

**DEPLOYMENT OF *IN SITU* MEASUREMENT TECHNIQUES AND THE MARSSIM  
PROCESS FOR CHARACTERIZATION OF THE BROOKHAVEN GRAPHITE  
RESEARCH REACTOR**

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**ABSTRACT**

This paper describes a DOE Accelerated Site Technology Deployment project being conducted at Brookhaven National Laboratory to characterize the Brookhaven Graphite Research Reactor facility, which is currently undergoing decontamination and decommissioning. The MARSSIM process is being implemented to provide guidance for survey planning and data evaluation. Innovative *in situ* analytical techniques are being deployed to quantify the type and extent of radiological contamination including ISOCS (Canberra Industries, Inc.) for gamma emitting radionuclides and BetaScint (BetaScint, Inc.) for Sr-90. These techniques provide a number of advantages compared with conventional characterization methods including near real-time data, ability to evaluate inhomogeneous materials, fewer samples required, and lower radiation dose exposure to personnel. Data has successfully been acquired and evaluated for several BGRR facilities and components including the Pile Fan Sump (PFS), underground piping for the PFS, parking lot areas, Above Ground Ducts, and contaminated cooling fans. Cs-137 is the predominant gamma-emitting radionuclide identified, with smaller quantities of Co-60 and Am-241 detected.

**INTRODUCTION**

The Brookhaven Graphite Research Reactor (BGRR) is a graphite-moderated, air-cooled, thermal neutron research reactor that operated at Brookhaven National Laboratory (BNL) from 1950 through 1968. Following shutdown, fuel was removed and the facility has been maintained in a safe shutdown mode since then. Many of the major BGRR sub-components are currently scheduled for near-term decontamination and decommissioning (D&D) including the Pile Fan Sump, above and below ground air ducts, and auxiliary buildings that house fans, filters, instruments, fuel transfer canal and water treatment systems. Figure 1 is a photograph of the Above Ground Ducts and Fan House that drew air through the reactor core for cooling. The Canal House used to facilitate removal of spent fuel and equipment is shown in Figure 2. Characterization of these facilities prior to, during, and after

dismantlement is required to minimize worker exposure, plan for appropriate disposition of materials and remaining facilities, and demonstrate compliance with applicable environmental regulations.



**Figure 1** Above ground ducts and Fan House at the BNL BGRR



**Figure 2** BNL BGRR Canal House structure used for removal of spent fuel and contaminated equipment

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Conventional baseline characterization requires the collection of thousands of surface smear, volumetric, and core samples, sending samples for on and off-site analysis, compiling the information in a database, and reviewing the data for quality assurance. Many of the areas requiring characterization are not readily accessible and/or are highly contaminated, further complicating the process. Thus, in addition to being time consuming and costly, the baseline characterization approach can result in excessive radiation exposures to personnel. This paper describes a project sponsored by the DOE Office of Science and Technology (EM-50) under the Accelerated Site Technology Deployment (ASTD) initiative to deploy state-of-the-art techniques and equipment for improved characterization of the BGRR. The approach includes utilization of the innovative Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) process and characterization using *in situ* measurement and analyses.

The MARSSIM approach provides guidance on planning, conducting, evaluating, and documenting environmental radiological surveys of soil and building materials to optimize the sampling process and demonstrate compliance with regulations. The final MARSSIM was published in December 1997 and has already attracted considerable interest from the D&D community. However, there is little experience in applying the MARSSIM methodology at actual sites. In those cases where it has been used, the emphasis has been on the final status survey design; its application for D&D characterization is novel. The MARSSIM process involves identifying Data Quality Objectives (DQO) to establish the types of data needed and the confidence levels required. Data validation and verification, as well as data quality assessment, are addressed through implementation of a Quality Assurance Project Plan. Valuable resources and time are saved by focusing on the proper data needed for evaluation.

Conventional gamma spectroscopy requires a major investment for purchase and eventual disposal of a variety of calibration sources that match the geometry and matrix of the expected contaminated medium. For each new geometry, a new calibration standard and hours of instrument calibration are required. This has limited *in situ* gamma spectrum analysis to simple geometries and contamination distributions. 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.

