Radiometric Characterization Process for Locating Radioactivity Hold-up and Measuring Non-uniformly Distributed Sources-15342

Richard Creed*, Tom Donohoue*, Erik Lindberg*, Marc R. Looman**, E. Ray Martin*, John A. Mason**, Daniel Pancake*, Cynthia Rock*** and Douglas J. Walraven*
*ANTECH Corporation 9050 Marshall Court, Westminster, CO, 80031, USA
**ANTECH, A. N. Technology Ltd., Unit 6, Thames Park, Wallingford, Oxfordshire, OX10 9TA, UK
***Argonne National Laboratory, 9700 South Cass Avenue, Lemont, IL 60439, USA

ABSTRACT

A novel approach to facility radiometric characterization for non-uniformly distributed radiation sources has been developed by ANTECH and is currently being employed at the Argonne National Laboratory. It is being applied to the measurement of a wide range of objects and the interiors and contents of laboratories and buildings. The integrated non-destructive assay measurement process involves the use of a scanning spectroscopic gamma ray camera as well as high purity Germanium detectors with different collimators and hence differing detector fields of view to measure and quantify non-uniformly distributed sources of radioactivity in laboratory equipment, glove boxes, storage tanks, pipework and general laboratory objects and process components. The purpose of the radiometric characterization is to quantify radiation sources and radioactive contamination levels in facilities containing nuclear material and to ensure compliance with facility characterization requirements and for decontamination and decommissioning planning and execution. The integrated measurement process also involves automated data capture and processing software and a database with which both measurement instrument operators and subject matter expert analysts can interact. An appropriate measuring instrument (gamma camera, scintillation detector, high purity Germanium detector system, neutron detector or alpha-beta counter) records the measurement data and the measurement site is recorded in a photograph. All of the data relevant to the measurement (or set of measurements for a particular object) are recorded in the database including measured gamma ray spectra, equipment description and relevant calibration, measurement site photograph and measurement descriptive data. Once data analysis protocols are established for specific geometries and measurement situations they can be applied to the measured data in a partially automated manner, reducing analysis and transcription errors. In addition to measurements and data analysis the process also utilises radiometric modelling computer codes including MicroShield and Monte Carlo Neutrons Photons in order to perform model calculations where appropriate. In a typical measurement, the ANTECH RadSearch spectroscopic gamma camera is utilized to find areas of concentrated activity so that these areas can be measured and calculated separately from areas with diffuse contamination. The use of the gamma camera in scanning mode helps to ensure that the assay does not overestimate the detected radiation source, which is a frequent consequence of using standard far field geometry and attenuation analysis codes that assume uniformity of both density and radioactive source distribution. Once the hot spots are identified, high purity Germanium detector measurements are made using various collimators with wide or narrow fields of view as required. These measurements are then processed through a semi-automatic calculation process assuming an appropriate source configuration as identified by RadSearch. These source configurations include point sources, line sources or surface sources. The Generalized Geometry Hold-up algorithms are employed to determine an estimate of the activity of the various sources that are identified in a measurement. The ANTECH integrated measurement process provides an automated calculation procedure where the analyst can select the source type, geometry and appropriate input data, and then the software calculates the activity by radionuclide for all sources identified in the measurement. The measurement and analysis process is controlled under the

ANTECH ISO9001: 2008 Quality Assurance System and managed by a site specific Quality Assurance Plan. Both Argonne National Laboratory and Department of Energy technical reviewers have vetted the Integrated Measurement and Analysis Process. The application of the measurement and analysis process is described and measurement examples are provided.

INTRODUCTION

This paper describes a continuing project, during which ANTECH has developed a comprehensive Integrated Measurement Process. Much of the work has been undertaken at the Argonne National Laboratory (ANL). The process is being refined and updated as the current project continues. The method combines multiple traditional non-destructive assay (NDA) techniques with state-of-the-art information management systems and mathematical modelling such as the Generalized Geometry Hold-up algorithms (GGA) [1] and Monte Carlo Neutrons Photons (MCNP) [2] simulation and modelling using codes such as MicroShield to enhance the benefits of expert analysis techniques for field measurements. International standards are employed in the determination of critical data parameters [3,4]. Using information technology applied to automate the gathering and analysing of measurement data as part of the measurement process, ANTECH Expert Analyst (EA) and subject matter expert (SME) staff are able to be involved in each NDA measurement, even at remote locations. The Integrated Measurement Process (IMP) has helped to ensure that NDA measurements are robust and reliable. As a consequence, the results can be employed by both facility and regulatory authority management in support of critical operational decisions.

INTEGRATED MEASUREMENT PROCESS

There is a wide range of techniques employed in field radioactivity assessments from simple health-physics instruments such as ion chamber survey meters to gamma spectroscopy using high-resolution spectroscopic detectors. Often, adequate measurements can be made with simple instruments. However, when more sophisticated instruments such as an High Purity Germanium (HPGe) detector are brought into play, the quality of the measurement becomes highly dependent on the skill of the operator and the person assessing and analysing the data. As will be described later in this paper, it is not uncommon to misuse a sophisticated instrument such as an object counting system and get an entirely incorrect result. This is where the ANTECH Integrated Measurement Process adds an additional layer of quality assurance.

