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ADVANCED TECHNICAL APPROACHES TO DETECT AND DISCERN TENORM IN COMMERCIAL TRANSPORTATION

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Abstract

Non-nuclear industries such as oil and natural gas production and processing, industrial coal combustion facilities, metal mining and processing, phosphate fertilizer and elemental phosphate production, and scrap metal processing (smelting) generate alarming quantities of TENORM by producing concentrated activities of NORM or by moving the radioactive materials where they potentially cause more human exposure. The existence of TENORM causes an increased risk for human exposure to radioactivity. Workers in TENORM-producing industries may be occupationally exposed to ionizing radiation. TENORM industries may release significant amounts of radioactive material into the environment, resulting in the potential for widespread exposure to ionizing radiation. Illicit trafficking of radioactive materials across the international borders is a global problem and currently being dealt with by establishing networks of sensitive radiation detection systems in the form of radiation portal monitors (RPM) at airports, national borders, highways, and ports of entry. The RPMs serve three major purposes: (1) the reliable detection and identification of threat materials, (2) the rapid characterization of non-threat materials comprised of NORMs and TENORMs and (3) the identification of legitimate radioactive materials in streams of commerce. Without proper adjudication the RPMs will create nuisance false alarms. The paper discusses advancement in spectroscopic analysis techniques and applications to examine gamma energy spectra collected by the RPMs. It discusses the methods to reduce noise and variance at different steps in data acquisition and analysis in the process of detecting the threat materials with low thresholds but without triggering on TENORMs. The practical applications of wavelet transform of gamma ray energy spectra will be explained. The success of reduction of dimensionality by principal component analysis (PCA) will be discussed within the framework of multichannel analysis spectroscopic data. The application of Detector Response Function (DRF) in the Gamma Detector Response and Analysis Software (GADRAS) software package developed at Sandia National Laboratories will be used to show how small amounts of threat materials masked by TENORM can be detected. The sensitivity to initial input parameters to GADRAS will be highlighted.

1. INTRODUCTION

Most soils and rocks contain low levels of naturally occurring radioactive material (NORM). Often NORM is concentrated through physical or chemical processing that results in technologically enhanced NORM called TENORM. It is formally defined as "naturally occurring radioactive materials that have been concentrated or exposed to the accessible environment as a result of human activities such as manufacturing, mineral extraction, or water processing." Examples of TENORM include fire brick, water and wastewater treatment residuals, coal ash, and decorative polished rock commonly used in building or home construction. It is also common for the native rock strata involved with the oil and gas extraction industries to contain somewhat higher levels of NORM, specifically natural uranium, thorium, and their decay products. TENORM spans a wide spectrum of raw materials and products meant for recycling or disposal. Below are listed some industrial sources of TENORM [1] that routinely produce ~25 pCi/g of radium-226 (in some cases orders of magnitude more than this nominal value)

- Petro-chemical industries including oil and natural-gas production, processing and refining industries
- Metal mining, processing including smelting facilities
- Phosphorus production and phosphate fertilizer manufacturing
- Coal industry and combustion of coal and its byproducts
- Water treatment facilities
- Geothermal energy production facilities

— Rare-earth extraction facilities
— Phosphoric acid manufacturing units

TENORM present unique problems because of their large volumes and widespread occurrence in industrial products, byproducts, and wastes. The physical, chemical, and radiologic properties of TENORM vary widely. Radium-226 and its decay products are the radionuclides of primary concern, but other uranium- and thorium-series nuclides should also be considered.

2. U.S. DOE TRIAGE SYSTEM

The United States Department of Energy (U.S. DOE) maintains continual (365 days a year, 24 hours per day, 7 days a week) scientific and technical support for the field responders deployed in radiological or nuclear emergencies, training, exercises, and perceived emergencies. A team of highly trained gamma spectroscopists—nuclear measurement specialists with considerable unique knowledge of improvised, state-built nuclear weapons and the nuclear fuel cycle—is on ready status to process and analyze field data and provide science-based resolutions of the problem in hand within an hour. Fig.1 shows gamma energy spectrum for an Eu-152 source taken by both a sodium iodide scintillator and a high-purity germanium (HPGe) semiconductor; the detectors have low- and high-energy resolutions, respectively. At a minimum, the information package sent out for Triage analysis should have the following items clearly identified:

(a) Gamma energy spectrum of background at "similar" environment as the target object
(b) Gamma energy spectrum of the target object
(c) Calibration gamma energy spectrum (with known nominal source strengths declared)

Other metadata, such as data obtained from contextual sensing (digital still or video pictures of the on-scene environment); neutron count rate and multiplicity data, any ground change detection data should also be included?

