A Sodium Iodide and Lanthanum Bromide Detector Equipped Gamma Survey System for Enhancing Discrimination between Contamination and Naturally Occurring Radiological Material - 11350

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ABSTRACT

A radiological survey system is described that employs thallium doped sodium iodide detectors and cerium doped lanthanum bromide detectors deployed on GPS enabled mobile platforms. The overall system includes digital signal processing to enhance detection confidence, use of GeoSoft's Oasis Montaj for mapping the survey data, and additional software for analyzing maps for effectiveness of remedial actions.

The system discussed takes advantage of the improved energy resolution provided by lanthanum bromide and utilizes this capability via software developed to evaluate the effectiveness of remedial actions. Data used to demonstrate the process and advantages focus on the ability to discriminate between naturally occurring radioactive material and contamination from an anthropogenic source. The paper includes data collected from surveys to locate site contamination with depleted uranium to demonstrate components and versatility of the system.

Maps of area surveys for DU contamination are presented to demonstrate the variability of total count rates as a result of variability in levels of naturally occurring radiological material in addition to the presence of DU metal or oxides. Survey maps are used to identify selected areas for collection of soil samples for at-site long-term count rates using lanthanum bromide detectors. Additional sites are identified for sampling where count rates are low and can be assumed to represent background levels. Survey spectral data collected using lanthanum bromide detectors for areas of elevated count rates and from low count rates are consolidated to produce spectra for determination of Pa-234m to Bi-214 isotopic ratios.

Isotopic ratios are computed from survey spectra and from soil samples. These ratios are compared to ratios of background soil samples and those that have been doped with sufficient DU oxide to one half of the regulatory threshold and to the regulatory threshold. Isotopic ratios for samples counted at-site using lanthanum bromide detectors are verified with equivalent values determined for the same samples counted in a laboratory setting using a high purity germanium detector.

Software for masking areas of survey maps likely to be DU contamination to generate consolidated spectra from areas that have been masked with those that are not masked. Isotopic ratios from the masked and unmasked areas can be compared to estimate the effectiveness of remedial actions targeting masked areas.

INTRODUCTION

The use of hand-held meters/detectors for conducting surveys of large outdoor areas has been the baseline technology for several decades. However, advent of advances in digital equipment has dramatically enhanced the speed, accuracy, and utility of such surveys. The ability to combine gamma detection data with GPS location data has opened up a variety of opportunities that did not previously exist.

Hand-held surveys are best completed by gridding off the area to be covered, typically on a one-meter spacing between the gridlines. Placing an ensemble of detection equipment on a mobile platform that also includes GPS capability does more than just allow GPS registration of detection data. This capability provides the potential to have a computer generated grid pattern and navigation system for conducting the survey. Elimination of manual gridding of the survey area reduces the number of individuals needed to conduct a survey and the time required.

Additional advantages of modern survey systems include the ability to survey at a higher speed. Typical survey speeds using hand-held devices is approximately two square meters per minute. Surveys of areas with relatively even terrain and that are not overgrown with vegetation can be as high as two square meters per second.

A most obvious advantage of modern survey systems is the ability to generate GPS registered maps of the area. These maps can be used for targeting areas to be remediated or simply as a management tool to monitor an area for changes in radiological contamination.

Large 10 x 10 x 40 cm (4 x 4 x 16 inch) thallium doped sodium iodide (NaI(Tl)) detectors are the most frequently used units for mobile survey systems. The four to one aspect ratio of these detectors makes them good candidates for conducting surveys on one-meter grid spacing. Coupling inorganic halide detectors with components like ORTEC's DigiBase with its on-board multichannel analyzer provides options such as auto calibration to prevent energy drift and different modes of operation.

Mobile survey systems can be characterized with respect to a variety of performance capabilities. Detector height and survey speed couple with the isotopes of interest to produce a detection limit for the system. The more energetic the emission, the deeper underground the contamination can be detected. The faster the survey speed, the shorter the dwell time over the target and the higher the detection limit. Other factors such as the ability of the system to navigate around obstacles, the stability of the system on slopes or in wet areas, and the vulnerability of the detectors to damage are significant.