Processing of field data is done in two distinct phases: data collection, then analysis and reporting. ANTECH has developed software to apply a high level of automation to the data collection phase. Trained field operators using the ANTECH Multi Geometry Gamma Analysis (MGGA) software are able to collect data in a structured manner, independent of the measurement technique. The data are vetted automatically during the actual measurement data collection process, minimizing data entry errors. Operator prompts are based on the expert analyst's needs and requirements, ensuring there are no data omission errors. Measurement Control is automated, ensuring that instrument failures are detected and corrected at an early stage in the measurement process. Finally, all of the data associated with each measurement are combined into a single file so data organization is simple and easy to transmit to the analyst. The MGGA code has been developed and is maintained under ISO-9001 and NQA-1 quality assurance standards.

The EA or SME performs the analysis and generates a report using a semi-automated system, which is also part of the MGGA software suite. The MGGA software allows the expert analyst great flexibility in the data analysis process. In complete contrast to the concept of a field-automated object counting system, where large measurement errors can arise and be undetected,

MGGA software relies on the EA to select and modify the data analysis parameters. This is to ensure that the measurement data is correctly analysed, considering the assumptions and constraints that apply to the measurement situation and that these constraints are considered in the analysis. In this way the data is analysed correctly and as a consequence robust and reliable results are generated. The MGGA software then stores the result of the EA analysis in a database. Reports are readily generated from the database and there is ease and flexibility in report formatting. In essence, the Integrated Measurement Process produces a report with an order of magnitude greater level of confidence and quality than the reports produced by fully automated object counting systems. Using the automation incorporated into the MGGA software, the EA can routinely generate reports within a day of data collection.

MEASUREMENT METHODS

The Integrated Measurement Process is adaptable to virtually any field radiation measurement. Currently, it is being used to process data collected with sodium iodide systems, high purity germanium systems, neutron slab counters, and the ANTECH scanning spectroscopic gamma ray camera, RadSearch [5]. The concept is being extended to other measurement techniques such as field HP measurements using hand held dual-phosphor scintillation probes for alpha/beta surface surveys, and sodium-iodide holdup measurements.

These systems are used to quantify activity in or on objects such as glove boxes [6], hoods, drums, waste boxes, pipes, etc. The type of field information collected in support of the measurement, and the manner in which the data are processed depend on what is being measured. This includes information about the measurement location and conditions, the area ambient radiation, the geometry of the item being measured and its spatial relationship to the detector, and any attenuators present. The Integrated Measurement Process incorporating MGGA is designed to support all of these disparate and varied measurement requirements.

Within the Integrated Measurement Process it is assumed that all measurements are based on a careful calibration performed under laboratory conditions. For the HPGe system, this involves measuring known standards at a fixed distance and geometry. The calibration data is captured in the measurement database under MGGA. Adjustments for geometry and attenuation are then made for field measurements to relate the field data to the known response obtained during the instrument calibration.

DETECTOR SYSTEM EXAMPLES

A wide variety of detectors have been employed as part of the Integrated Measurement Process for measurements of radioactive holdup and radiation surveys at the Argonne National Laboratory. These can range from hand held alpha-beta counters to sophisticated spectroscopic scanning systems. In the following sections a typical selection of four different detector systems employed at ANL are considered and examples of their use presented.

Sodium Iodide Detector

In some instances, quantitative measurements are not necessary, or perhaps the ability to discriminate radioisotopes within a contaminant mixture is unnecessary. For this situation we have chosen to use an NaI detector, although a plastic scintillator would also be suitable and provide higher sensitivity. Sodium Iodide based instruments (such as the HMS4) do not need external cooling and are the instruments of choice for holdup measurements. They are often employed

where the measurement setup conditions remain constant for a series of measurements where a difference in response is related to the held-up activity.

As an example ANTECH deployed NaI detector at ANL to determine whether radioactivity was present in some very difficult to reach, 10-meter deep sump wells within hot-cells. Figures 1a and 1 b show the measurement geometry and the spectrum showing that no significant activity.
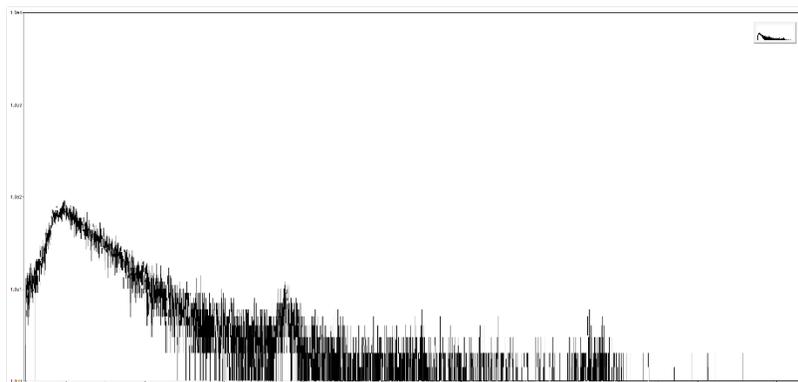

Fig. 1a – Sodium Iodide Detector Suspended in a Sump

Fig. 1b – Typical Sodium Iodide Gamma ray Spectrum from Sump Measurement

High Purity Germanium

The HPGe detector systems are at the core of the Integrated Measurement Process. Different HPGe systems are employed with different energy resolution and collimation for different measurement requirements. Figure 2a shows a safeguards quality HPGe detector with liquid nitrogen cooling and a wide field of view (FOV) collimator measuring a glove box.