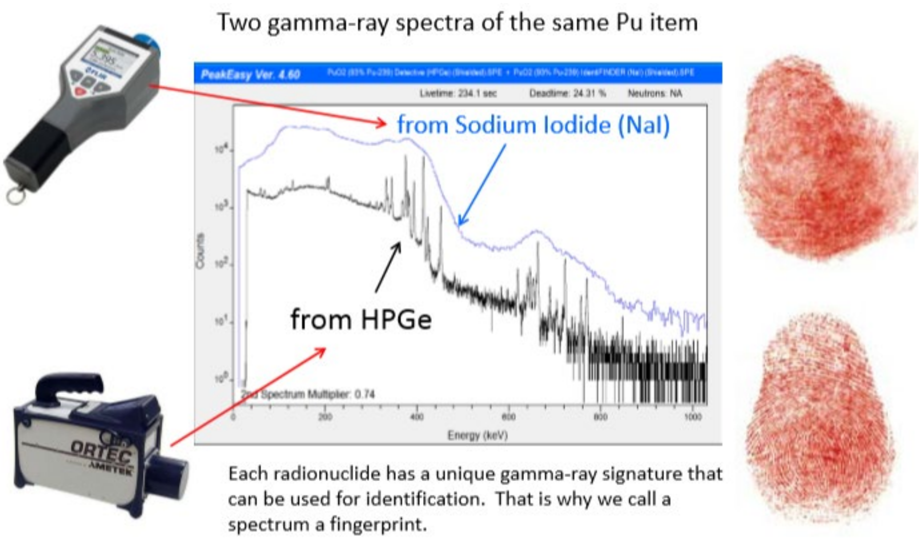

FIG. 1. A spectrum is a fingerprint. Comparison of a sodium iodide spectrum (low resolution) to a high-purity germanium spectrum (high resolution) for an Eu-152 isotope.

3. SYNTH

A simple but elegant parametric simulation code developed at the Pacific Northwest National Laboratory (PNNL) in the 1990s called SYNTH [2] (now in version 7) is still used to synthesize gamma ray energy spectra. SYNTH is a gamma ray spectroscopy interface to a deterministic physics engine designed to quickly model gamma ray measurements. It uses simplified models and physics (e.g., "transport" is handled by exponential attenuation,
$e^{-\mu x}$, without scattering or buildup, etc.). The emphasis is on peak energies, intensities, and shapes of the spectral lines; the details of the continuum are of secondary importance. The distributed pieces of original Visual Basic

codes organizes the problem into five logical components; users specify physical characteristics of a gamma ray source, the quantity of the nuclides producing the radiation, the source-to-detector distance, the type and thickness of absorbers, the size and composition of the detector (Ge or NaI), and the electronic set up for spectral acquisitio[2]. A spectrum is generated by first adding the full-energy peaks specified in the source term module to the spectrum. The form of an individual peak is a Gaussian with a single-exponential tailing function. The area of the peak is computed from the intensity of the specified source term (corrected for gamma ray branching ratios), the counting time, the geometry factor, the self-attenuation in the source, transmission through any absorbers, and the intrinsic detector efficiency.

The Compton continuum is added to the spectrum by taking the theoretical shape of a Compton spectrum, and normalizing it with a peak-to-Compton ratio appropriate for the volume of the detector. The impact on the spectrum due to other physical effects is also included, such as multiple Compton scattering (the region between the full-energy peak and the Compton edge), single and double escape peaks (from pair production), the difference in peak shape of gamma and x-rays (Gaussian vs. Lorentzian), the variation of resolution as a function of energy, and others.

The process of generating gamma energy spectra is deterministic (hard-coded algorithms), as opposed to stochastic (Monte Carlo methods); the spectrum is generated in minutes and is independent of count time. The codes produce a gamma energy spectrum that can be viewed and saved in a number of different spectral data formats. Fig. 2 describes a simplified overview of the SYNTH simulation process.