Measurement of gamma emitting radionuclides is being accomplished using a field deployable gamma spectrometer (*In Situ* Object Counting System or ISOCS) manufactured by Canberra Industries, Inc. The battery-operated system provides traditional spectra of counts as a function of gamma energy, which are then converted to radionuclide concentration by applying pre-defined geometry templates in the analysis software. Thus, complex contamination distributions (e.g., an inaccessible contaminated pipe within a wall) and resulting quantification of the contamination therein can be identified.

Detection of Sr-90 is being accomplished by means of a field deployable high energy beta scintillation detector manufactured by BetaScint, Inc. This system, can measure Sr-90 and U-238 at approximately 1 pCi/g above background with a 5-minute count time. Soil samples of 2 -3 kg are collected, analyzed using the BetaScint system and then quantified based on data from a series of known standards prepared using similar media. Measurements are conducted in a field laboratory set up in close proximity to soil removal operations while D&D activities are continuing.

## APPLICATION OF THE MARSSIM FOR CHARACTERIZATION

The MARSSIM approach emphasizes the use of statistical planning and data analysis for demonstrating compliance with a final status survey. There are few examples of how to apply the DQO process for other types of surveys where such formal analyses are not necessary, or even appropriate. One of the objectives of this project is to provide a concrete example of how the MARSSIM methodology can be applied to characterization surveys and to develop a framework for the design of characterization survey plans that can be used to implement the MARSSIM at the BGRR and other DOE sites.

The DQO process is the basis for the performance-based guidance in planning MARSSIM surveys. The steps of the DQO process specified in the MARSSIM include:

1. state the problem
2. identify the decision
3. identify the inputs to the decision
4. define the boundaries (spatial and temporal)
5. develop a decision rule
6. specify limits on decision errors
7. optimize the design for collecting data

Through the implementation in this project, the first four steps of the DQO process are common to both characterization and final status surveys. In the final three steps, there is significant difference in interpretation and application to the characterization survey.

The fifth step in the DQO process is the specification of a decision rule. For the final status survey this usually takes the form of a statistical hypothesis test. For a characterization survey such a highly structured rule will not generally be appropriate. However, it should be possible to identify:

- (a) a range of results that clearly indicates that there is no need for remediation in an area,
- (b) a range of results that clearly indicates that there is need for remediation in an area, and
- (c) an intermediate range of results that may indicate the need for more data before a decision is made.

Such a scheme is loosely patterned after sequential testing procedures, but is primarily intended to differentiate the easy decisions from the more difficult ones so that more resources can be devoted to the areas that need it.

Specifying the acceptable limits on decision errors is the sixth step in the DQO process. For final status surveys, this means specifying Type I and Type II error rates for statistical hypothesis tests. Again, such precision is usually neither desirable nor necessary in a characterization survey.

In a final status survey, the decision errors are used to determine the number of samples it is necessary to collect. The same is true for the characterization survey, except that extensive use of professional judgement must be made to balance the costs of additional measurements against the risk of drawing the wrong conclusion from the data. Optimizing the design of a characterization survey (step seven of the DQO process), involves using all the information available, together with professional judgement, to assess the worth of the information to be gained from additional data in terms of increasing confidence in a remediation decision. This is where the width of the "gray region" expressed by choice (c) of step 5 is used to separate, as efficiently as possible, the easy decisions from the difficult ones. The cost of data

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collected early in the characterization can be balanced against the possibility that new data will be needed. The consequence of incorrectly classifying an area as needing remediation when it does not should be balanced against the cost of discovering during a final status survey that an area thought to be clean actually is not. Remediation costs are also balanced against the cost of characterization measurements.

The seven specific elements of the overall DQO process, outlined above, are addressed by the ASTD project team through development of individual project-specific survey plans (PSSPs) in support of individual BGRR D&D campaigns. The PSSP considers the goals of the intermediate D&D objective, the baseline characterization elements, and the targeted components of the facility, to identify the scope and content of the *in situ* characterization efforts using the DQO process. The PSSP provides details on field of view, shielding, and detection levels necessary for the *in situ* evaluations and identifies sample number designations for items and views of items for tracking and reporting purposes.

