying of soil samples. A Frisch chamber alpha-spectroscopy device will be used for alpha-emitting radionuclides. Initial data and analysis from the PFT leak detection study are presented. The outcome of the data analysis is a three-dimensional (3-D) visualization of the potential leak pathways from the below grade ducts. The information from the PFT study will be used to optimize subsurface soil data collection in the second phase of this program. In addition, subsurface soil characterization data beneath the Fuel transfer Canal House are presented.

INTRODUCTION

The Brookhaven Graphite Research Reactor (BGRR) was the world's first research reactor constructed solely for study of peaceful uses of atomic energy. After operating from 1950 until 1968, the BGRR was placed in a safe shutdown mode. It is currently on an accelerated decommissioning schedule with a completion date projected for 2005. During operation, the reactor was air-cooled. The air was vented through an exhaust plenum, subsurface air ducts, through a filter, and out a 320-foot high exhaust tower. During operation, fuel failures released radioactive contamination through the exhaust system. After closure of the BGRR, rainwater intrusion into the ducts and plenum has been observed. Leakage of contaminated rainwater out of the ducts leading to potential subsurface contamination is suspected. Figure 1 displays the BGRR complex and illustrates the approximate location of the below grade ducts.

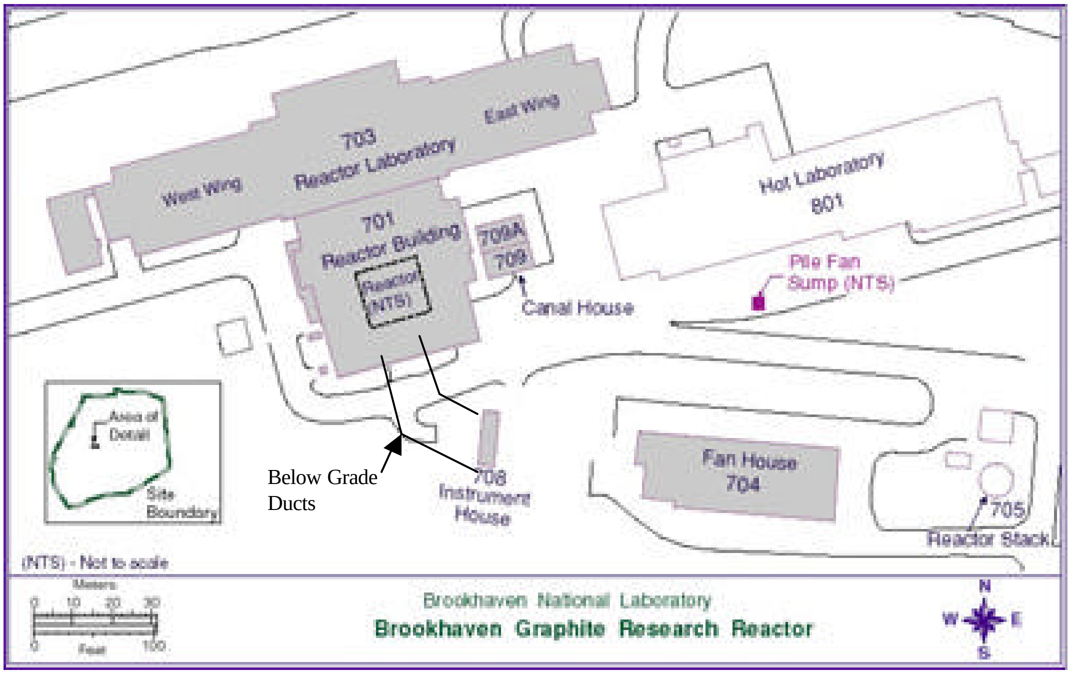

Fig. 1. Surface view of the BGRR complex and approximate location of below grade ducts.