Two of the most obvious performance capabilities of the system are its ultimate detection sensitivity for the contaminants of interest and how accurately do the maps produced from survey data correlate to actual location of the contamination. These two parameters work against each other. For a given survey speed, the shorter the summing period for counts or spectra development, the more accurate the GPS positioning of the map. However, shorter count times will reduce the detection sensitivity, particularly if detection is to be made using the energy of emission from the isotope of interest. Ultimately, a combination of survey speed, summing time

for the count/spectra, and necessary detection sensitivity (tolerance for false negative detection) must be balanced to achieve a functional system.

This paper describes a radiological survey system that includes the use of cerium doped lanthanum bromide ($LaBr_3(Ce)$) detectors and specialized software for evaluating the survey maps. Lanthanum bromide detectors have better sensitivity, energy resolution, and temperature stability than NaI(Tl) detectors. Their anisotropic thermal expansion has restricted availability of crystals to small sizes until recently. Detectors are now available in the 76 x 76 cm (3 x 3 inch) size for approximately $35,000 each. Clearly, these detectors are not going to replace the standard sized NaI(Tl) unless their superior performance justifies it.

Table I gives the general comparative data between NaI(Tl) and $LaBr_3(Ce)$ detectors. This table demonstrates the physical properties of each scintillator crystal and the advantages of the $LaBr_3(Ce)$ units. The photo absorption coefficient for $LaBr_3(Ce)$ crystals is larger than the equivalent value for NaI(Tl). This means that the $LaBr_3(Ce)$ detectors do a better job of absorbing an incident gamma ray. This is reflected by the Xo value in cm, the distance of travel through the crystal that is required to completely absorb energy from a 511 keV photon. The quantum yield value Y column giving the number of scintillated photons emitted from a single 1.0 MeV gamma reflects the greater detection efficiency of the $LaBr_3(Ce)$ detectors. Finally, the relaxation time τ in ns shows the much shorter time required for emission of scintillated photons. The shorter relaxation time provides for greater energy resolution.

Table I. Key parameter data for cerium doped lanthanum bromide and thallium doped iodide detectors.

Scintillator	ρ (g cm^{-1})	Z_{eff}	Photo Absorption Coefficient 511 keV, (cm^{-1})	x_o (cm)	Y (ph MeV)	τ (ns)
$LaBr_3(Ce)$	5.29	46.9	0.065	1.64	61,000	17-35
NaI(Tl)	3.67	50.8	0.058	2.56	43,000	230

Figure 1 provides a graphic example of the differences in detection sensitivity and energy resolution between the $LaBr_3(Ce)$ and NaI(Tl) detectors. The two spectra provided in Figure 1 were collected a 10 second spectrum using 76 x 76 cm (3 x 3 inch) detectors for a 10 mCi Cs-137 sealed source.
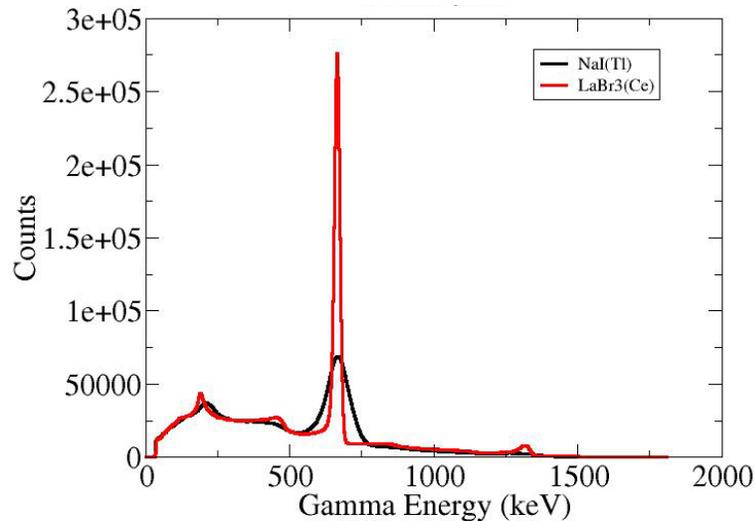
Figure 1. This figure clearly shows the sharper energy resolution of the $LaBr_3(Ce)$ detectors and the more Gaussian distribution of each peak. The combination of these two parameters enhances the applicability of digital signal processing to enhance detection capabilities.