Fig. 2a – Liquid nitrogen cooled HPGe coaxial safeguards detector with wide FOV collimator

A versatile HPGe system used at ANL is the ORTEC IDM-20-V, which has a Stirling cooler, an internal digital multichannel analyser and wide-angle collimation. It is mounted on the ANTECH ANTCart®, a highly mobile and versatile wheeled platform or another positioning system. The detector can be positioned in nearly any orientation up to a height of 2.7 metres using ANTCart.

Fig. 2b – IDM-200-V Stirling cooled HPGe detector mounted on the ANTCart®

Neutron Slab Counter

The ANTECH neutron detector module or slab counter consists of a rectangular module containing two He-3 tube detectors. The He-3 detectors are typically Reuter-Stokes RS P4-1620-209 tubes, 2" diameter by 20" (5 cm x 51 cm), mounted in a block of polyethylene 24" x 4.5" x 7.75" (61cm x 11.5 cm x 19.7 cm). The tubes are offset in the polyethylene such that one face is 2" (5 cm) from the tube center, and the other face is 2.5" (6.25 cm) from the tube center. Thus, the module provides some directionality in response to neutrons. The entire module is wrapped with a 0.5 mm cadmium sheet and encased in a stainless steel enclosure.

Neutron slab counters with total neutron counting electronics can be used to locate and determine relative intensity of a variety of neutron sources. Counters were used at ANL to locate and assist in the remediation of a faint source of neutron personnel exposure from a spill of Cf-252 in a Glove box.

Gamma Camera

The RadSearch Gamma Camera [5] is a device for measuring and quantifying radiation in the environment. It is particularly applicable to decommissioning and nuclear clean-up activities. RadSearch is capable of searching for, locating and quantifying radioactivity distributed on surfaces, within equipment and in pipe work and determining radiation dose in the environment. It consists of a detection head with a combined lanthanum bromide ($LaBr_3$) scintillation detector and photomultiplier, video camera and distance measuring laser range-finder system.

Fig. 3 – RadSearch spectroscopic scintillation gamma camera

Field of View Corrections

Most of the detectors have a fixed field of view (FOV). The Generalized Geometry Holdup (GGH) areal-source geometry correction model includes a correction for the effect of collimation on the FOV. A FOV calibration is performed, and mathematical corrections are applied to raw data based on the GGH method [1]. A FOV response function is displayed below in Figure 4.
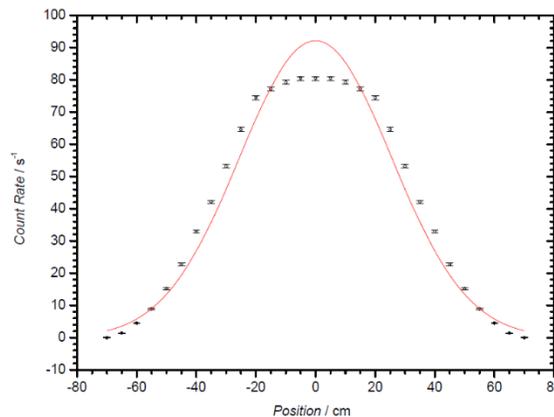

Fig. 4 – Detector FOV measured data and Gaussian fitted function

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MULTIPLE GEOMETRY GAMMA ANALYSIS

The MGGA code is written under the ANTECH ISO9001: 2008 Quality Assurance (QA) System and managed by a site specific QA Plan. It is a key component in the Integrated Measurement Process in that the software directs the operator in data collection and controls both the measurement and analysis process. The data acquisition screen is shown in Figure 5 below.
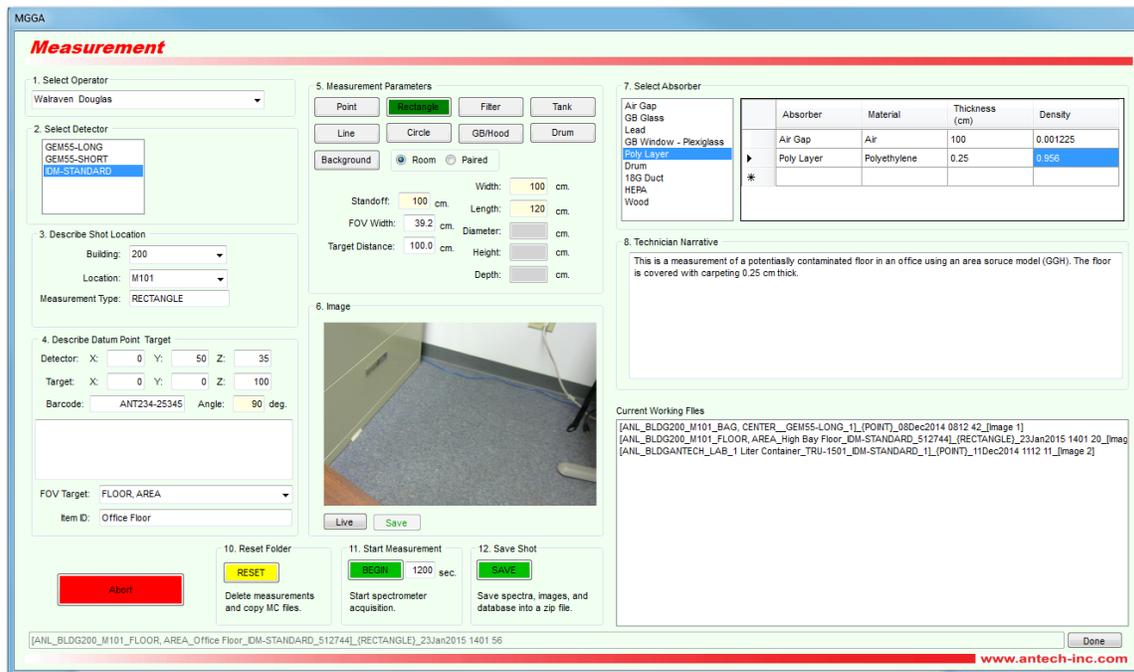