A major issue facing the BGRR Decommissioning Project involves determining the location and extent of contamination on the exterior of underground ducts and canals and in the soils beneath these structures. While it is likely that most of the soils and substructure exterior surfaces are not contaminated above site cleanup levels, the current baseline calls for complete removal of all the subsurface structures and three feet of soil beneath them. As part of this Accelerated Site Technology Deployment (ASTD) program sponsored by the U.S. Department of Energy Decontamination and Decommissioning Focus Area (DOE DDFA), innovative technologies are being deployed to complete subsurface characterization and provide a visualization of the levels of soil contamination below the underground structures at the BGRR. The objective of this program is to obtain a risk-based rationale for leaving the ducts and canal in place in regions where contamination is not present. The innovative technologies include the use of:

- A small footprint Geoprobe to install sampling ports in the soil and beneath the buildings
- Perfluorocarbon gas tracers (PFTs) to define the leak pathways
- EVS-PRO a three-dimensional (3-D) data visualization and analysis tool
- In Situ Object Counting System (ISOCS), for rapid field gamma surveys of soil (core) samples
- BetaScint, for beta surveying of soil (core) samples
- Frisch chamber alpha-spectroscopy device for alpha-emitting radionuclides.

The innovative technologies will permit an increased quantity of characterization data. This will be combined with graphic 3-D visualizations of the extent of contamination, which will result in more rapid decision-making and consensus building with external stakeholders about the need for complete substructure removal. If the data supports leaving the ducts in place, the potential cost savings to the project is $3.4 million.

CHARACTERIZATION OF POTENTIAL LEAK PATHWAYS

As part of the overall characterization efforts, state-of-the-art gaseous tracers have been applied to determine leak pathways from the ducts to identify which soil regions under or adjacent to the ductwork should be emphasized in the soil characterization process. Brookhaven National Laboratory (BNL) has developed a suite of PFTs (1,2), which were originally used in atmospheric and oceanographic studies and have since been applied to a great variety of problems. These include subsurface barrier continuity verification (3), detecting leaks in buried natural gas pipelines, and locating radon ingress pathways in residential basements (1). PFTs have regulatory acceptance and are used commercially (e.g. detecting leaks in underground power cable systems) (2). PFTs allow locating and sizing of leaks at depth, have a resolution of fractions of an inch, and have been used in a variety of soils (3).

At the BGRR, the subsurface ducts are approximately 170 feet long, 20 feet in height and expansion joints occur approximately every 40 feet. There are two separate exhaust ducts in the system. Each subsurface duct consists of a primary carbon steel duct, which was used to exhaust the air, and a secondary duct, which jacketed the primary duct and was used to cool the air in the primary duct by flowing air in the opposite direction of the exhaust air. Although, the primary and secondary ducts were originally distinct, corrosion over the years has lead to communication between the two. The outer duct is surrounded by approximately one foot of concrete. Contamination of soils is expected to coincide with the leak pathways out of the outer subsurface air duct and concrete. The most likely leak pathways will occur at the expansion joints along the duct as determined from internal video surveillance of the ductwork. However, the video surveillance is not accurate enough to rule out other pathways. Therefore, PFT tracers were used to determine all potential water pathways. Because PFTs can be detected at extremely low levels (10 parts per quadrillion is routine), very small leaks are easily identified. Testing for leaks in the BGRR underground duct was accomplished by injecting the PFT(s) inside of the secondary air duct and monitoring for that tracer(s) in the soil surrounding this duct. The location and quantity of tracer detected outside of the duct determines the location and the size of any leaks. Larger openings in the duct mean that greater concentrations of tracer are transported out more rapidly. A detailed discussion of the use of PFT's to determine leaks at the BGRR can be found in a companion paper presented in these proceedings (4).

Although a gas tracer is not an exact analogue for the water leakage pathway, the probability of contamination occurring is highest in the areas defined by leakage of the PFT tracer. Stated differently, if gas does not leak through an area, there is high confidence that contaminated water will not leak through that area. The probability for water leakage is higher along the bottom of the ducts and decreases as one nears the top of the ducts. For this reason, the baseline soil characterization efforts emphasize the regions adjacent to and below the expansion joints. However, the use of the PFT tracers determines more precisely and with higher confidence the areas where leakage can occur.

A more exact determination of leak pathways has several advantages. The use of PFTs will determine whether the suspect areas are in fact leaking (and the relative magnitude of the leaks), but more importantly will determine if any additional areas of the duct are leaking (e.g., significant cracks in the concrete duct). If additional leakage is found, additional sampling in this region can be performed to improve the subsurface characterization. This will allow the regulators and stakeholders to have more confidence in the sampling scheme that emphasizes suspect/known leak pathways and uses exploratory sampling elsewhere. Another advantage to using PFTs is that they may be able to eliminate some of the suspect contamination pathways by determining that they are not leaking. We can then perform exploratory type sampling in these areas saving considerable funds.