The justification for using $LaBr_3(Ce)$ detectors for general field surveys comes not because of their higher performance than NaI detectors, but rather from their ability to substitute for high purity germanium (HPGe) detectors. The $LaBr_3(Ce)$ units do not have the energy resolving capability of HPGe systems, but they are capable of sufficient resolution to provide decision quality data in many applications. Additionally, they do not have the cryogenic cooling and energy requirements of HPGe detectors and are much more field-able. This makes them worthy of consideration for field applications when attempting to discriminate between background concentrations of naturally occurring radiological materials (NORM) and anthropogenic contamination. The example used in this paper is enhancing extraction of valuable information from surveys to discriminate between naturally occurring uranium and depleted uranium.

Description of Methodology

The Mississippi State University (MSU) Institute for Clean Energy Technology (ICET) gamma spectroscopy survey system utilizes two different mobile platforms; a motorized cart and a man-portable three-wheeled cart. Each is equipped with NaI(Tl) and $LaBr_3(Ce)$ detectors. These two platforms are shown in Figure 2. Position data are collected using differential GPS equipment. The three-wheeled cart uses a single Leica antenna and the motorized unit utilizes a four antenna Javad Triuph based system. The three-wheeled cart is used in areas not as amenable to the motorized system. Both utilize a process computer for data recording and monitoring operational status of all instruments.

Figure 2. Photos of two survey platforms used by the MSU ICET survey system. The right photo is a motorized platform equipped with a single 10 x 10 x 40 cm (4 x 4 x 16") NaI(Tl) and four 7.6 x7.6 (3 x 3") LaBr$_3$(Ce)detectors. The right photo shows a man-portable three wheeled cart equipped with a single 17.8 x 17.8 (7 x 7") NaI(Tl) and two 7.6 x7.6 (3 x 3") LaBr$_3$(Ce) detectors. Both platforms have on-board computers and GPS systems.

Data collected during the survey are stored by the onboard computer and ported for post collection processing and mapping using GeoSoft Oasis Montaj software. Maps are routinely generated using total counts; however, map layers are also generated using selected energy ranges and enhanced count rates enhanced by digital signal processing. Figure 3 provides an example of a total count map produced using Montaj using survey from a 10 x 10 x 40 cm NaI detector. This survey is typical of areas where naturally occurring variation of NORM exists.

It has been stated earlier that this paper uses surveys conducted to determine the location of DU contamination to demonstrate application of the hardware and software system. Depleted uranium contamination is frequently encountered at DOE and DoD sites. It is important to be able to discriminate areas that are contaminated with depleted uranium from areas that simply have elevated levels of naturally occurring uranium in the surface or near-surface soil.

The survey map shown in Figure 3 demonstrates variability in count rates that are not uncommon, particularly when the survey is attempting to identify low levels of contaminating radioactive material. Three areas of this map are identified for further evaluation. One area with elevated count rates, one appearing to be background, and another elevated count rate area that is a parking lot covered in crushed stone. Many mapping algorithms allow user control of the false color scale threshold levels. This allows the user to visualize small differences in low count rates as an aid in determining locations to collect soil samples. The color scale of this map has been set to enhance differences in count rates at or near background.
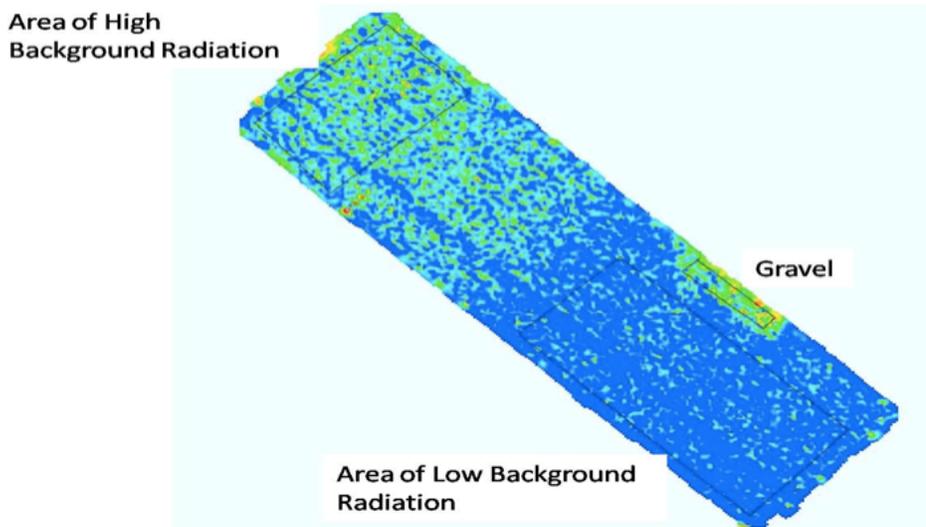
Figure 3. Map of area generated using total counts from a large NaI detector and demonstrating variability of count rates due to presence of naturally occurring radioactive material.