Fig. 5 – Data Acquisition Screenshot

When using MGGA for data acquisition, the operator is presented with drop-down lists of active detectors, locations, measurement control sources, attenuators, etc. A narrative is collected where the operator describes the measurement details. The measurement geometry is selected, and the operator is prompted to supply geometry details depending on the type of measurement. The MGGA software also controls the data acquisition hardware, for example an ORTEC HPGe digital spectrometer. The spectrum is obtained and all measurement details, including an image from the web-camera are combined into a single, easy to transmit dataset. ANTECH has experienced a significant improvement in a data quality by implementing this level of automation.

Typical Gamma ray Spectroscopy Analysis

All measurements performed with an HPGe detector system result in an ORTEC spectrum (.SPC) file. These files are analyzed as histograms (spectra) that relate the detected gamma ray counts to their energies over the calibrated energy range of approximately 50 to 1500 keV. The spectra are processed through the GammaVision® WAN32 peak fitting algorithm to report counts in a specific region of interest (ROI) as a discrete peak. The region of interest represents a full energy peak that is related to a specific isotope based upon the peak energy profile search of the GammaVision® isotope library. The measured intensity and detector efficiency are used by GammaVision® to calculate and report the measured activity of the identified radionuclide. The

activity reported by GammaVision® is the calculated activity for a point source at a measurement distance of 100 cm.

The analysis begins with an automated search to calculate the total activity for each peak that is identified by the WAN32 peak search engine. This is followed by a thorough review of the spectrum by an SME, during which peaks not identified by the analysis engine are manually identified, and "false" peaks are rejected. The ORTEC WAN32 analysis engine does a preliminary library-based peak search of the spectrum. The analysis assumes that all gamma rays listed in the library exist in the spectrum so it tries to fit a peak at each energy listed in the library. A Mariscotti peak search is then implemented on the remainder of the spectrum. The analysis selects from standard peak fitting and background correction algorithms consistent with the GammaVision® User's Manual. Each identified peak is associated with an isotope and the measured counts related through the detector's efficiency calibration and the specified isotope's branching ratio enables the analysis software to calculate and report the individual isotope's activity. A summary report of the nuclides identified in the sample is printed to a report that is then reviewed by the SME. A typical HPGe spectrum is shown in Figure 6.

Each reported nuclide may have one or more associated test codes which show whether or not the peak passed various automated tests. The analyst reviews these codes. In general, the presence of a code is highly suggestive that no real peak is present; the peak is not Gaussian, could not be de-convoluted, is inconsistent with other photo peaks from the same nuclide, etc.
An analysis library is built iteratively by visual inspection of all spectra, identification of potential nuclides present in the spectra, and adjusting until all nuclides are accounted for.
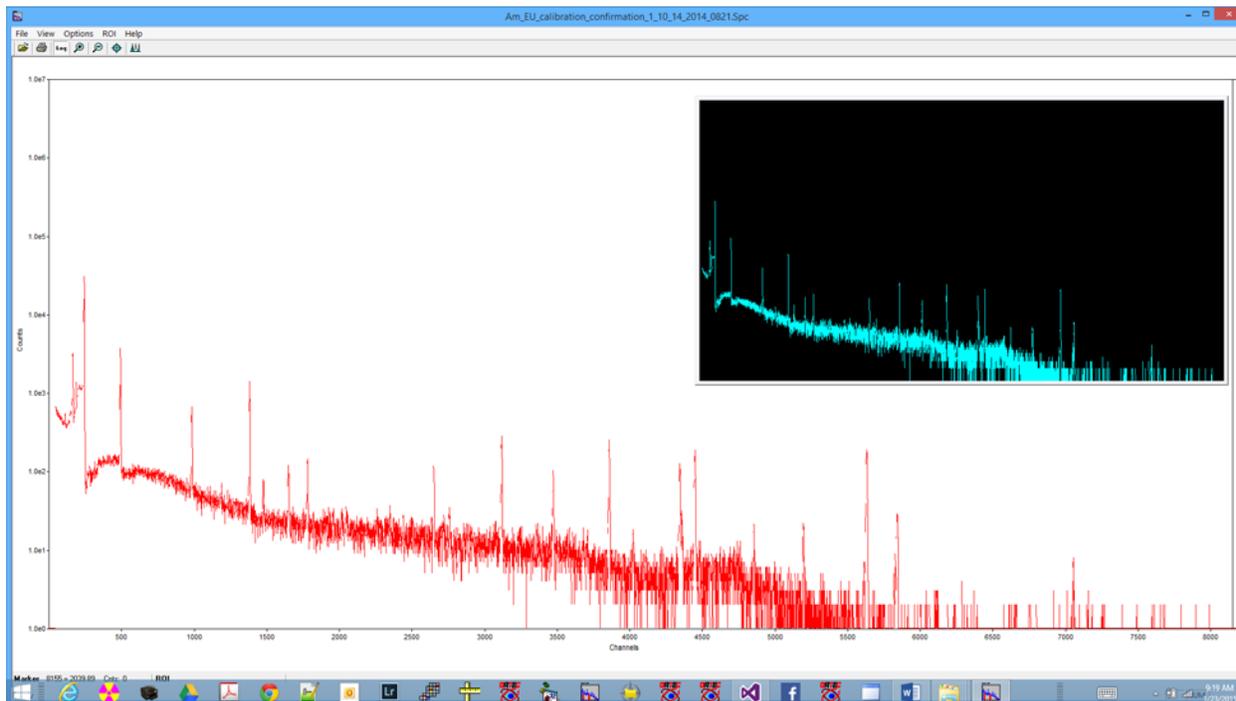

