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PORTABLE NON-DESTRUCTIVE ASSAY METHODS FOR SCREENING AND SEGREGATION OF RADIOACTIVE WASTE

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ABSTRACT

Significant cost-savings and operational efficiency may be realised by performing rapid non-destructive classification of radioactive waste at or near its point of retrieval or generation. There is often a need to quickly categorize and segregate bulk containers (drums, crates etc.) into waste streams defined at various boundary levels (based on its radioactive hazard) in order to meet disposal regulations and consignor waste acceptance criteria.

Recent improvements in gamma spectroscopy technologies have provided the capability to perform rapid in-situ analysis using portable and hand-held devices such as battery-operated medium and high resolution detectors including lanthanum halide and high purity germanium (HPGe). Instruments and technologies that were previously the domain of complex lab systems are now widely available as touch-screen "off-the-shelf" units. Despite such advances, the task of waste stream screening and segregation remains a complex exercise requiring a detailed understanding of programmatic requirements and, in particular, the capability to ensure data quality when operating in the field. This is particularly so when surveying historical waste drums and crates containing heterogeneous debris of unknown composition.

The most widely used portable assay method is based upon far-field High Resolution Gamma Spectroscopy (HRGS) assay using HPGe detectors together with a well engineered deployment cart (such as the PSC TechniCART™ technology). Hand-held Sodium Iodide (NaI) detectors are often also deployed and may also be used to supplement the HPGe measurements in locating hot spots. Portable neutron slab monitors may also be utilised in cases where gamma measurements alone are not suitable.

Several case histories are discussed at various sites where this equipment has been used for in-situ characterization of debris waste, sludge, soil, high activity waste, depleted and enriched uranium, heat source and weapons grade plutonium, fission products, activation products, americium, curium and other more exotic nuclides. The process of acquiring and analyzing data together with integration of historical knowledge to resolve and delineate waste streams (for example between low-level waste and transuranic waste) is described.

INTRODUCTION

There is a growing need for on-site non-destructive assay (NDA) characterization activities in order to streamline the packaging and sentencing of radioactive waste in a cost effective manner. Portable and hand-held devices, particularly high purity germanium (HPGe) systems, are now widely deployed for this purpose benefiting from recent improvement in rugged digital electronics and battery operated detectors. The process of waste stream screening and segregation is nevertheless, a non-trivial exercise requiring expert knowledge and rigorous procedures to ensure data quality requirements are maintained in challenging environments.

DESCRIPTION OF EQUIPMENT

Portable far-field High Resolution Gamma Spectroscopy (HRGS) assay is usually performed using High Purity Germanium (HPGe) detectors, a DigiDART™ multi-channel analyzer, laptop computer with a suitable universal cart such as the Pajarito Scientific Corporation (PSC) TechniCART™. Figure 1 shows a typical arrangement (showing the collimator removed).

The TechniCART™ is engineered for adaptability and flexibility to support up to 250 kg of detection & measurement equipment including heavy collimators. The HPGe (liquid nitrogen or electrically cooled) can be accurately positioned up to 8 feet (243 cm) above the ground. The system allows for rapid reconfiguration including adjusting slide mechanism and a variable tilt adjustment. Flat free wheels permit movement through warehouse, dirt and gravel type terrain. This type of multi-use platform is ideal for portable NDA because it is lightweight, rugged and ergonomically efficient.

Figure 1. PORTABLE NDA: TechniCART™

Hand-held Sodium Iodide (NaI) detectors may also be used to supplement the HPGe measurements in locating hot spots. Extensive evaluation of the assay requirements presented at DOE complex sites has determined that this is the most effective means to attain a quantitative assay suitable for the in-situ characterization.

In addition, portable neutron slab monitors are used for assays where gamma measurements are not suitable. The total neutron count is correlated to the primary neutron emitter after (alpha,n) emissions are corrected for from a knowledge of the chemical and isotopic composition of the waste (the latter may be measured by HRGS).

CALIBRATION

Every gamma detector has a unique intrinsic efficiency profile. The standard [2] definition of efficiency is related to the probability that a gamma-ray emitted from a point source 30 cm from the detector will be counted in a photopeak. This efficiency is a function of gamma-ray energy. Efficiency calibration is represented by a mathematical function that defines the system efficiency response as a function of energy – referred to as the efficiency curve. The efficiency curve is usually determined by taking measurements with a traceable source of known radionuclide composition and quantity. A suitable source emits gamma-rays that span the operating spectrum range of the detector. Commonly used standards [3] include mixed gamma sources with activities from 59.5 keV to 1.836 MeV. Thus Eu152, Eu154, or combinations of mixed gamma sources including Cs137, Co57, and Co60. The efficiency curve is constant for a particular detector and does not require adjustment so long as the detector is not damaged and has no demonstrated deterioration.