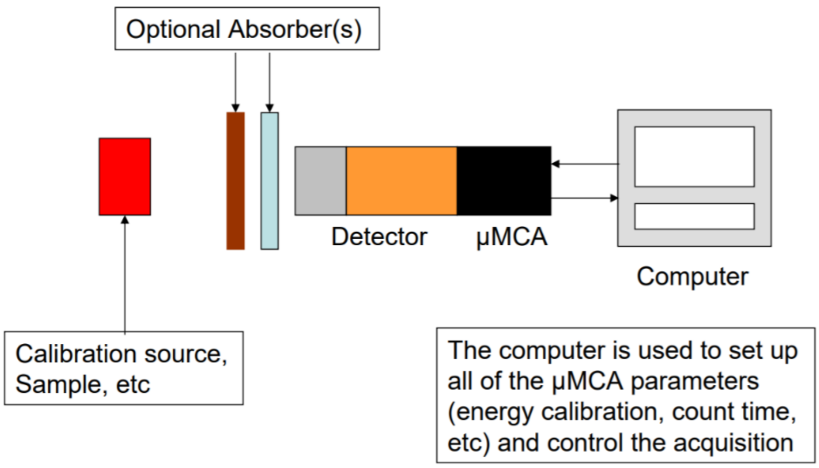

FIG. 2. SYNTH schema describing the processes involved in generating the gamma ray spectra.

Super SYNTH, a gamma ray spectroscopy interface to MCNP designed to simplify modeling of laboratory and real-world measurements requires MCNP to be installed and functioning to generate spectra, but MCNP is not needed if only an input deck is being generated. It organizes the problem into six logical steps. Super SYNTH generates a fully commented and functional MCNP input deck. It runs MCNP, then parses the generated *mctal* (Monte Carlo tally) file to produce a spectrum that can be viewed and saved in a number of different spectral data formats. Fig. 3 shows a gamma energy spectrum generated from combination of calibration sources (10 µCi of Cs-137, Co-60, Am-241 and Ba-133) measured by an HPGe sensor.

4. PEAKEASY

PeakEasy is an easy-to-use gamma ray spectroscopy analysis tool with a powerful graphical user interface. The analysis power of PeakEasy comes from the resident libraries of 500 common radioisotopes. This radionuclide reference tool enhances the speed and accuracy of the radionuclide identification and quantitative analysis of gamma ray spectra. PeakEasy can read and display spectra from over 200 different file formats. It can convert spectral data to several of the most popular file formats used by other analysis tools (for example **.cnf* files from Canberra Industries, **.chn* files from Ortec Industries, **.spc* files from FLIR Technology Systems; etc.). PeakEasy also includes a searchable gamma ray database (~30,000 gamma ray energies), data on each radionuclide, a batch mode processor, and multi-peak area analysis tools.

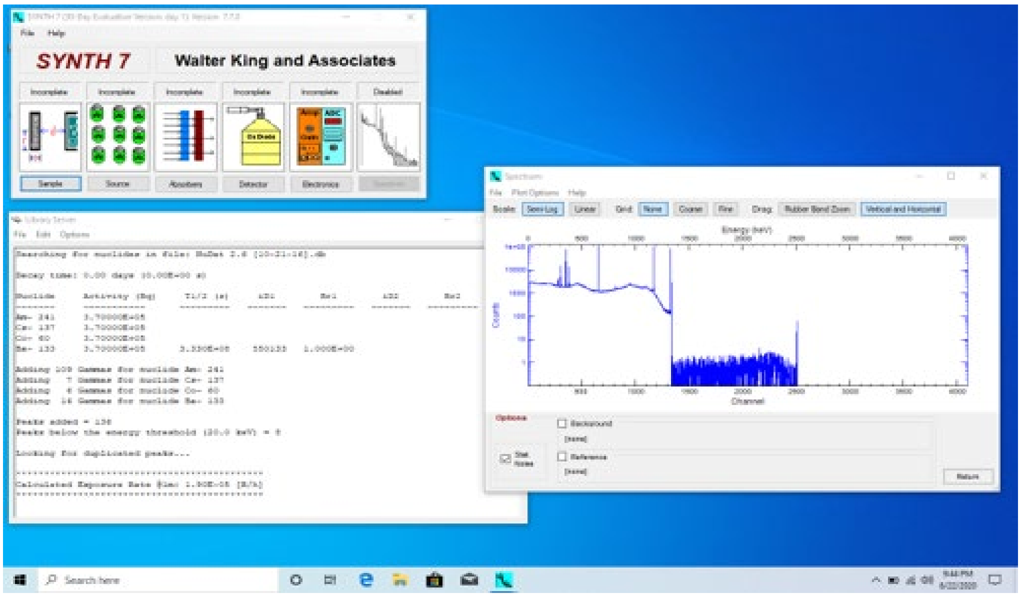

FIG. 3. A high-resolution simulated gamma-energy spectrum from a combination of calibration sources.