Continuity of spectrum analysis and interpretation among the PSSPs is assured by compliance with the ASTD Project *In Situ* Analysis Quality Assurance Project Plan (QAPP).<sup>1</sup> This QAPP provides a description of the individuals, organizational responsibilities, and control measures necessary to achieve, verify and demonstrate compliance with both federal and industry quality assurance requirements. This QAPP has been developed using the guidance in EPA QA/G-5 to ensure that appropriate requirements for project data quality have been adequately addressed.<sup>2</sup>

### ***IN SITU* GAMMA SPECTROSCOPY**

*In situ* gamma spectroscopy has been shown to be cost-effective in almost all applications where field sampling and laboratory analyses are the baseline technologies. Results can be obtained immediately following field acquisitions, thereby reducing the time delays incurred by physical sampling and laboratory analysis. *In situ* measurements can be performed on sealed systems (i.e., without breaching a containment barrier) or remotely (i.e., at a distance from an external radiation source), reducing personnel exposures and/or work hazards. Where independent lab analysis is required prior to free release of materials, *in situ* measurements serve as a screening technique, eliminating the unnecessary analysis of samples above derived concentration guidelines (DCGLs). Large areas or volumes can be assayed with a large field of view to reduce errors arising from non-homogeneity, providing a more accurate estimate of average radionuclide concentrations. These advantages make *in situ* spectroscopy an attractive tool for many characterization applications. The Canberra ISOCS system couples previously proven detector hardware with innovative calibration software to produce an integrated instrument capable of quantified analysis in the field comparable to laboratory-grade analysis.

*Germanium Detector:* The radiation detector utilizes a high purity germanium crystal for high resolution and high efficiency gamma radiation detection. For this application, a Canberra Broad Energy Germanium (BEGe) detector was selected because it enhances the efficiency below 1 MeV while exhibiting increased transparency to high energy gammas, such as those from naturally occurring K-40 and Tl-208 (thorium series progeny). The detector shape (50 mm diameter by 30 mm thick) is optimized for analysis of real-world objects in the detector's field of view, but it has less sensitivity to a Marinelli beaker sample geometry than a traditional cylindrical-shaped coaxial detector. The enhanced detector efficiency for low energy gammas (from 30-100 keV) provides a field capability for detection of Am-241 and low energy gammas associated with actinide alpha-emitters that greatly exceeds the capability of traditional detectors.

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*Modular shields and cart:* Useful mechanical components of the ISOCS system include a field deployable mobile cart and a modular system of stainless-steel covered lead shields. Annular side shields of either 19 mm or 44 mm lead thickness effectively reduce the detection of interfering radiation from items in the vicinity of the detector and background radiation, resulting in improved system sensitivity. The field of view can be further restricted, from 180E to 90E or 30E, by installing lead collimators on the cart's mounting rails, so that items adjacent to the object of interest can be significantly reduced or eliminated from the analysis. In addition, a completely shielded sample chamber can be assembled by combining the components of the two thickness annular shield systems to enable timely, low-background analysis of samples in the field.

*Analytical Software:* The ISOCS efficiency calibration software provides the user with the ability to quantify nuclide activity easily and reliably. This software employs a mathematical calibration technique that includes detector-specific characteristics, accounts for collimators and/or shields, and models the physical object to be assayed. It uses a combination of Monte Carlo calculations and discrete ordinate attenuation computations to derive efficiency curves (fraction of gammas emitted from the object that interact in the detector for an energy interval) for each specific *in situ* analysis. Objects are modeled from one of a set of generic sample shapes, such as boxes, cylinders, planes, spheres, pipes, etc. These basic geometry templates have many parameters that can be modified to create an accurate representation of the sample object and detector geometry. Efficiencies can be generated in a few minutes in the field and can be modified easily if needed.

*Technology Application:* 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. Final results have been reported the same day, following data review and validation. Lower activities or more difficult to measure objects, such as enclosed systems, buried sources, and low-level surface contamination, can take much longer to measure and evaluate. However, large surface areas or volumes with heterogeneous material distributions can be assayed with a single *in situ* measurement, thus saving time over other, more manual, methods, such as sampling and remote laboratory analysis.