Maps like the one shown in Figure 3 are quite valuable for determining where to collect soil samples and or locating areas to be remediated. However, additional valuable insight can be extracted from the survey data. Additional software tools are necessary to accomplish a more extensive interpretation from the survey data.

Linear survey speeds for most systems like the one described in this paper are typically on the order of one to two meters per second. Survey data collected at these speeds are correlated with position data collected from the GPS equipment for generating maps. The time constant for determining count rates, survey speed, limits of detection, and accuracy of position are interrelated. The faster the survey speed, the greater the uncertainty associated with location or the higher the detection limit. The survey system described in this paper uses a time constant of 100 ms for determining count rates for speeds from one to two meters per second. This produces a relative small number of counts for each map position pixel. A spectrum produced from such a small number of counts has little definition, particularly in background areas.

Software has been developed to facilitate selecting a polygon portion of the map and composite all of the count data within that region into a single spectrum. The ability to collect detection data from a larger area increases total counts and spectral definition if the areas for compositing are judiciously selected. Detector data from the three survey areas identified in Figure 3 have been composited into spectra and are shown in Figure 4. The LaBr$_3$(Ce) spectra show higher resolution and both sets of spectra demonstrate that the variability in count rates originate from emissions in energies below 500 keV. Little or no variability is seen in the 1001 keV region.
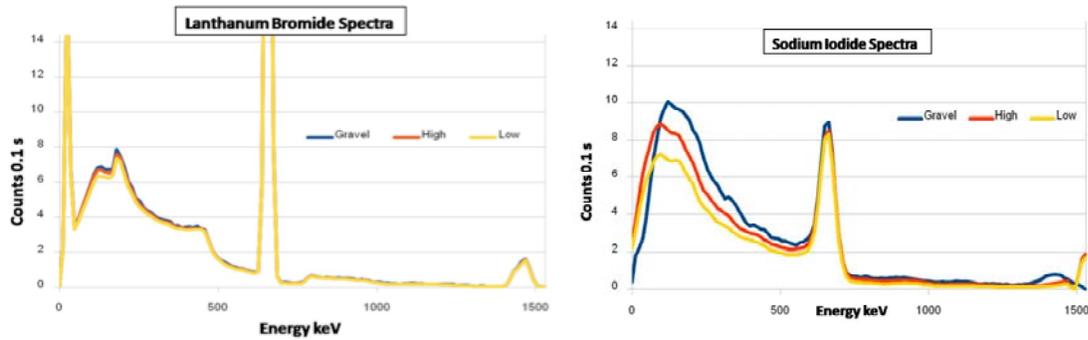
Figure 4. Spectra generated by compositing all count data from three different areas identified in the map shown in Figure 3. The left plot shows three spectra generated from LaBr$_3$(Ce) data and the right plot provides equivalent spectra from NaI(Tl) data.

The U-238 isotope is primarily an alpha emitter and difficult to determine in field surveys. The short-lived daughter Pa-234m is much easier to detect by its 1001 keV gamma. Virtually all field surveys for uranium oxides employ the 1001 keV photon of Pa-234m as the indicator for U-238 detection.