Small Footprint Geoprobe

Geoprobe Systems has developed a compact subsurface soil investigation system capable of driving standard penetrometer type probes and sampling devices, including continuous soil sampling systems capable of retrieving discrete core samples. The Geoprobe Model 54 is a 31-in. wide unit that will fit within standard doorframes and other restricted space areas where standard penetrometers would not be able to fit. The unit uses hydraulics to hammer and/or push a metal sampling tube into soil and then withdraws the sample to the surface. Typical sampling tubes allow 1.25-in. or 2-in. diameter cores to be taken. The length of the sample that can be taken with each withdrawal is two feet. The system weighs less than 725 pounds and is equipped with wheels so it can be moved around similar to a cart. The unit is connected to a remote hydraulic power unit using flexible hydraulic hoses.

The compact Geoprobe will be used to augment samples taken using conventional soil boring techniques. It has been lowered into the spent fuel canal area to take samples directly underneath sumps and joints suspected of leaking. This same unit can also be used to place supplemental boreholes for further characterization should the sampling optimization show areas requiring more data to reduce uncertainty or if unexpectedly high contaminant levels are found in any area.

The boreholes used to install the PFT monitors were placed using the small footprint Geoprobe Model 54 LT unit. Boreholes were placed along both sides of the ductworks and along the central axis of each individual duct. The monitoring wells along the sides each have multiple sampling ports evenly spaced vertically such that the first port is at the same elevation as the top of the ductwork and the last port at the same elevation as the bottom of the ductwork. The distance from the duct to the surface changes with the southern end only 2 feet below-grade while the northern end, near the main building, is approximately 15 feet below grade..

Use of EVS-PRO for PFT Data Interpretation

Once a complete data set for one day of PFT sampling has been analyzed, the information is input into C Tech's EVS-PRO for visualization (5). The software unites interpolation, geologic modeling, geostatistical analysis, and fully 3-D visualization tools into a software system developed to support environmental contamination issues. One of EVS-PRO's strengths is its integrated geostatistical analysis, which provides quantitative assessment of the quality of site characterization.

Figure 2 presents representative data for the tracer PMCP at the south duct on February 12, 2001, 5 days after the start of the injection in the internal duct. Evidence of PMCP in the surrounding soils indicates a leak pathway from the internal duct. The diagram contains the façade of the building, windows, which are represented in blue, the supports for the building, the underground ducts, and the sample locations. Sample concentrations are color coded where red denotes the highest concentration and blue the lowest. Peak concentrations were approximately 15 ppb.