Once the detector's efficiency is established, models can be developed to accurately determine the detection efficiency for the containers that are required to be assayed such as rectangular containers (boxes) or cylinders (drums).

MEASUREMENT PROCEDURE

After the portable NDA equipment has been set up, calibrated and commissioned on site, the measurement campaign usually begins with a set of one or more background measurements. These are performed with the portable NDA system on a daily basis or, at a minimum, whenever the system is deployed in a new environment.

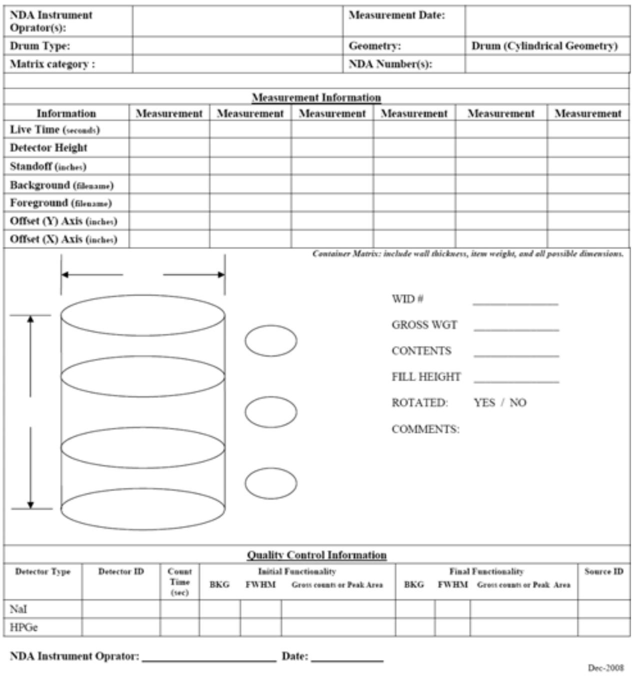
In accordance with ANSI N15.36-1994 [1], the system is tested for proper operation before and after use in its operating environment. This is accomplished by performing a functionality measurement (bracketing each shift's work) of a Quality Control (QC) source of known radionuclide composition and quantity. Using both a high energy and a low energy reference gamma-ray for the QC check evaluates the full operating range of the spectrometer. Three parameters are evaluated and tracked to determine the systems functionality: peak centroid, peak Full Width at Half Maximum (FWHM) and peak net area. The peak centroid check will maintain the energy calibration, the peak FWHM check will maintain the FWHM calibration and the net area test will maintain the efficiency calibration.

The results from the QC functionality check are entered into the QC tracking software by the operator in the field. This process confirms the functionality of the system and after the shift the data is usually uploaded into an electronic database for review by an NDA Professional. Quality Control Charts are generated to provide long term tracking information that allows the analyst to rapidly diagnose systematic trends in system performance (e.g. slow degradation in detector resolution) and take early preventive action as appropriate.

Each container in the assay campaign is measured one-at-a-time with the HPGe detector. The analyst must determine suitable collimation and shielding to use and the number of positions / heights to be measured, as appropriate, for each container. Counting is usually performed until a preset statistical precision is achieved or until a default maximum count time is reached. For example when counting low-enriched uranium it is usual to count until the photopeak at 258 keV contains at least 1000 net counts. However, for high-enriched uranium the preference is to obtain at least 1000 net counts for the 1001keV photopeak. The count time for drums and smaller items is usually in the range of 30 minutes to 1 hour whereas boxes are typically counted for between 1 and 3 hours.

The usual procedure for identifying ‘hot-spots’ (for extended assay) is to use a 3x3 NaI Detector. Firstly the operator will determine the gamma background of the measurement area by acquiring data in different directions around or near the item to be scanned. The item is then scanned by pointing the detector toward the item and moving the detector across all surface areas at a speed of about 10 cm per second with the detector as close to the surface as possible. For large boxes, the operator often uses the “18 point” scan method - a scan technique developed using 18 points on a box type container to detect gamma-ray anomalies or hotspots within a containerized matrix to assist with modeling.