A typical ISOCS application can be illustrated by reviewing the characterization of core-cooling exhaust fans, prior to their removal, volume reduction, and shipment from the site. Each fan is a massive squirrel-cage type blower, nominally 8 ft x 10 ft x 12 ft, and 14,000 lbs. The fans became internally contaminated, likely as a result of fuel element failure, but the identity, extent, and quantity of radioactive material in the fan internals were unknown. External surveys revealed non-uniform internal deposition with highest readings in the vicinity of the fan volutes, where entrained dust particles would have had a higher probability of settling out due to eddies and dead spaces in air flow currents. Three of the five fans had been upgraded/replaced during the operating life of the reactor. Thus, physical configurations, dimensions, and radionuclide quantities were different from those in the other two fans.

The ISOCS was mobilized to the Fan House containing the five contaminated fans and *in situ* gamma spectra were acquired from Fan #5 (representative of Fans #5 and 4) and Fan #3 (representative of Fans #3, 2, and 1). Figure 3 is a photograph of the ISOCS deployed at the BGRR Fan House. Each fan housing was scanned using 44 mm annular shields and 90<sup>E</sup> field of view collimators to reduce interference as much as possible from adjacent contaminated structures. Because of the equipment layout, there was no position where gamma spectra could be acquired without structural components

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(concrete supports and carbon steel struts) shielding a portion of the field of view. The ISOCS cart was positioned so that the detector was oriented diagonally downwards at the fan housing volute bottom, where surveys indicated an accumulation of radioactivity. Spectra were accumulated for 15 minutes each from two symmetric positions: NE of the housing facing SW and NW of the housing facing SE. Equipment setup, spectrum acquisition and equipment-breakdown required less than two hours, with only minimal health and safety oversight and no breaching of contamination containment barriers. Radioactivity in the fan housing was modeled as a layer of surface dust, uniformly covering the interior of the carbon steel fan volute (horizontal or diagonal rectangular plane). Due to the complex geometry with intervening structural members, several alternative geometry models were defined. The intervening structural members were adjusted in the models until the results from the symmetric scans were similar. The modeling and analysis of both fans required about six hours.

The results of the analysis are provided in Table 1. The range in the value is representative of the uncertainty in the analysis, and is primarily associated with assumptions on unobserved inner structures of the fan. The table demonstrates that even when using a detector with enhanced low-energy response, the detection level can still be high when the source is shielded by a highly attenuating medium such as this example, inside a carbon steel fan housing. With the use of the ISOCS modeling software, a quantified estimate of the activity in the fan was provided in approximately eight hours, without fabricating a physical radioisotope calibration standard, without breaching contaminated barriers, and without handling and transporting contaminated samples.

Table I Results of *In Situ* Measurements at BGRR Fan House

Fan Unit	Measured Activity, FCi			
	Cs-137 low estimate	Cs-137 high estimate	Co-60	Am-241
Fan No 5	75 ± 3	600 ± 20	# 0.1	# 180
Fan No 3	114 ± 10	330 ± 30	4.8 ± 0.5	# 800

Note: The uncertainties in the table represent ±2s counting error; values expressed as “#” represent the minimum detectable activity and indicate that the radionuclide was not detected.



**Figure 3** Using *in situ* ISOCS system to evaluate internal contamination at the BNL BGRR Fan House #5

Another convenient use of the ISOCS technology is the rapid quantification of soil activity in the field. The use of the modular shields to construct a low-background counting chamber on the mobile cart/stand provides a laboratory quality, quantified analysis that is available in almost real time at the point of sampling. The typical detection levels attained by a five-minute count of a soil sample in a one liter high density polyethylene bottle are illustrated in Table 2.

Table II Typical Minimum Detectable Activity for 5-min Assay of 1L Soil Samples

Radionuclide	Co-60	Cs-137	Eu-152	Th-232 <sup>(a)</sup>	Ra-226 <sup>(b)</sup>	U-235	Am-241
MDA (pCi/g)	0.26	0.24	0.32	0.35	0.43	0.20	0.24

<sup>(a)</sup> Th-232 activity inferred from Pb-212 assuming secular equilibrium.