PeakEasy provides a few additional ease-of-use features:

- Easy and rapid energy calibration adjustments using slider bars
- Advanced calibration functionality with automated peak finder and polynomial least-squares calibration
- Batch mode processing (summing, file converting, appending, region-of-interest analysis)
- GPS locations extracted from file (must be connected to the Internet for this option)
- Easy summing of spectra taken over a selectable range of time interval during data acquisition
- Enhanced viewing and analysis tools for handling portal and search detector data
- Multiple Gaussian peak fitting with real-time display of fit results as calibration or region of interest is adjusted

PeakEasy deliberately does not perform any automated identification because it is meant to support the national laboratory spectroscopists in US DOE Triage system where individual expertise is required to make time-sensitive, situation-dependent, complex, difficult decisions regarding how to handle the materials involved within the context of the threats they pose. Fig. 4 shows ambient background gamma energy spectra taken with a sodium iodide scintillator (low resolution) and an HPGe semiconductor detector (high resolution). It provides a canvas to visually compare and contrast two spectra at a time. Fig. 5 shows the gamma energy spectrum collected with an identiFINDER spectrometer system. PeakEasy allows the analyst to determine the net counts under each of the two overlapping full energy peaks by defining nonlinear background counts under the peaks. The net counts under the multiple peaks from the same isotope, associated resolution of the spectrum at specific energy enhances the analysis process.

5. GADRAS

The GAmma Detector Response and Analysis Software (GADRAS), developed at Sandia National Laboratories by Mitchell and Mattingly [3] is a powerful gamma ray spectral analysis software toolset that is gaining applications in isotope identification and quantification by field deployed gamma detection systems. Carefully planned trade-offs are made while developing threat analysis metrics between very low false alarm rates and high throughput of cargo in commerce. GADRAS facilitates rapid spectroscopic analysis by parametrizing the detector response function to shorten the time to adjudicate a monitoring anomaly. GADRAS can compute and provide detector responses quickly and accurately without having to go through full-blown Monte Carlo simulation of the scenario and surroundings, giving users the ability to obtain usable results in minutes or less. GADRAS uses a one-dimensional discrete ordinate calculation to simulate gamma ray energy spectra. GADRAS

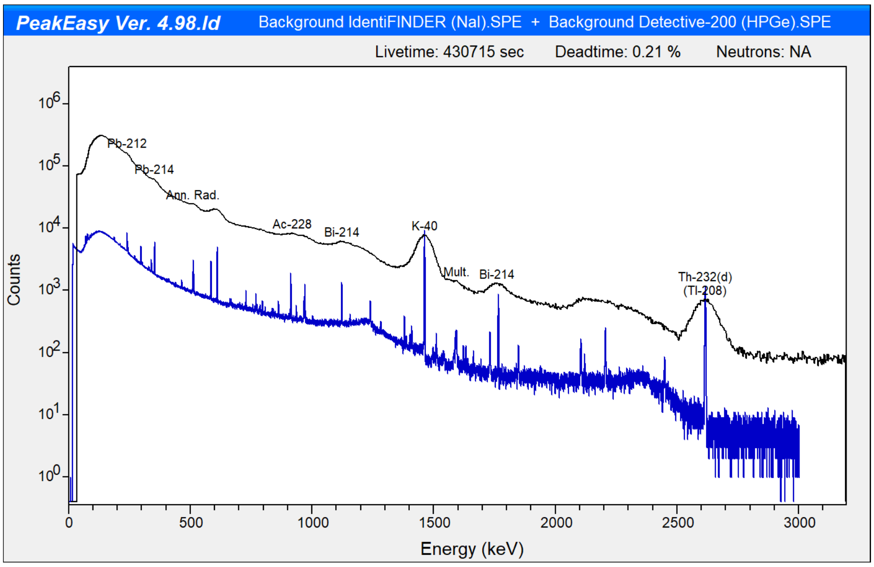

FIG. 4. Background gamma energy spectra taken by sodium iodide scintillator (low resolution) and HPGe semiconductor (high resolution) from PeakEasy library.