The operator will always carefully document the scan rate measurement results and the distance / orientation of the detectors or on the NDA Field Worksheet. An example worksheet in is shown in Figure 2.



NDA Instrument Oprator(s):		Measurement Date:	
Drum Type:		Geometry:	Drum (Cylindrical Geometry)
Matrix category :		NDA Number(s):	

Measurement Information

Information	Measurement	Measurement	Measurement	Measurement	Measurement	Measurement
Live Time (seconds)						
Detector Height						
Standoff (inches)						
Background (filename)						
Foreground (filename)						
Offset (Y) Axis (inches)						
Offset (X) Axis (inches)						

Container Matrix: include wall thickness, item weight, and all possible dimensions.

WID # ________
GROSS WGT ________
CONTENTS ________
FILL HEIGHT ________
ROTATED: YES / NO
COMMENTS:

Quality Control Information

Detector Type	Detector ID	Count Time (sec)	Initial Functionality BKG	FWHM	Gross counts or Peak Area	Final Functionality BKG	FWHM	Gross counts or Peak Area	Source ID
NaI									
HPGe									

NDA Instrument Oprator: ________ Date: ______

Dec-2008

FIGURE 2. EXAMPLE FIELD WORKSHEET

The acquisition software used is ORTEC’s GAMMAVISION supplemented by the ISOTOPIC modeling software. Isotopic analysis codes such as PC-FRAM may also be used.

GAMMAVISION is used for acquisition and basic peak fitting of spectra. This process calculates net peak area counts for gamma-rays of interest. The results of the peak fitting can then be entered into a gamma modeling software application to identify peaks, model the measured item, correct for matrix, geometry and other factors and calculate relevant isotopic activities and activity densities.

Nuclide quantification and sample modeling corrections are performed using ISOTOPIC. This provides a practical solution to a wide range of gamma-ray measurement problems encountered in site characterization including the measurement of large boxes and drums. ISOTOPIC has been developed from work done originally at several US DOE sites in the analysis of thousands of fissile waste containers [5]. The analysis engine includes methods developed at the US Energy Measurements Laboratory [6], [7] to measure wide-area contamination of soils and surfaces.

A semi-empirical approach to efficiency calibration is used. ISOTOPIC provides a number of standard geometry "templates" from which a specific measurement template may be developed. These include cylinders, lined cylinders (pipes), boxes, point source (far field), and infinite plane. For the counting of packages, pipes and surfaces, the detector is characterized by a single point-source measurement, even when a collimator is to be used. This primary calibration, which can be traced to a certified standard, for any detector, is extrapolated or modelled to match the physical situation of the sample; container geometry, material, and matrix composition.

Minimum Detectable Activity (MDA) is calculated for each isotope. Unknown peaks are identified in the report file so that any nuclide not already found in the analysis library will be available to the analyst. The library can then be changed to include these nuclides. Activities, grams of a particular nuclide, or MDAs from multiple measurements may be reported as weighted averages. The weighting is user definable by the analyst.

Differential Peak Analysis

This method relies on the observation of several photopeaks with different energies that originate from the same radionuclide. It provides a means of identifying and eliminating potential biases involved from the assumptions used to model the item. Inaccuracies such as matrix composition and density manifest themselves by an energy dependency in the activity calculated at the various photopeak energies. The form of this energy dependency can be used to determine if the bias is due to the matrix composition or the matrix density. If the reported activity constantly increases or decreases with energy then the inaccuracy is most likely attributed to the matrix density. If however, the variation in the reported activity has a more complex form then this is an indication of either self-absorption in a lump of the radionuclide being analyzed or deviations between the assumed and actual matrix composition. Careful data review is essential to remove this and other sources of bias in the model used to analyze the measurements.

Peer Review Process

A critical component of an accurate portable NDA program is the peer review process. The purpose of peer review is to identify and, where possible, correct reported results that may affect data quality due to conditions outside the bounds of the calibration assumptions. Possible conditions, if any, are identified and appropriate remedial action steps may be initiated (e.g. re-analyse or re-measure). Figure 3 illustrates the Portable NDA Data Flow and Review Process. The procedure includes initial Data Quality review by the NDA Technical Specialist or Operator and an Independent Review once the data analysis is completed.