Fig. 6 – Typical HPGe Spectrum

Nuclide data is derived from the comprehensive ORTEC Nuclide Navigator® master library. With each radionuclide added to the library, there may be many (hundreds) of gamma-ray energies. These are further culled to remove gamma-ray lines that are outside of the 50 keV to 1.5 MeV

calibration range specified in the Statement of Work for the project. Note that some of the nuclides are hard to detect; for example, U-238 has only two photons, both below the calibration range. It is hard to detect due to its low-yield, low energy emissions, but it decays to Th-234, which decays to Pa-234m which has a strong 1001 keV peak. The presence of U-238 can be inferred from Pa-234m. An SME, who is an experienced gamma spectroscopist, reviews the GammaVision® analysis report. A typical spectroscopic analysis report is shown in Figure 7.
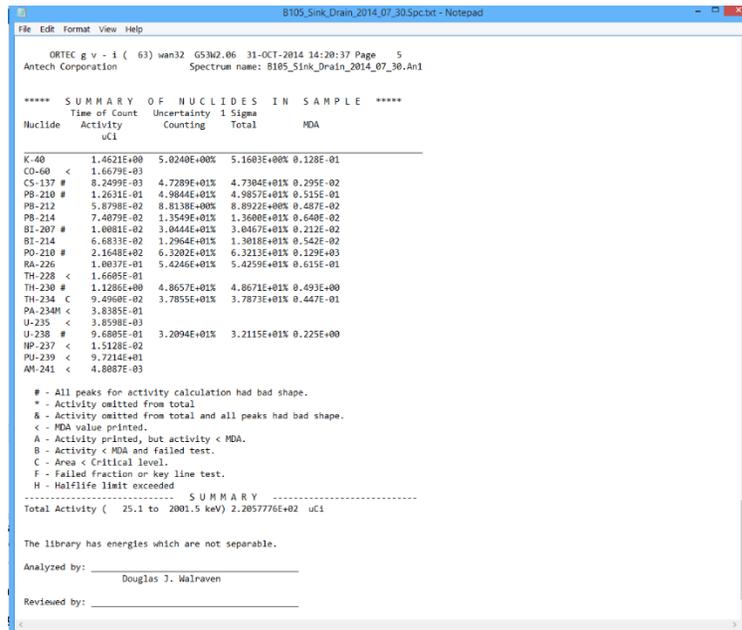

Fig. 7 – Typical Spectroscopic Analysis Report

During his review, the analyst transfers the results of the automated analysis to a database and compares these results with the graphical display of the spectrum. Identified peaks are confirmed. Unidentified peaks are identified where possible. More importantly, "false" peaks are identified. The sensitivity of the WAN32 engine is such that, in addition to identifying real photo -peaks, it tends to identify electronic "noise" artefacts as peaks. The SME analyst identifies these "false" peaks in the final report by indicating Rejected, MDA reported in the worksheet. Rejected peaks are discarded and not considered for further inclusion in the report. The WAN32 software attempts to fit a Gaussian peak to each photo-peak. If counts are present, but the peak-fit statistics are poor, a Bad Peak Shape flag is shown. In most cases, a bad peak shape flag is consistent with a "false peak", and that nuclide is rejected.

Point-Source Geometry Correction

On-site personnel determine which type of geometry correction is most appropriate before making a measurement. Typically items with a simple geometry such as pumps or valves, or even short pipes may be best modelled as point sources. In these cases, the geometry correction is given by:

$$k = \left(\frac{d_{measurement}}{d_{calibration}}\right)^2 \qquad (Eq.\,1)$$

Where d is the distance in cm.

Line-Source Geometry Correction

Items that are more complex than a simple point source, and behave like a line, such as a pipe or a duct are modelled using a line source geometry correction given by:

$$k = \frac{d_{measurement}}{2 \, \theta \, d_{calibration}^2} \qquad (Eq. \, 2)$$

Where θ is the physical half-angle in radians.

The geometry corrected result is in units of activity per unit length. In order to obtain the total activity the result is multiplied by the length of the line source.

Area-Source Geometry Correction

Many items with flat or nearly flat surfaces such as a glove box or hood floors, walls, and filters can be measured using the Generalized Geometry Holdup (GGH) model. This method is used to calculate the areal activity within the field of view. The geometry correction is given by:

$$k = \frac{1}{2 \, \pi \, \sigma^2} \qquad (Eq. \, 3)$$

Where σ is the Gaussian Parameter for the specific detector/collimator combination.

Complex Geometry Correction – Microshield and MCNP

The collimated/shielded detector field of view (FOV) is typically a conic projection calculated and based upon the depth and diameter of the collimator. The center of the detector FOV is targeted to be approximately normal to the center-line of the tank or object. A fundamental assumption for tanks is that measurements are performed on radioactive materials that are homogeneously fixed to the inside surfaces of the tank. Since this assumption does not support the use of a point, line, or area (GGH) model, a point-kernel attenuation or Monte Carlo type model is needed. The Microshield code has the ability to model a surface-contaminated cylinder, and is chosen for calculating the geometry correction for tanks. The tank measurement geometry is shown in Figure 8, below.