DU is almost exclusively U-238. The process of extracting uranium from ore separates it from virtually all of its decay daughter products. The enrichment process removes the majority of U-235 (and other U isotopes) from U-238 leaving it as the overwhelming constituent of DU. The U-238 isotope has a 10^9 year half life so the ingrowth of daughter products is a very slow process. One way to discriminate between naturally occurring uranium and DU is to determine the ratio of Pa-234 to Bi-214. This can be done chemically using ICP mass spectrometry or radiologically using a HPGe detector.

Figure 3 provides a typical HPGe spectrum of uranium ore. The 1001 keV photon for Pa-234m and the 609 keV photon of Bi-214 are both identified and it can be seen that the Bi-214 peak is much more clearly visible than the Pa-234m peak. An equivalent spectrum for DU oxide is almost devoid of the energies signifying U-238 daughters.
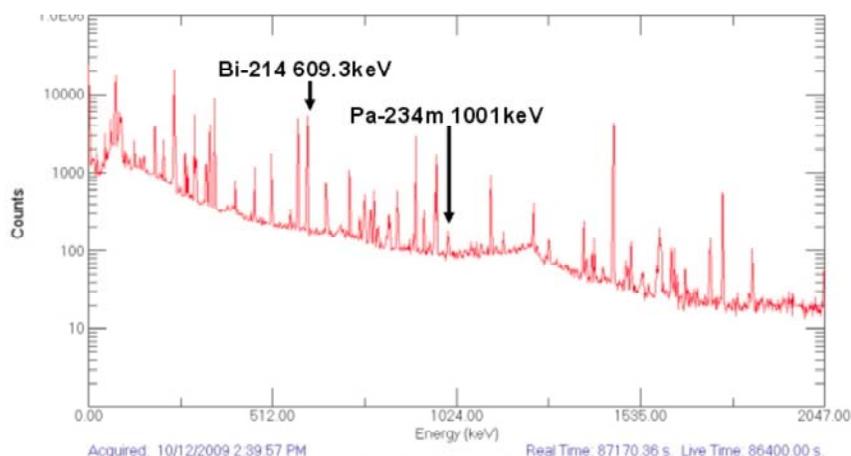
Figure 5. High purity germanium detector spectrum of naturally occurring uranium oxide ore sample.

The ratio of Pa-234m to Bi-214 should be constant for either DU oxides or for naturally occurring uranium oxides and its daughters. Comparison of spectral components for these two isotopes (Pa-234m to Bi-214 ration) yields an indication of the extent to which naturally occurring uranium has been contaminated with DU. Table II provides the ratios for naturally occurring uranium ore and for DU oxides. The dramatic difference in these rations (0.012 to 31,380) gives a sensitive measure of the extent to which DU adulterates the naturally occurring uranium.

Table II. Table of counts associated with Pa-234m and Bi-214 isotopes taken from the naturally occurring uranium ore high purity germanium detector spectral dataset displayed graphically in Figure 4. Also included are the equivalent data for a sample of depleted uranium oxide.

Source of Uranium	^{234m}Pa Net Counts (1001 keV)	^{214}Bi net Counts (609 keV)	Count Ratio ^{234m}Pa/^{214}Bi
Natural Uranium Ore	75,338	6,226,253	0.012
Depleted Uranium	5,334,559	170	31,379.76

Application of Methodology

The survey system described in this paper consists of the following tasks and capabilities shown in Table III.

Table III. Listing of individual components of the survey system along with infrastructure requirements for each activity.

Activity	Necessary Infrastructure
Survey data collection	Survey Equipment
Mapping	GeoSoft Oasis Montaj
Soil Sampling	Soil Sampling Material
Counting Soil Samples	Lanthanum bromide detectors
Compositing Spectra	Software for Production of Composite Spectra
Map Masking	Masking Software

Application of the full range of activities and capabilities of the method is demonstrated using data from two different surveys. Figure 6 provides a map segment from a survey conducted along a road where elevated count rates had been associated with DU contamination. This map shows locations where two soil samples were collected (sample numbers 9 and 10) and also shows areas selected for composite spectra production (high spectra and low spectra). This map represents two important aspects of the overall system of survey equipment, mapping software, soil sampling, and software tools for making maximum use of the survey data.
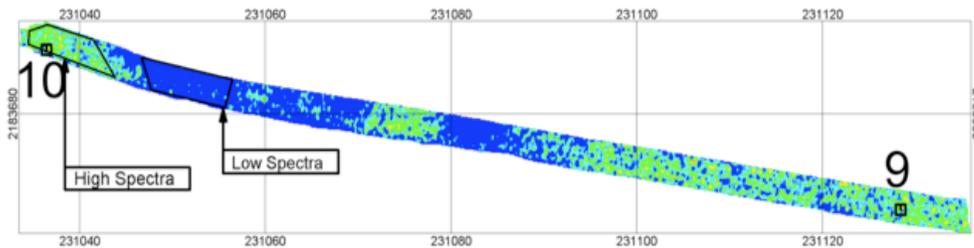