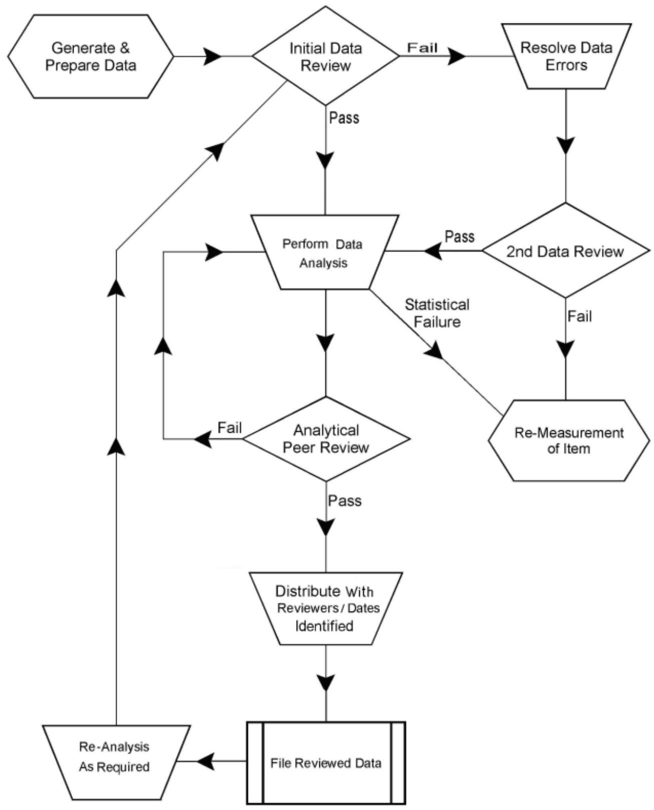


FIGURE 3. DATA FLOW AND REVIEW PROCESS

An initial review of the measurement data is carried out by the operator to ensure that the data quality is satisfactory. This includes spectral quality checks of the data associated with the assays and the data associated with measurements of the daily check sources. These checks include spectral resolution, detector efficiency and energy calibration stability. For example, acquisition dead time is reviewed to ensure that it is consistent with the observed count rates and peak shapes.

An independent review is performed on each measurement. In this process a statistical assessment is performed on all data analysis results produced from field measurements. All technical documentation created for supporting NDA activities are reviewed. This may include a walk-through of technical documents, quality control, and spectral analysis performed on a measured item. If during the analysis process, the electronic media is determined to be bad or erroneous results are generated from known data, it shall be considered a statistical failure or if the quality control is outside the guidelines established for a given detector it may be considered out-of-control. In the event a statistical failure occurs, an independent analyst may analyze the data for failure confirmation. If confirmation is obtained, the data is omitted and re-measurement of the item should be considered. During the measurement process, the portable NDA team must maintain all measurement equipment both quantitatively and qualitatively within the bounds of control. Once the Independent Review process is completed, the results are then processed for reporting.

Training requirements

Training is another critical component of a successful measurement protocol. All staff involved in portable NDA (i.e., technical analysis, data review, operating assay equipment etc.) must be, at a minimum, trained and qualified in accordance with ASTM C-1490-04 [7] which stipulates the minimum education levels, training and experience necessary for the team members.

MEASUREMENT UNCERTAINTY

The total measurement uncertainty (TMU) associated with an individual container may be determined based on the observed gamma-ray spectrum and the geometry model that is applied during far field analysis. For example, in the case of a uranium measurement, the analysis will determine both the U235 and U238 content. Where the U235 enrichment is known (from process knowledge or isotopic evaluation) the U238 content may be used to determine an independent measurement of the U235 content. By combining these two independent measurements of U235 using a weighted average approach the uncertainty in the reported U235 may be minimized.

The activity concentration, AC is calculated from the measured activity, A and the net weight, NW. This relationship is presented in Equation 1.

$$AC = \frac{A}{NW} \qquad (1)$$

The uncertainty in the activity concentration, σ_{AC} , is calculated from the measured activity and its associated uncertainty, σ_A , together with the net weight NW and its associated uncertainty, σ_{NW}. This relationship is presented in Equation 2.

$$\sigma_{AC} = AC\sqrt{\left(\frac{\sigma_A}{A}\right)^2 + \left(\frac{\sigma_{NW}}{NW}\right)^2} \qquad (2)$$

It is likely that the uncertainty in the net weight is negligible in comparison with the uncertainty in the measured activity. For this reason Equation 2 can be simplified to the expression given in Equation 3.