For consistency, attenuation correction is performed in the Excel® workbook rather than within Microshield. Tanks are modeled with a material density of 10^{-6} g/cm^3. This causes the model to not make any substantive attenuation corrections, and yields a pure geometry correction. Each cylinder is modeled and the un-collided fluence at a measurement point is calculated. This is repeated for a point source at the same distance and intensity. The geometry correction is:

$$k = \frac{Fluence_{Cylinder \, source}}{Fluence_{Point \, source}} \qquad (Eq. \, 4)$$

ANTECH uses MCNP - *A General Monte Carlo N-Particle Transport Code,* Versions 4 and 6 for modelling complex geometries [2]. Application of geometry correction factors is performed case-by-case in the event that MCNP is used.

ANTECH has its own point-kernel modelling code, ISOCorr [7], which can be used to generate geometry corrections for homogeneous drums and waste boxes.
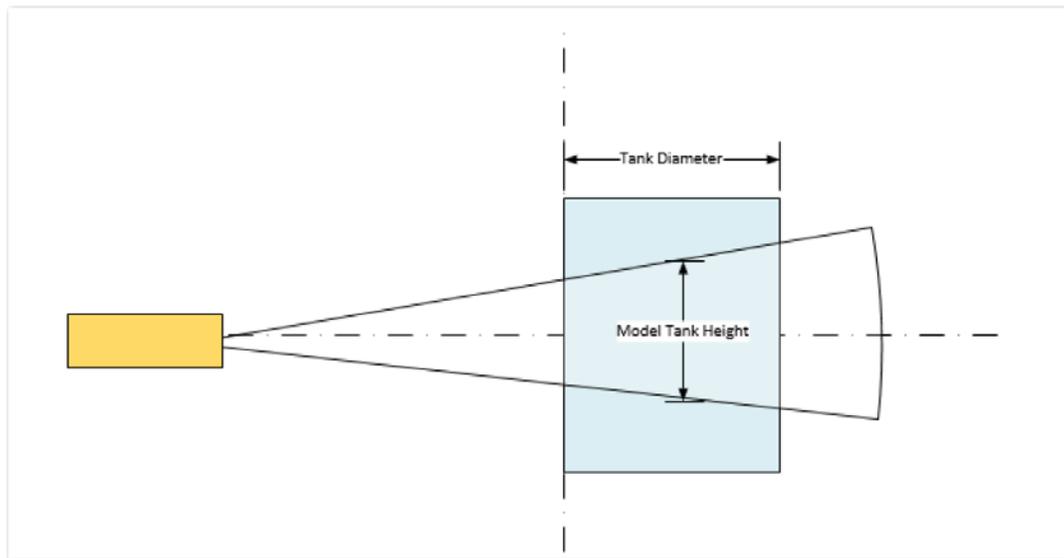
Fig. 8. Tank Measurement Geometry

Attenuation

Corrections are made for gamma ray attenuation for both air and any other shielding between the detector and the measured item. Mass attenuation coefficients (μ_m) are adapted from the National Institute of Standards and Technology website. Linear interpolation is used to calculate the specific μ_m for each nuclide at its key energy line as identified in the GammaVision® library. An attenuation correction factor is calculated as follows:

$$CF_{Attenuation} = \frac{1}{e^{(-\mu_m * density * thickness)}} \qquad (Eq. 5)$$

Corrections Applied to GammaVision® Output

Each measured activity reported by GammaVision® is corrected as follows:

$$Activity_{Reported} = Activity_{GammaVision} * k * CF_{Attenuation} \qquad (Eq. 6)$$

Measurement Control

Quality Control measurements are employed to demonstrate detector system functionality and control. Daily Performance Checks are an integral part of the HPGe Measurement Control Program. Control limits for the HPGe system are developed during the initial set up of the measurement system in each facility using compiled data from measured backgrounds, certified source standards, and HPGe calibration verification measurements. The MGGA software automates the measurement control process and displays graphs showing the behaviour of centroid position, resolution, and efficiency over time. A typical Measurement Control screen from the MGGA code is shown in Figure 9.

Fig. 9. Display of a typical Measurement Control screen from the MGGA code.

The detector system is operated only if its performance is within the established range. If a performance check falls out of range, the result is investigated, and corrective actions are taken. Due to its extreme stability of HPGe detectors, it is rare for performance check measurements to fail with HPGe detectors systems.