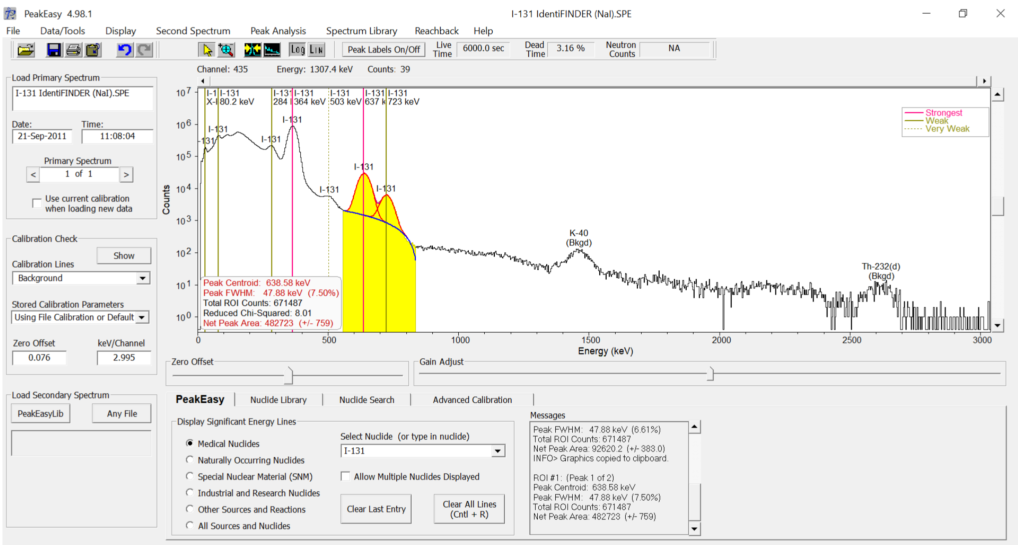

FIG. 5. Peak fitting and other gamma energy spectral analysis capabilities of PeakEasy are demonstrated.

performs a full-spectrum analysis, not just a full energy peak analysis. It can be used in analyzing gamma ray spectra of various energy resolutions from sodium iodide, cadmium zinc telluride (CZT), or HPGe detection instruments. The most powerful component of GADRAS is the detector response function (DRF) computation tool, which helps define detector characteristics using a minimum number of parameters. GADRAS is widely used across the DOE complex including the Triage system to analyze unknown gamma ray energy spectra acquired from a variety of handheld, portable and portal monitoring systems. The capabilities include characterization of detector response parameters, plotting and viewing measured and computed spectra, analyzing spectra to identify isotopes, and estimating source energy distributions from measured spectra. GADRAS-DRF can compute and provide detector responses quickly and accurately, giving users the ability to obtain usable results in seconds or minutes [4]. Fig. 6. describes pictorially how GADRAS can be used to analyze gamma ray energy spectrum from a unknown source with some knowledge of the environment under which the spectrum was collected and quantitatively determine the radioactive materials involved (Ra-226 in this case, 3.915 ±0.001 mCi) with high degree of certainty.