Figure 6. Map segment showing data collected during surveying a road segment to determine the presence of depleted uranium.

Acquisition of soil samples from areas known or believed to be free of contamination is an important activity that can be conducted before deployment for conducting the survey. These samples can be counted for 24 hours using both $LaBr_3(Ce)$ and HPGe detectors. They can then be amended by addition of an amount of DU oxide to equal half the threshold limit for remediation followed by recounting. This process is then repeated by increasing the concentration of DU oxide to the remediation threshold level.   Table IV contains isotopic ratios for derived by the above process. These isotopic ratios derived using soil samples native to the area can be used to evaluate those computed from survey data.

Table IV. Spectral ratios generated using background soil samples that have been doped with depleted uranium oxide to half the remedial threshold and to the threshold. Ratios are shown for data collected using both HPGe and LaBr$_3$(Ce) detectors.

	ICET LaBr and HPGe Detector Net Count Ratios for Controlled Samples											
Contamination Factor	**Th-234(63keV) : Bi-214(609keV)**				**Th-234(93keV) : Bi-214(609keV)**				**Pa-234m(1001keV) : Bi-214(609keV)**			
	LaBr		**HPGe**		**LaBr**		**HPGe**		**LaBr**		**HPGe**	
	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)
Neat Sample	0.011	0.014	0.247	0.016	0.026	0.016	0.513	0.016	0.010	0.006	0.027	0.006
Sample + 0.5 Threshold	1.277	0.039	11.478	0.147	6.543	0.179	18.176	0.228	0.060	0.020	0.790	0.016
Sample + Thres.	1.717	0.052	19.629	0.237	9.285	0.265	31.825	0.379	0.106	0.017	1.594	0.023

Survey data for areas identified as the high and low spectra areas on the map in Figure 6 are composited and spectral ratios are computed. These ratios are compared to values displayed in Table IV. Soil samples collected at locations of highest count rates such as samples 9 and 10 shown in Figure 6 are placed in Marinelli beakers and counted for 24 hours at-site using LaBr$_3$(Ce)detectors. Spectral ratios for these high-count locations are also compared to reference data displayed in Table V. Laboratory based analysis of the same samples using HPGe detectors have verified the accuracy of the conclusions drawn at-site using spectral ratios generated from the LaBr$_3$(Ce) at-site data.

Table V. Spectral ratios produced from analysis of soil samples collected from high count rate locations using LaBr$_3$(Ce) detectors.

On-Site Soil Analysis						
Sample Code	**Th-234(63keV) : Bi-214(609keV)**		**Th-234(93keV) : Bi-214(609keV)**		**Pa-234m(1001keV) : Bi-214(609keV)**	
	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)
2010-AUG03-00	0.032	0.022	0.116	0.026	0.001	0.010
2010-AUG03-01	0.000	0.000	0.078	0.046	0.011	0.013
2010-AUG03-02	0.000	0.000	0.033	0.023	0.000	0.000
2010-AUG03-03	0.024	0.040	0.097	0.024	0.000	0.000
2010-AUG03-04	0.000	0.000	0.050	0.019	0.000	0.000
2010-AUG03-05	0.095	0.043	0.062	0.024	0.005	0.009
2010-AUG03-06	0.034	0.020	0.007	0.022	0.013	0.008
2010-AUG03-07	0.047	0.018	0.059	0.020	0.023	0.015
2010-AUG03-08	0.033	0.017	0.072	0.019	0.011	0.014
2010-AUG03-09	0.040	0.020	0.046	0.022	0.020	0.016
2010-AUG03-10	0.055	0.020	0.066	0.022	0.007	0.008
2010-AUG03-11	0.035	0.017	0.050	0.020	0.000	0.000
Control Sample Ratios						
Contamination Factor	**Th-234(63keV) : Bi-214(609keV)**		**Th-234(93keV) : Bi-214(609keV)**		**Pa-234m(1001keV) : Bi-214(609keV)**	
	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)	Count Ratio	Uncer. (±)
+ 0.0x DU (Background)	0.011	0.014	0.026	0.016	0.010	0.006
+ 0.5x DU	1.277	0.039	6.543	0.179	0.060	0.020
+ 1.0x DU (Threshold)	1.717	0.052	9.285	0.265	0.106	0.017