MEASUREMENT EXAMPLES

Example No. 1 - Sink Drain

A single measurement was made with the HPGe detector horizontal to the sink drain trap and a point-source model was used to approximate the residual activity at that location. Attenuation through plexiglass was used as an approximation to the drain material. A photograph of the measurement geometry and gamma ray spectrum are shown in Figure 10.
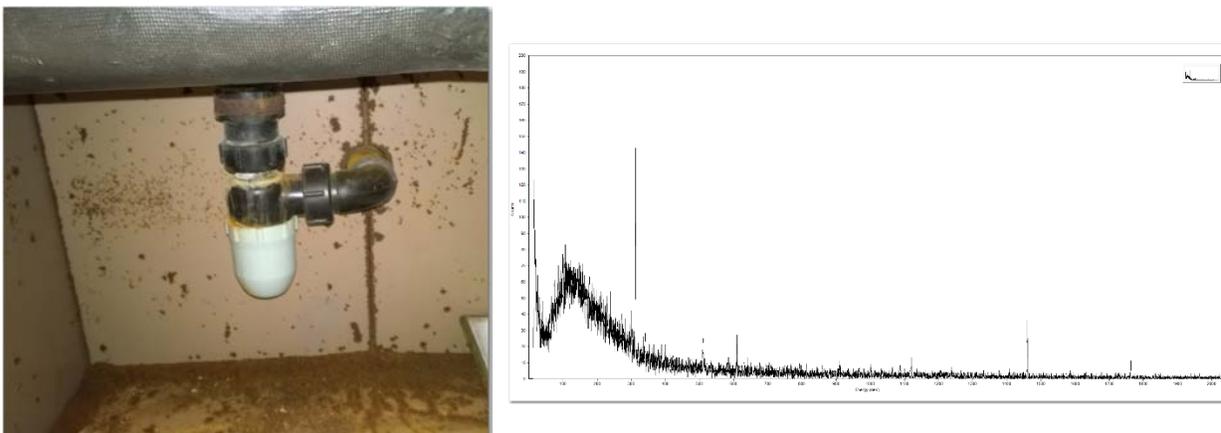

Fig. 10. Photograph of measurement location and measured gamma ray spectrum – Sink Drain

Example No. 2 – A Compactor Exhaust Duct

A measurement was made with the HPGe detector oriented vertically to include a section of a compactor exhaust duct. The residual activity in the entire duct was determined using a line-source model and by extrapolating the linear activity over the entire length of the duct. The attenuation correction was applied for the 18G steel wall of the duct and for air. A photograph of measurement geometry and the gamma ray spectrum are shown in Figure 11.
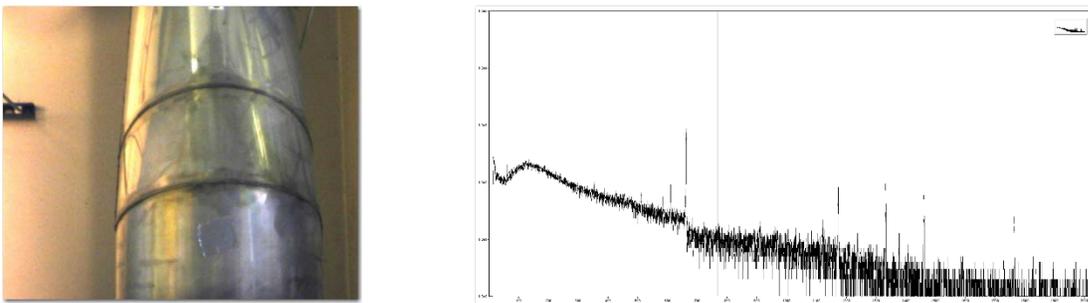

Fig. 11. Photograph of the measurement location and the measured gamma ray spectrum – Compactor Exhaust Duct

Example No. 3 – A Hood

A measurement was made with the detector oriented at a downward angle such that the field of view covers the bottom of the hood. An areal-source GGH model was used to determine the areal activity, which was extrapolated to the bottom surface of the hood and to 20% of the sides, back, and front (movable sashes). Since the sash was open during the measurement, attenuation correction is made for air only. A photograph of the measurement geometry and gamma ray spectrum are shown in Figure 12.
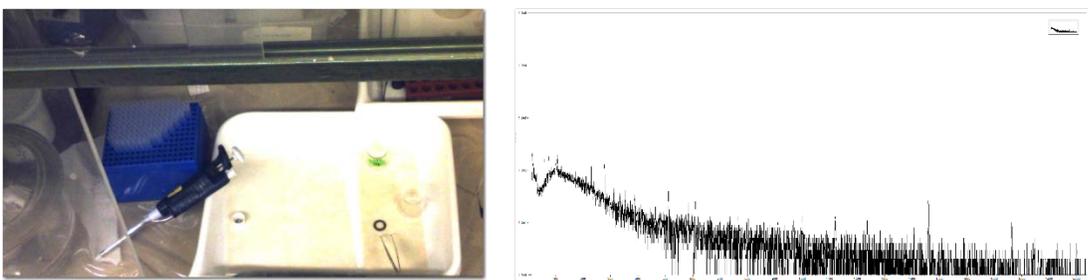

Fig. 12. Photograph of the measurement location and measured gamma ray spectrum – Hood

Example No. 4 – An Empty Waste Tank

In this example, the detector was horizontal to the center of the waste tank. A complex geometry model is used for geometry correction in this case. For the tank, a surface contaminated cylinder Microshield® model was developed to correct for geometry and attenuation through the steel wall of the tank. A photograph of the measurement geometry and gamma ray spectrum are shown in Figure 13.
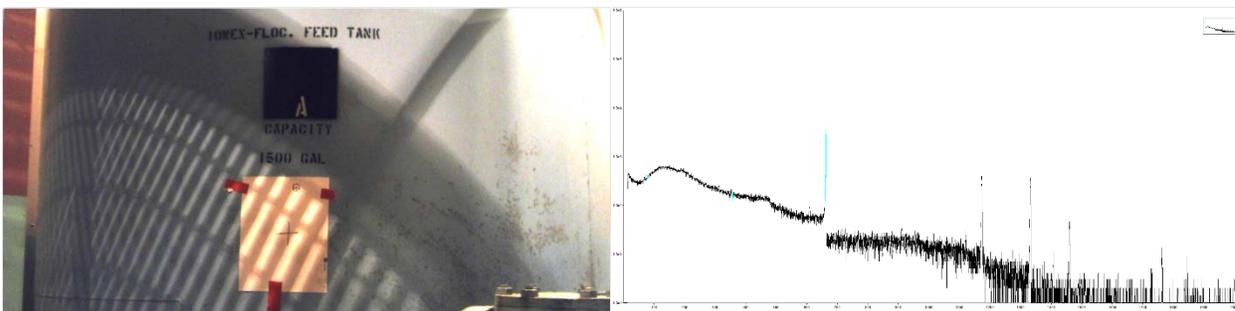
Fig. 13. Photograph of the measurement location and measured spectrum – Empty Waste Tank