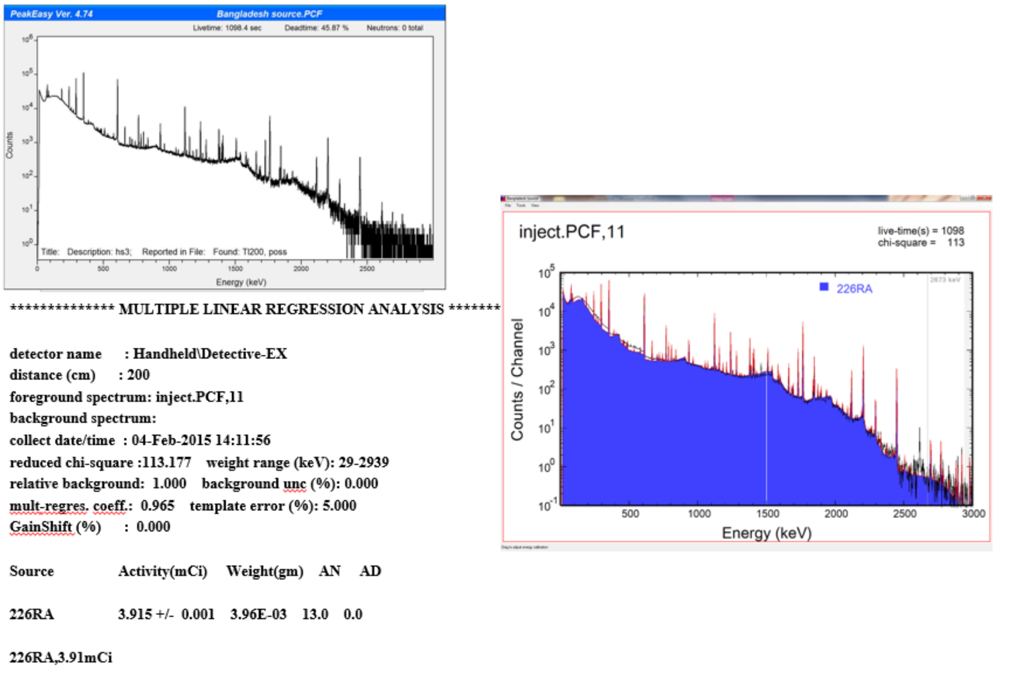

FIG. 6. Unknown source analysis. Example of application of the GADRAS toolset to quantitatively analyze radioactive materials.

GADRAS also uses neutron flux data. Since neutrons pass more readily through high-Z material and gamma rays pass more readily through low-Z material, different materials, such as in a container, will affect the total radiation output differently. As a result, using both gamma ray and neutron data improves the analysis.

6. PRINCIPAL COMPONENT ANALYSIS

Principal component analysis (PCA) is a multivariate statistical tool that uses an orthogonal transformation to convert a set of observations of possibly correlated variables into a set of values of linearly uncorrelated variables called principal components. The number of principal components are fewer than the number of original variables. The transformation is defined in such a way that the first principal component has the largest possible variance and accounts for as much of the variability in data as possible. Each succeeding component in turn has the highest variance possible under the constraint that it is orthogonal to the preceding components. The resulting vectors are an uncorrelated orthogonal basis set. The principal components are orthogonal because they are eigenvectors of the covariance matrix, which is symmetric. PCA is sensitive to the relative scaling of the original variables.

The goals of PCA, dimensionality reduction and independence, are achieved by transforming a multivariate dataset in such a manner that only a small fraction of new, uncorrelated dimensions (principal components) are needed to retain nearly all of the variation present in the original dataset. For any particular energy resolution, the number of principal components (dimensions) required to explain a particular desired fraction of the variation in the original data (e.g., 99.9%) provides a measure of the information content available in the original spectral population. As a qualitative example of PCA application we use three pairs of individual radioisotopes, Cs-137, U-235 and Pu-239, to obtain six separate spectra of gamma ray energy as collected by a sodium iodide–based detector, a typical radioisotope identification device, identiFINDER. The scaling of the counts was done by first converting counts C to square root of counts, $\sqrt{C}$, and then dividing the $\sqrt{C}$ values by the maximum value of $\sqrt{C}$ so the entire spectra is normalized to unity at the maximum. Fig. 7 shows the six spectra, two for each of the above mentioned isotopes.

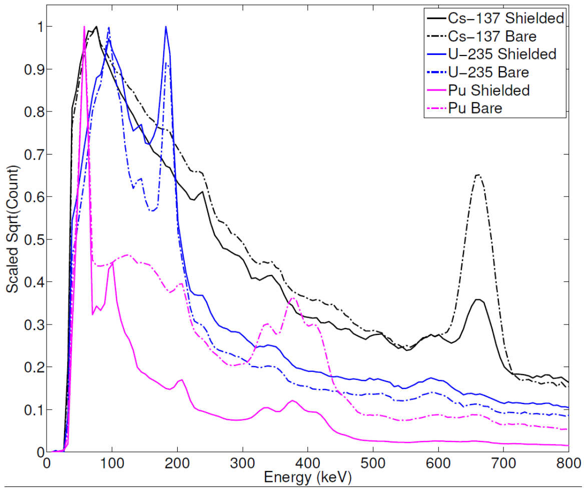

FIG. 7. Two scaled spectra from each of Cs-137, U-235 and Pu-239 are shown above between the gamma energy range of 0–800 keV.