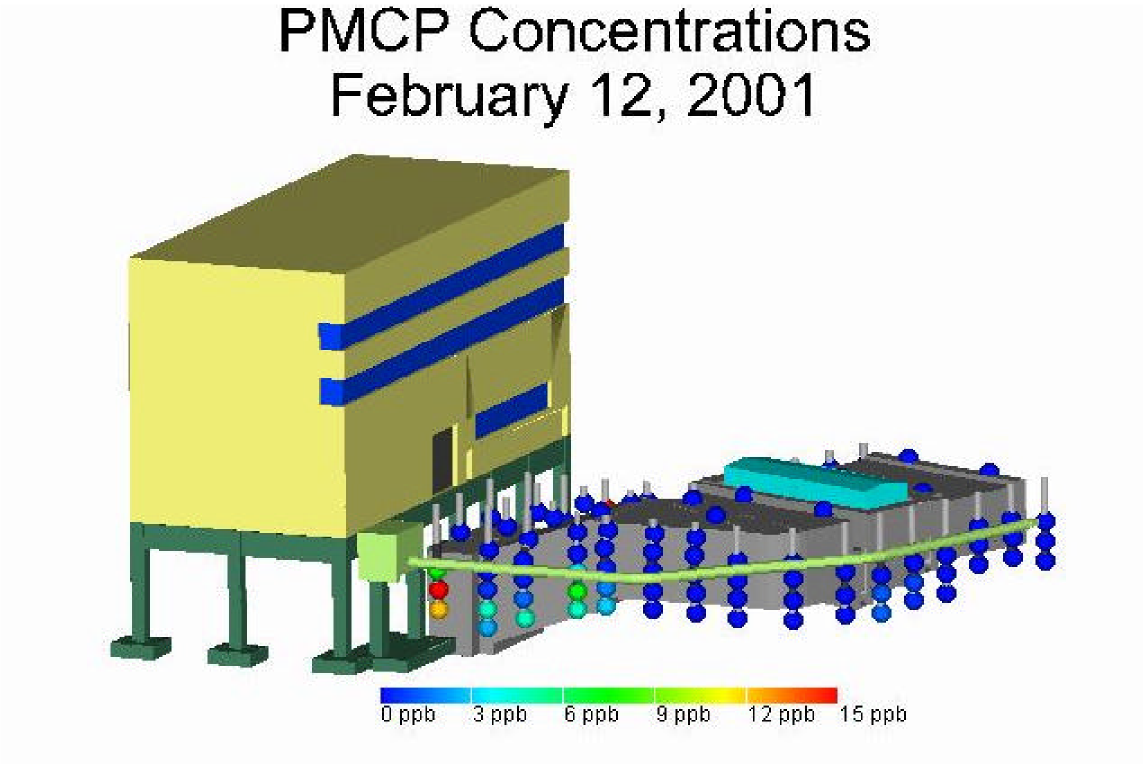

Fig. 2. PMCP measured concentrations at the South Duct on February 12, 2001

This is close to 10% of the value found in the injection loop inside of the ducts, which indicates that a substantial hole exists in the duct at this area. This peak value was found in the first well from the building at a depth of approximately 27 feet below grade near the bottom of the duct. As was mentioned earlier, regions of minimal or no leakage are depicted in blue. The data clearly indicate that there are substantial areas where leakage is minimal. This would suggest soil characterization should be focused on the areas with the highest leak rates. If a region is not susceptible to gas leakage, it is not susceptible to water leakage.

Figure 3 presents a representative data set for the tracer PMCP at the north duct on February 12. In this figure, the building is not shown to provide a clearer view of the data. There are several indications of leaks at this duct and the concentrations are typically higher than on the south duct. The peak concentrations on this side of the duct were 50 ppb, approximately 1/3 of the value in the ducts and again indicate a substantial size flaw in the duct allowing release of the gas. Values near 50 ppb were detected in the first well away from the building and in one other well.

EVS-PRO was useful in determining the leak pathways in the following areas:

- *Data Interpretation:* Determination of the spatial distribution of the PFT as a function of time.

- *Communication:* Visual presentation of subsurface ducts and PFT concentrations is critical for effective communication. EVS-PRO was used to integrate geologic information, PFT concentration data, and site maps (showing buildings and the ducts in 3-D).