Example No. 5 – A "Hot Spot" in a Glovebox

In this example, one is able to "zoom in" on a single object within a Glovebox. Here, a lead pig containing a radioactive source is assessed. Using a long tungsten collimator with a very narrow field of view, it is possible to measure a single object, rather than the entire Glovebox. By attenuating through the known thickness of the lead pig, one can determine the activity inside the pig. Photograph of measurement geometry and spectrum are shown in Figure 14.
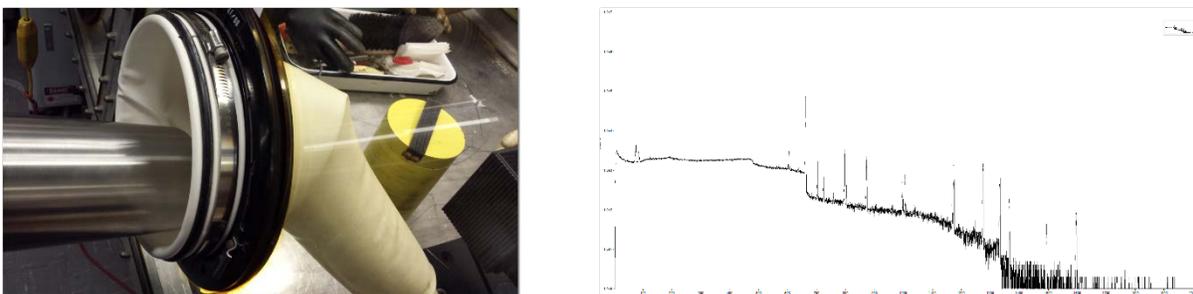

Fig. 14. Photograph of measurement location and measured spectrum – Hot Spot in a Glovebox

Example No. 6 – A non-Homogeneous Waste Drum

This is a measurement of a "Hot Spot" of radioactivity identified by RadSearch® within a 30-gallon waste drum. The contents were described as waste rags and PPE. A point-source model is used to assay the hot spot. The attenuation correction was applied for the air, the drum wall, and the drum contents that overlay the Hot Spot. Figure 15 is an image of the scanned drum.
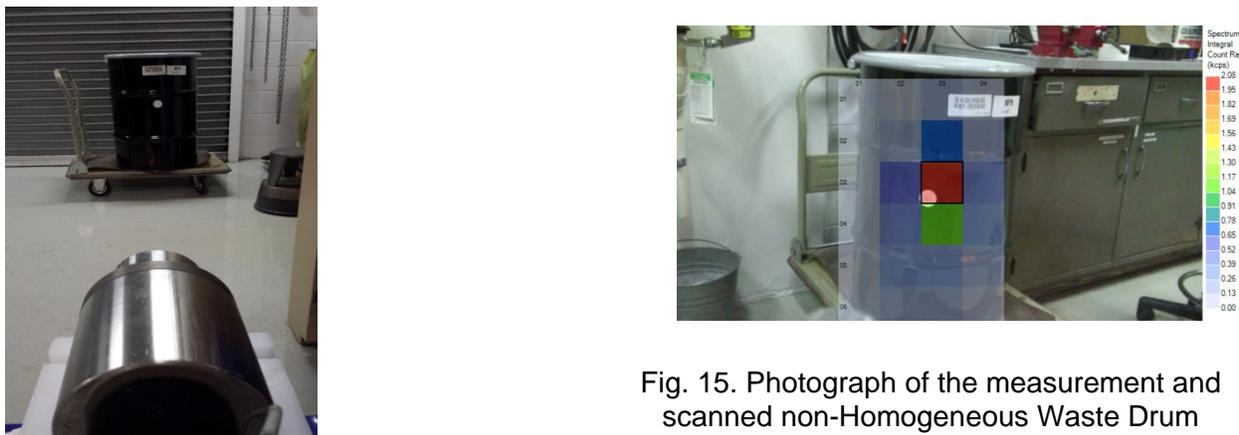

Fig. 15. Photograph of the measurement and scanned non-Homogeneous Waste Drum

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CONCLUSIONS

An Integrated Measurement Process to facility radiometric characterization for non-uniformly distributed radiation sources has been developed. It involves the use of the Multi Geometry Gamma Analysis code to control and manage the data acquisition as well as facilitating the semi-automatic analysis and reporting of measurement data under the guidance of a Subject Matter Expert. The process can employ a range of radiation detectors and includes a comprehensive range of geometry corrections including the GGH protocols and complicated modelling using codes such as MCNP. The analysis of uniformly distributed sources using object measuring codes like ISOTOPIC and ISOCorr [7] can also be incorporated if required giving the process universal applicability.

A number of examples of the application of the Integrated Measurement Process have been considered in the paper. During its application to measurements at the Argonne National Laboratory the Process has improved efficiency of both measurements and data analysis and has reduced errors in data handling.

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