Following the PCA analysis method prescribed in Ref. [5] the "within isotope" variability is displayed as well as "between isotope" variability. Fig. 8 describes the same six spectra reduced to two dimensions by three pairs of points (clusters). The values of the two coordinates in Fig. 8 are chosen so that the distances between each of the 15 ($^6C_2$) possible spectral pairs are very closely approximated (using multidimensional scaling) by the distances as computed using ordinary Euclidian distance applied to the 2-component values. Each isotope has a unique thumbprint that can be regarded as a cluster of points in the 2-component space.

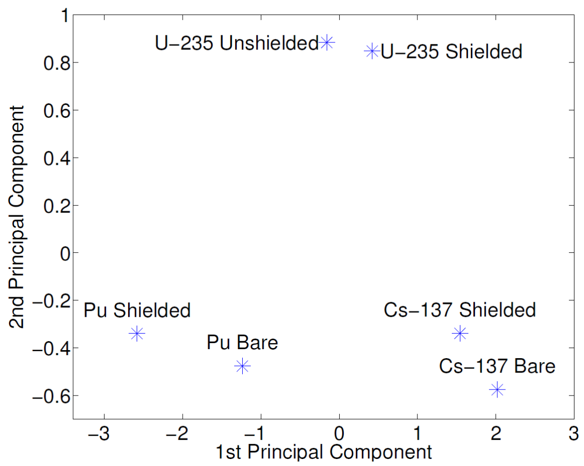

FIG. 8. There is a recognizable clustering of same isotope data points whether the spectrum was from bare or shielded source for each of the three isotopes. The complex energy spectrum has been reduced to two-dimensional plot still bearing the identity of the isotopes.

7. WAVELET TRANSFORM AND WAVRAD

7.1 Wavelet transformation of gamma ray energy spectrum

The gamma energy spectra from the field-deployable detectors are sometimes collected under complex situations of rapidly varying background radiation from nearby objects. It is conceivable that gamma signals from radio-nuclear threat objects can be masked by overwhelming TENORM signals. Gamma ray background is cluttered with transient collections from NORM and medical isotopes. Potentially threatening sources will likely be hidden in this noise; they may be encountered at large standoff distances and with low count rates. A rapid-response detection system derives an anomaly detection algorithm that characterizes low count sources as either threatening or non-threatening, and it operates well in the presence of high benign source variability. To reduce the background fluctuations and increase the signal-to-noise ratio observed by a detection system several de-noising techniques have been successfully used in recent years. One of the techniques utilizes wavelet transforms of a gamma energy spectrum by introducing a wavelet function ψ for timescale transformations. A wavelet is a square integrable function ψ whose translations and rescalings also provide a basis of square integrable functions. An example of a continuous wavelet is the "Mexican Hat" wavelet where, up to a normalization factor defined as

$$\psi_s(x)\, \alpha \left(1 - \frac{x^2}{s^2}\right) e^{\frac{-x^2}{s^2}}.$$

This function is dilated with a scale parameter α, and translated in time by τ as

$$\psi_{\alpha,\tau} = \frac{1}{\sqrt{\alpha}}\, \psi\left(\frac{t-\tau}{\alpha}\right).$$

A number of wavelets functions can be defined, choosing the proper scaling factor α. A continuous wavelet transform (CWT) [6] is defined by folding in the spectrum f(t) as

$$W_s(\alpha,\tau) = \frac{1}{\sqrt{\alpha}} \int_{t=-\infty}^{\infty} dt\ f(t)\, \psi\left(\frac{t-\tau}{\alpha}\right).$$

The CWT transforms one-dimensional information contained in a signal into two-dimensional plane. The wavelet transforms $W_s(\alpha, \tau)$ denote this transformation, which is termed the wavelet coefficients. An essential feature of the wavelet transform is that the wavelet coefficients provide information about both the central location and the spatial extent of the structures in the original signal. Identifying the structures that are present only on the lowest wavelet levels allows one to tag noise and small-scale structures and remove these from the spectrum.

7.2 WAVRAD

WAVRAD [7], or wavelet-assisted variance reduction anomaly detection, is an algorithm that mathematically compares features of a spectra to a collection of the previous spectra over a given time period. This algorithm has been shown to reduce the number of spurious alarms and does not depend on a source library of data. In addition, this algorithm points to areas in the spectra that give cause for the alarm, providing the field expert with additional information beyond the occurrence of the alarm itself. Finally, this algorithm is insensitive to slow changes over background radiation levels in the field.