<sup>(b)</sup> Ra-226 activity inferred from Bi-214 assuming secular equilibrium.

*Observations:* Data has successfully been acquired and evaluated for several BGRR facilities and components including the Pile Fan Sump (PFS), underground piping for the PFS, parking lot areas, Above Ground Ducts, and contaminated cooling fans. Through deployment of ISOCS to date, the ability to provide individual isotopic identification and quantitative assays in the field quickly and reliably has highlighted several advantages of the technology:

- C reduction of the cost and time delay associated with sampling and laboratory analysis
- C reduction of the potential hazards of contaminated system entry and sampling/measurement
- C the ability to derive efficiencies for objects without purchasing or fabricating radioactive standards
- C enhanced detection sensitivity by using the annular shields and collimators



One design limitation has been identified that affects the efficient use of the BEGe system in the characterization phase of a D&D project. The 30E collimator shields over 90% of the sensitive area of the detector, thus negating any increased sensitivity of the detector. Having to rely on only the 90E collimator results in some assays being more complex and less specific, since adjacent sources cannot be readily screened from the primary object spectrum.

### **FIELD DEPLOYABLE SR-90 ANALYSIS**

*Technology Description:* 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.

Initially, the strontium-90 detection efficiency of the BetaScint system is established by measuring its response to site-specific calibration standards. These calibration standards are prepared by spiking clean (non-contaminated) site soils with known quantities of strontium-90. The net system response is directly proportional to strontium-90 activity concentration because it is almost entirely due to strontium-90 beta interactions.

Routine daily operations of the BetaScint system include the performance of daily quality control checks and background measurements. Quality control checks consist of analysis of a calibration standard of known activity. The results of the quality control checks are compared against established acceptance criteria to determine whether the instrument is functioning properly. Background checks are performed by counting with no samples in place (i.e., bare detector.) These measurements, which are performed daily at a minimum, are subtracted from gross sample counts to establish net detector response. For calibration and operation, a 2 - 3 kg sample is typically dried, sieved to remove organic matter and rocks over 6 mm in size, and spread evenly over a large area counting tray. The sample tray is then positioned beneath the window of the detector and counted for five minutes. Following the analysis, the system reports the number of coincident events and the counts are converted to Sr-90 activity concentration in the soil, using the detection efficiency correlation established with the spiked site soils.

*Implementation at BNL:* The BetaScint system was deployed at Brookhaven for a two week demonstration during the period of December 6 - 17, 1999. Soil samples from the BGRR site and other environmental restoration areas at BNL were collected and analyzed using the system. Figure 4 is a photo of the BetaScint equipment system set up at BNL.

Four calibration standards were prepared from a NIST traceable Sr-90 solution and uncontaminated site soils. Following preparation, the calibration standards were analyzed by the BetaScint system and a Sr-90 detection efficiency correlation was established. It should be noted that the observed efficiency, 0.67 counts/second per pCi/gram, was virtually identical to the efficiency established by BetaScint at other sites.<sup>3</sup>

During the two-week demonstration, a total of 145 evaluations were performed on 35 samples. The analytical count time for these analyses was 5-minutes. Based on the BetaScint results, Sr-90 activity

concentrations in these samples ranged from non-detectable to approximately 70 pCi/gram. The 5-minute count time yielded a minimum detectable activity concentration of approximately 1.2 pCi/gram at the 95% confidence level, which is considerably less than the BGRR DGCL for Sr-90 of 15 pCi/gram.

The calibration standards were re-analyzed by the BetaScint following system calibration in order to evaluate the precision of the BetaScint analyses,. The results of these analyses (Table 3) are within  $\pm 8\%$  of the calibration standard activity concentrations, indicating that the system exhibits acceptable levels of reproducible detection. To assess instrument accuracy, 7 soil samples and aliquots from the 4 calibration standards were sent off-site for conventional baseline Sr-90 analysis to facilitate comparison; the results of these analyses are still pending.