All of the activities and data generation described above can be accomplished while on-site and a preliminary report can be rapidly compiled. Data displayed for the case reference above can be taken as evidence that high count areas are not contaminated with DU. Soil samples can be sent to a laboratory for conformational assessment using either HPGe detectors and/or mass spectroscopy.

An additional opportunity exists for using survey data to make remediation decisions when preliminary data indicate the presence of contamination.   Figure 7 shows segments of a survey map with localized contamination. Three views of the map show the standard display (left) detection locations that have been extracted from the background areas (center) and the map with detection areas masked (right). Software developed for automatically masking (extracting) detection locations from the survey data set facilitates development of composited spectra for detection areas and from the background. Spectral ratios from these spectra can be employed to evaluate the effectiveness of remediating contaminated areas
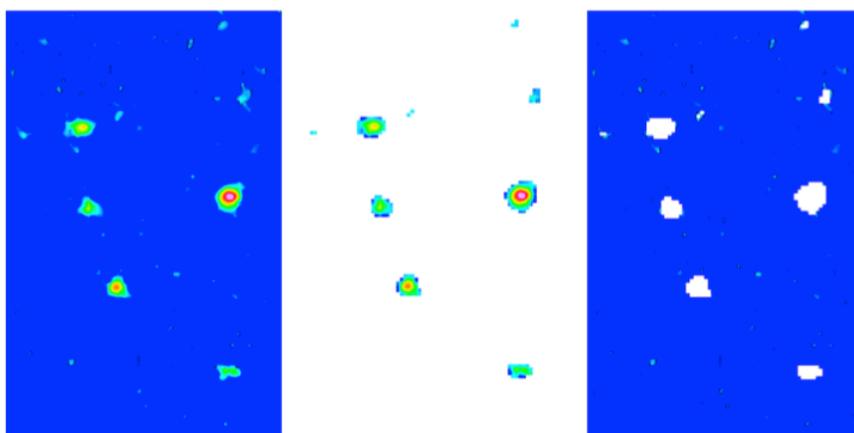

Figure 7. Three displays of a survey map that show detection locations (left), specific detection areas extracted from the overall map (center), and the map with detection areas masked (right).

Spectral ratios produced using the masking and spectra compositing software compared to reference data such as those contained in Table IV can be used to evaluate the effectiveness of remedial actions for large survey areas. The software criterion for selecting areas that are masked is under user control. This allows the user to identify locations for removals using either a total count threshold or a masking level derived to achieve acceptable isotopic ratios for the remediated area. This technique can be employed in consultation with regulators to increase confidence in the adequacy of remedial actions.

Conclusions

Lanthanum bromide detectors have been demonstrated as a valuable tool for collecting field survey data. These detectors have significantly better energy resolution and do not require the supporting infrastructure of HPGe detectors. Additionally, $LaBr_3(Ce)$ detectors have much

better counting efficiency than either NaI(Tl) or HPGe detectors. Spectral ratios generated using LaBr$_3$(Ce) detectors have proved effective in discriminating between areas with elevated background count rates and those contaminated with DU.

Software packages developed for conducting surveys and interpreting survey data have proved effective in compositing spectra from selected areas of a survey map for the purpose of calculating composite spectra. Masking software has demonstrated utility in evaluating the effectiveness of remediation of selected areas as a planning effort to optimize the remediation process. Post remediation surveys are used to verify effectiveness; however, a pre-action planning tool that can accurately predict the adequacy of removing selected detection material can optimize the process.

Acknowledgement