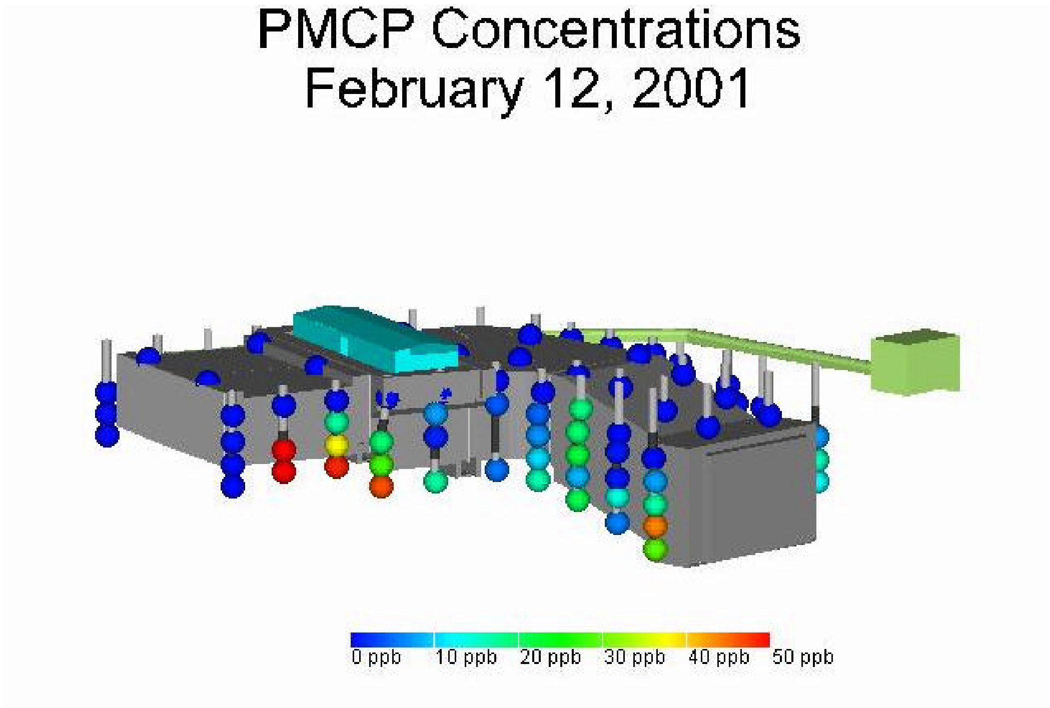

Fig. 3. PMCP concentrations at the North Duct on February 12, 2001.

SOIL CHARACTERIZATION SAMPLING PLAN

Based on process knowledge of the history of the BGRR, visual inspection, and the PFT leak characterization data, a subsurface soil-sampling plan will be developed. During operation of the BGRR, fission products were released through the ducts due to fuel failures. Contaminants detected in the exhaust system included Cs, Sr, Co, and transuranic elements such as Pu. The objective of the sampling plan will be to delineate any region that has soil contamination at levels above pre-specified soil cleanup levels for every contaminant.

Grab samples will be taken at locations determined from the sampling plan. These samples will be brought to the surface and measured using two field-deployable radiation detection systems, ISOCS (gamma) and BetaScint (Sr-90) that were successfully deployed at the BGRR in FY 00 (6). Alpha-emitting radionuclides will be evaluated through the Frisch Chamber Alpha Spectroscopy. The spot samples will be used to determine what areas (boreholes) under the duct should be surveyed first, which boreholes are candidates for full surveys (gamma, beta, and alpha) and which boreholes are expected to be clean. If Sr-90 in the groundwater is an issue, the 3M Empore Disk system will be used.

The data from ISOCS, BetaScint, and other sources will be analyzed using EVS-PRO. Similar to the PFT studies, EVS-PRO will be used to optimize sample location, define zones of contamination above the cleanup levels, quantify the uncertainty in the size of the remediation zones, and visualize the data to facilitate communication with other interested parties.

In Situ Object Counting System

Traditional gamma spectroscopy has meant a major investment for purchase and eventual disposal of a variety of calibration sources, matching the geometry and matrix of the expected contaminated medium. For each new geometry, a new calibration standard and hours of calibration is required. This has limited *in situ* gamma spectrum analysis to simple geometries and contamination distributions. The In Situ Object Counting System (ISOCS), developed by Canberra Industries, Inc., overcomes these problems. The ISOCS system is equipped with a broad energy-intrinsic germanium detector capable of covering energies from 3 keV to 3 MeV with high efficiency and resolution. Its sensitivity allows easy detection of both low-energy spectra associated with transuranic isotopes and higher energy fission product spectra. The ISOCS unit is mounted on a field-deployable cart and includes shielding and collimators, a battery powered multi-channel analyzer, and a portable computer.

The versatility of the ISOCS system has been demonstrated in numerous situations during initial characterization and decommissioning efforts at the BGRR. Surface soil detection sensitivities of less than 1 pCi/g have been attained with count times as short as 10 minutes for common gamma emitters such as Cs-137.