Table III Results of Calibration Standard Measurements

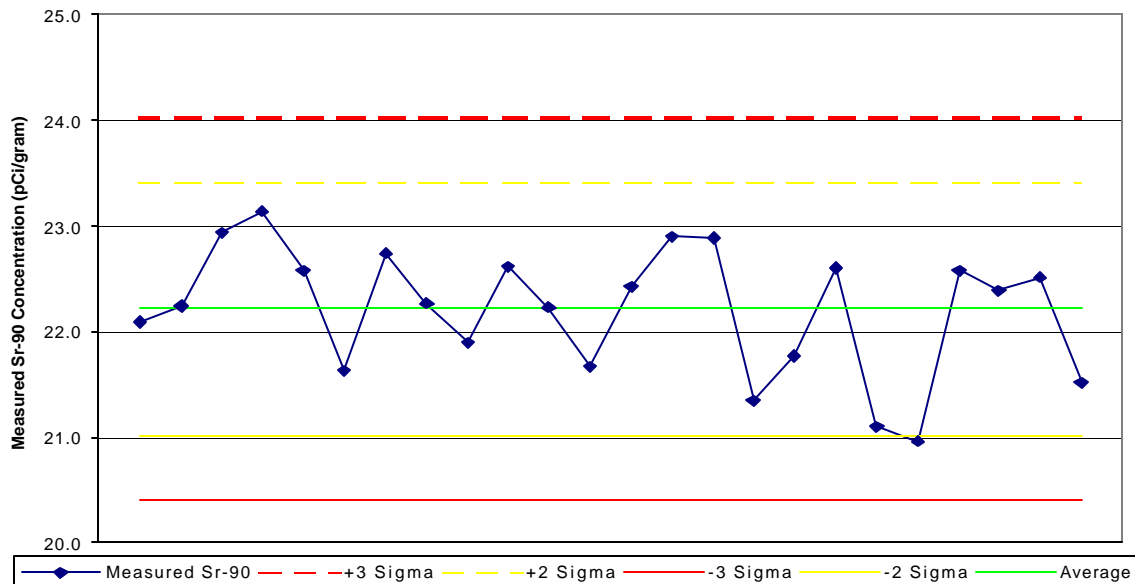
Spiked Sr-90 Concentration (pCi/g)	Measured Sr-90 Concentration <sup>(a)</sup> (pCi/g)	% Difference
7.5	7.9 $\pm$ 1	5
15	16 $\pm$ 2	7
22.5	22.4 $\pm$ 2	1
610	562 $\pm$ 40	8

<sup>(a)</sup> Errors reported at the 95% confidence level

Daily, or more frequent, quality control checks were performed (a total of 25) by analyzing the 22.5 pCi/gram calibration standard that had been prepared. The average result of these analyses was 22.2 pCi/gram (see Figure 5.). All results were within  $\pm 2 s$  of the average value, with the exception of one result that was slightly less than 2 s, indicating that the system response is stable over time and exhibits acceptable levels of precision. Duplicate analyses were also performed on most samples to evaluate the precision of the system. In general, the results of duplicate analyses were within acceptable statistical bounds.



**Figure 4** Loading soil sample for Sr-90 BetaScint analysis at BNL



**Figure 5** Results of quality control measurements for BetaScint in response to 22.5 pCi/g calibration source

## SUMMARY AND CONCLUSIONS

The MARSSIM guidance, with its emphasis on final status survey design, has been applied in the characterization phase through a modification of the final steps of the DQO process. *In situ* data have successfully been acquired and evaluated for several BGRR D&D objectives, and the ability to provide individual isotopic identification and quantitative assays in the field quickly and reliably has highlighted several advantages of the technology. In particular, the Canberra ISOCS system can be used to effectively measure gamma emitting contamination in areas difficult to assay (e.g., pipes and equipment or areas with high radiation levels) or as a field deployable gamma spectroscopy laboratory for volumetric samples. BetaScint provides a field deployable system for near real-time (approx 20 min) evaluation of Sr-90 with good detection limits (1 pCi/g concentrations). Together, the use of MARSSIM with *in situ* characterization techniques is enabling accelerated, accurate, and cost-effective evaluation of equipment, structures, and materials at the BNL BGRR.

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