$$\sigma_{AC} = \frac{\sigma_A}{NW} \tag{3}$$

Calculations Associated with a Group of Containers

If it is found that the calculated uncertainty associated with the activity concentration of an individual container is greater than the threshold value, then a group of containers may be selected for which the activity concentration will be recalculated. This approach will serve to reduce the reported uncertainty associated with the activity concentration that is calculated for the group. Calculations of the total activity concentration, TAC and its associated uncertainty, σ_{TAC} for a group of containers, where n is the number of containers in the group, are presented in Equation 4 and Equation 5.

$$TAC = \frac{\sum_{i=1}^{n} A_i}{\sum_{i=1}^{n} NW_i} \tag{4}$$

$$\sigma_{TAC} = TAC\sqrt{\left(\frac{\sqrt{\sum_{i=1}^{n} \sigma_{A_i}^{2}}}{\sum_{i=1}^{n} A_i}\right)^2 + \left(\frac{\sqrt{\sum_{i=1}^{n} \sigma_{NW_i}^{2}}}{\sum_{i=1}^{n} NW_i}\right)^2} \tag{5}$$

The uncertainty in the net weight will most likely be negligible in comparison with the uncertainty in the measured activity. For this reason Equation 5 can be simplified to the expression given in Equation 6.

$$\sigma_{TAC} = \frac{\sqrt{\sum_{i=1}^{n} \sigma_{A_i}^{2}}}{\sum_{i=1}^{n} NW_i} \tag{6}$$

Example Calculation

To illustrate the application of this technique, consider a hypothetical uranium site where a limit exists on the uncertainty per waste consignment of 285 pCi/g. The simulated results for a group of five containers are presented in Table 1 together with the results for the group. For each of the individual containers activity concentrations in the range 1500pCi/g to 1900pCi/g were selected using a random number generator. Likewise the uncertainties in the activity concentrations were randomly sampled in the range of 25% to 40%. The net weight for each container was populated over the range 50kg to 100kg. The activity concentration and its associated uncertainty for the group of five containers were calculated using Equation 4 and Equation 6.

In each case, the uncertainty in the activity concentration exceeds the site limit of 285 pCi/g for the individual containers. However, when the activity concentration and its uncertainty are calculated for the group of containers the calculated uncertainty of 237pCi/g meets the requirement.

TABLE 1. EXAMPLE TMU CALCULATION

Drum	Net Weight (kg)	U235 Mass (g)		Activity Conc. (pCi/g)	
		Value	TMU	Value	TMU
Drum 1	74.7	57.5	16.7	1694	491
Drum 2	81.8	68.8	18.6	1851	500
Drum 3	78.7	54.5	18.5	1520	517
Drum 4	78.7	60.7	18.8	1699	527
Drum 5	58.4	49.1	17.2	1848	647
Total	372.3	290.5	40.2	1717	237

DATA REPORTING

After the completion of the analysis, measurement uncertainty calculation and peer review process a final data report is generated for the operating site. In this report the nuclide activities are listed together with associated uncertainty. The minimum detectable activity is often reported for each nuclide. Where required, the activities or mass values are summed e.g. to determine total alpha activity, total uranium mass etc. Cross-referencing information is supplied to reference documents such as the background and QC measurement reports, field worksheets and operating procedures used. The analysis software and version numbers are also listed on each report.

BENEFITS SCREENING AND SEGREGATION

Portable NDA equipment can quickly categorize and segregate bulk containers (drums, crates etc.) into waste streams defined at various boundary levels (based on its radioactive hazard) in order to meet disposal regulations and consignor waste acceptance criteria. Typically for LLW / TRU sorting this boundary is at 100 alpha nanoCuries per gram of waste (3700 Bq/g). Early screening and segregation of waste with portable NDA equipment allows the site operator to efficiently sentence and repackage the waste. Thus the throughput of containers to any downstream high sensitivity assay units (e.g. systems using passive or active neutron) can be more effectively controlled and managed. Repackaging of the waste at this stage can also ensure dose uptake minimization and reduce transportation and storage costs.

PORTABLE NDA EXPERIENCE

The following case histories provide valuable experience and lessons learned at various sites in North America where many different waste streams and isotopic mixtures have been encountered.

Portsmouth Gaseous Diffusion Plant (PORTS)

NDA services have been performed at PORTS since 2005. The portable assay systems have measured thousands of items including drums, boxes, cans, bottles and HEPA filters. Uranium is the main element of interest for these measurements but all radionuclides detected are reported. Many of the waste items assayed at PORTS have been disposed of at Nevada Test Site (NTS) using this characterization data.