BetaScint

Strontium 90 (Sr-90), a fission product commonly associated with nuclear reactors, is a pure beta emitter and thus is not directly detected by gamma spectroscopy. Conventional Sr-90 analysis requires chemical separation of the strontium from the sample matrix, followed by in-growth of the Yttrium 90 (Y-90) progeny for analysis, a time consuming procedure that often takes 1 - 4 weeks. The BetaScint system consists of a multi-layer beta scintillation detector array with a beta radiation entrance window measuring 30-cm by 60-cm. Scintillating fibers are fashioned into ribbons, which are stacked vertically. Soil samples are prepared, transferred to large area counting trays, and positioned beneath the detector window for analysis. Beta particles that pass through the detector window excite electrons in the scintillating ribbons resulting in the emission of light pulses, which are counted by photomultiplier tubes. Coincident circuitry to detect simultaneous events in several ribbon layers distinguishes high-energy betas (Sr-90) from lower energy contaminants and background.

The cost of conventional baseline Sr-90 analysis (including transportation) is approximately \$200/sample and usually requires 2 - 4 weeks. BetaScint analyses cost about \$20 -\$30/sample and can be completed within minutes of sample collection. Additional cost savings are realized because the equipment is already available at BNL. In addition, the BetaScint sensor evaluates a relatively large surface area (~2,000 cm^2), so that results are more representative than those obtained from typical sample aliquots of several grams of material.

This system can measure Sr-90 and U-238 at approximately 1 pCi/g above background with a 5-minute count time.

CHARACTERIZATION OF SOIL CONTAMINATION AROUND THE SPENT FUEL CANAL

Although soil characterization has not started underneath the ducts, it has been started in the soil surrounding the spent fuel transfer canal (Figure 1). Operational history indicates contamination may have been released in this area. The primary contaminant of concern is Cs-137. Soil characterization was performed using a 5 meter grid at the surface and measuring Cs-137 contamination using ISOCS and standard laboratory techniques. Soil samples were collected at the surface, and at several discrete depths down to two feet below grade at all locations. At selected locations, deeper soil samples were collected and analyzed. In the spent fuel canal, the small footprint geoprobe was lowered into the canal and used to drill into the subsurface and collect soil samples. Sampling focused on the areas around sumps and joints in the building.

Complete characterization has not been obtained at this point and sampling is continuing. EVS-PRO has been used to evaluate the soil contamination around the spent fuel canal. The results of this work are presented in Figure 4, which depicts a plan view of the area with a surface map of the footprint of buildings. Each bore location is labeled. Data is represented by a blue sphere if the measured value is less than 23 pCi/g, the soil free-release limit, or a red sphere if the concentration is above the limit. The results of this analysis indicates that there are localized areas with Cs-137 levels above the site free-release limit of 23 pCi/g. Most of these areas are under the canal building. One area above the cleanup limit exists beneath a sump at the edge of the building.

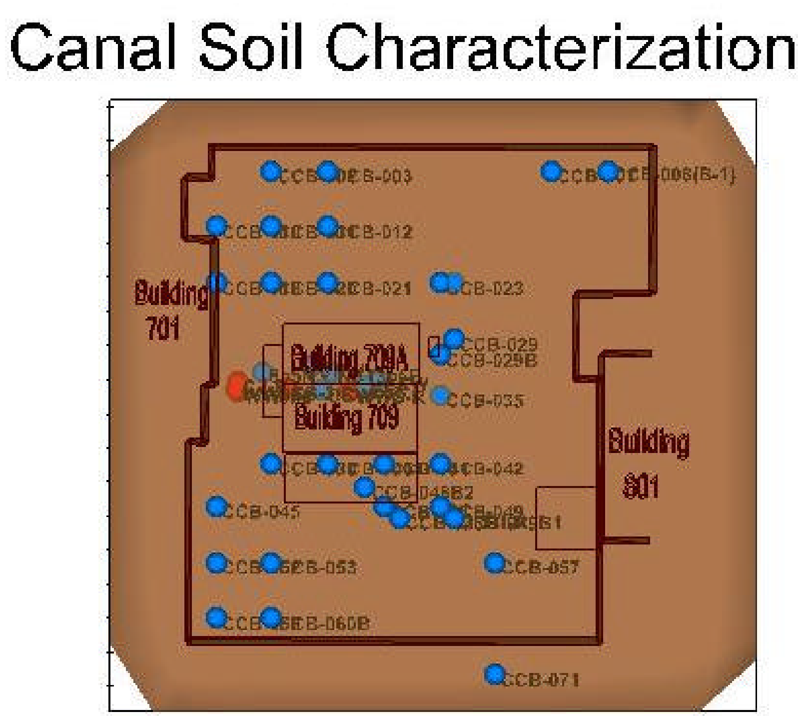

Figure 4. Plan view of spent fuel canal buildings (Buildings 709 and 709A) and soil sample locations. Samples below the soil cleanup guideline are in blue. Samples above the guideline are in red.