WAVRAD provides a general algorithmic approach for real-time anomaly detection, impervious to common detector limitations (e.g., poor resolution, imperfect or nonlinear calibration, gain shift due to environmental reasons). The algorithm is based on the continuous wavelet transform for variance reduction, formation of an expectation from recent measurements, followed by evaluation of the deviation between the current measurement and the expectation using methods from linear algebra.

8. NSCRAD

The nuisance-rejection spectral comparison ratio anomaly detection (NSCRAD) method defines as independent variables certain ratios of spectral regions of interest called spectral comparison ratios (SCRs). The NSCRAD method (developed at PNNL) is designed to detect spectral anomalies and not to identify particular isotopes. However, the algorithm uses spectra from likely threat sources in choosing the broad energy regions that will be tested for count rate anomalies. Spectral regions of interest for use in calculating the SCRs were determined for three classes of special nuclear materials. Our particular partitioning of threats into three classes is a common sense one that is based on their gross spectral characteristics [8].

(1) Depleted uranium (DU)-like sources: Have characteristic features near 766 and 1001 keV from Pa^m-234, the first daughter of U-238. The DU-like spectra differ from that of natural uranium in that features from isotopes further down the decay chain are absent in DU.

(2) Highly-enriched uranium (HEU)-like sources: Exhibit the strong emission near 186 keV from U-235 in HEU.

(3) Plutonium (Pu)-like sources: Have a broad spectrum from about 100 to 500 keV from Pu-239 with no distinct features.

In practice, the embedded NSCRAD algorithm used in mobile radiation data analysis creates an anomaly metrics. In Fig. 9 the anomaly metric is compared to the nominal gamma alarms created by analyzing the gamma count-rate changes. The NSCRAD anomaly metric is much superior and easily distinguishable from the nominal alarm metrics.

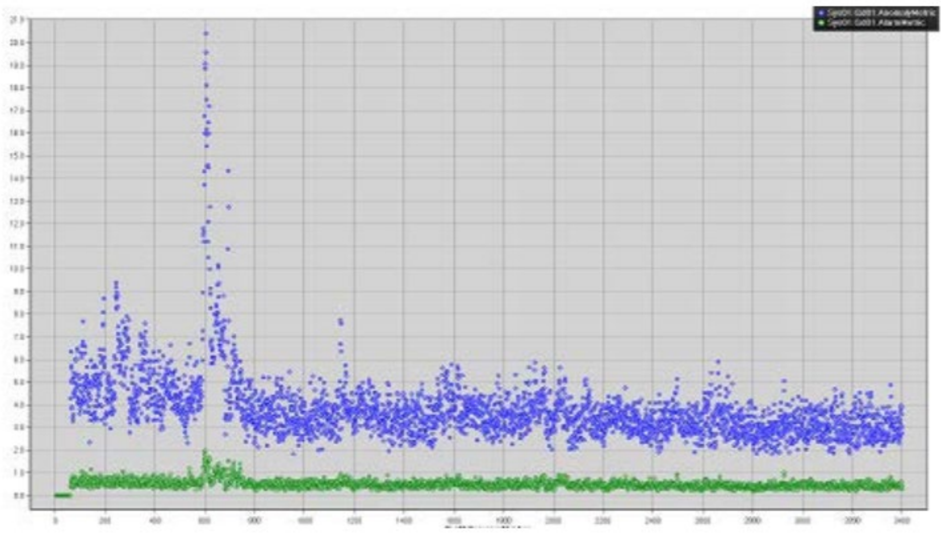

FIG. 9. NSCRAD anomaly metric compared to nominal gamma alarm metric.

9. CONCLUSION

Coexistence of radioactive materials out of regulatory control along with TENORM in the commerce poses a major problem for the national nuclear security postures. Ideally, law enforcement or other first responder groups should not be bothered with radiation alarms caused by NORM or TENORM. Simple alarm triggering algorithms based on count rate anomalies or even energy spectral anomalies are not sufficient to reduce the false alarm rates in a static or dynamic measurement system to an acceptable minimum. Several complex techniques and protocols have been developed in the USA that allow radiation alarms to trigger if and only if threat materials are present that are different from NORM and TENORM. These methods take advantage of mathematical toolsets like principal component analysis or wavelet transformation of gamma energy spectra to accentuate the true radio-nuclear threats. Additional measures, such as incorporating contextual sensor data along with the radiation measurement data, may alleviate some of these problems in commerce.

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