Los Alamos National Laboratory (LANL)

Portable far-field HRGS HPGe assay systems have been deployed at most technical areas at LANL to measure a wide variety of items including drums, boxes, cans, bottles, HEPA filters and glove boxes [8]. A wide and sometimes exotic range of radionuclides are encountered routinely from the measured items at LANL, including but not limited to Pu239, Pu238, Pu240, Pu241, Am241, Am243, Cf249, Cm243, Eu152, Eu154, U235, U238. Since 2005, the portable NDA program set up at LANL has assisted in the measurement of thousands of items and continues to be part of the gamma spectroscopy group. The assay results are primarily used for transuranic (TRU) low-level (LL) sorting with many of the waste items having been disposed of at NTS.

Hanford

Portable far-field HRGS HPGe assay systems have been utilised at Hanford's 100 and 300 areas to characterize a wide variety of items including glove boxes, HEPA filters, pipe arrays, ducts and tanks. Innovative characterization techniques have been used to overcome significant challenges including remote characterization of the highly contaminated building 324 B-cell, and in-situ characterization of the building 327 dry fuel storage carrousel. A wide range of radionuclides are often encountered from the measured items at Hanford , including Pu239, Pu238, Pu240, Pu241, Am241, Am243, Cf249, Cs137 Eu152, Eu154, U235, U238. These assay results are primarily used to guide and optimize D&D activities on the Hanford site.

Lovelace Respiratory Research Laboratory (LRRI) – at Sandia National Laboratory

In 2008 a portable far-field HRGS HPGe assay system was deployed by PSC at LRRI to assay a small number of boxes and drums for TRU/LLW sorting. Measured items were identified as suspect TRU or LLW which allowed LRRI to determine the best waste disposal option.

Advanced Mixed Waste Treatment Facility (AMWTP) – at Idaho National Laboratory

The Advanced Mixed Waste Treatment Facility in Idaho processes transuranic (TRU) waste for shipment to the Waste Isolation Pilot Plant (WIPP). At AMWTP, various fixed and portable measurement instruments have been used to solve challenging measurement problems including assay of sludge, soil, high activity waste, depleted and enriched uranium, heat source and weapons grade plutonium, fission products, activation products, transuranics and other more exotic nuclides. Key to this success is the building of a team of NDA professional with wide range of innovative characterization techniques that have been applied within both the TRU and LLW programs. These include; characterization of LLW for disposal at NTS and the Energy*Solutions* facility in Clive, Utah, characterization of lead lined drums and identification of remote handled waste, and screening level characterization of containers using both historical data and field NDA measurements.

Characterization techniques at AMWTP have addressed a wide range of challenges posed by waste from sites throughout the DOE complex [9]. These sites include; Rocky Flats, Mound, Hanford, Bettis Atomic Power Laboratory, Battelle Columbus Laboratory, Idaho National Laboratory, and Argonne East. To date Pajarito Scientific Corporation has been directly involved in the characterization for disposal of over one hundred thousand drums of TRU and LLW.

CONCLUSIONS

A variety of complex and challenging waste streams have been successfully analyzed using high quality portable NDA measurement programs. Equipment and processes have been developed to allow site operators to rapidly screen and segregate waste for more efficient packaging, storage, transportation and sentencing.

Key to the success of the portable NDA program is the rigor attached to data acquisition and analysis. Routinely performed background and QC measurements are essential as is a thorough and detailed independent review of each and every measurement. The lessons learned from decades of operations is that 'bad data quality' is usually symptomatic of the NDA supplier not implementing or understanding the need for data review and simply treating the equipment and software as a black box. Peer review is an essential part of the portable NDA program.

Field measurements of this type must often sacrifice an element of accuracy in order to achieve pragmatic goals for container throughput and the mobility of the equipment. One way to reduce the total measurement uncertainty is by using a container grouping approach. This method will reduce the TMU associated with small groups of containers that will be defined as packages. Target measurement uncertainties at less than +/- 15% are achievable with this approach.

The on-site surveying & measurement technology described in this paper can be utilized for short term or project specific deployment at client sites. These systems provide rapid and cost effective characterization data without the capital expenditure for a single use item. Full technical and operational support is provided with personnel who have worked for many years within the operating infrastructure of nuclear facilities. To meet the needs of the field operations a suite of flexible ruggedized detector deployment platforms have been developed that provide stable and dependable operation over a wide range of site conditions and terrains.

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