Figure 5 shows a side view to provide a perspective on depth. In this view, the interface between the two brown shaded areas represents the elevation of the foundation of surrounding buildings. Excavation of soil becomes difficult at depths greater than this. The blue layer represents the water table, which is approximately 67 feet below grade. The contamination remains well above the water table at all measured locations. Additional data are being collected beneath areas of high concentration to confirm that contamination has not reached the water table. One advantage of this visualization is to demonstrate the highly localized nature of the contamination. High levels of contamination extend only a few feet in any direction parallel to the ground surface. This is confirmed by adjacent sampling a few feet from the hot spots; these show up clean. High levels of Cs-137 concentrations also correlate with sumps and joints in the building. Cs-137 concentrations in randomly selected areas beneath the canal building were always below the free-release criteria.

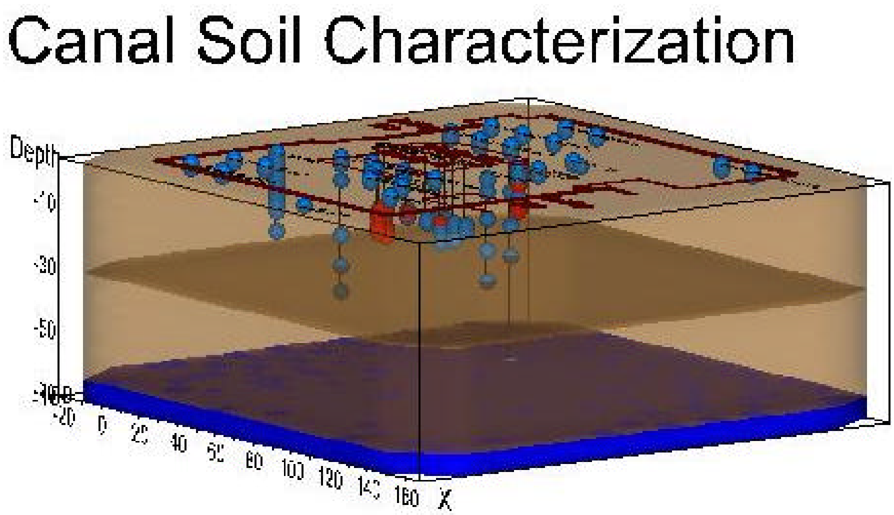

Fig. 5. Side view of spent fuel canal buildings and soil sample locations. Samples below the soil cleanup guideline are in blue. Samples above the guideline are in red. The blue layer at the bottom represents the water table.

CONCLUSIONS

A major issue in the decontamination and decommissioning of the Brookhaven Graphite Research Reactor is the potential for soil contamination underneath the underground ducts and transfer canals. Water was used in the canals to provide shielding from radioactivity from spent fuel that was transferred through the canals. Rainwater was known to enter these ducts in the 30-year period after shutdown of the reactor. Although much of this water was retained in sumps, it is suspected that water also leaked out along the ducts. A goal of this study is to deploy innovative technologies to improve subsurface characterization. The first phase of this ASTD study was to use gas tracers (PFTs) to help define the most likely leak pathways. These tests have been conducted and the data are being analyzed. Preliminary review of the data using EVS-PRO indicates several regions that are likely places to look for soil contamination. Based on the final analysis of the PFT data and site operational experience, a sample analysis plan will be developed for soil characterization. The soil characterization will use additional innovative technologies including ISOCS, BetaScint, and Frisch Chamber Alpha Spectroscopy. EVS-PRO was also used to interpret and visualize the data from subsurface characterization of soils around the spent fuel transfer canals and will be used in a similar manner for the soils beneath the below grade ducts. Placing the data in a spatial context is valuable for data interpretation. Ultimately, it is hoped that the improved characterization achieved through the innovative technologies will lead to better, more defensible decision-making for determination of which parts of the subsurface structures can be left *in-situ* and which parts need to be removed.

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