DEVELOPMENT OF THE SPECTRAL DECONVOLUTION ANALYSIS TOOL (SDAT) TO IMPROVE COUNTING STATISTICS AND DETECTION LIMITS FOR NUCLEAR EXPLOSION RADIONUCLIDE MEASUREMENTS

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ABSTRACT

The Spectral Deconvolution Analysis Tool (SDAT) is being written to improve counting statistics and detection limits for nuclear explosion radionuclide measurements. SDAT will utilize spectral deconvolution spectroscopy techniques to analyze both β–γ coincidence spectra for radioxenon isotopes and high-resolution High Purity Germanium (HPGe) spectra that are utilized for aerosol monitoring. Spectral deconvolution spectroscopy is an analysis method that utilizes the entire signal deposited in a gamma-ray detector rather than the small portion of the signal that is present in one gamma-ray peak. Counting statistics are improved by utilizing the entire detector response and the deconvolution algorithm directly handles interferences between radionuclides. This method shows promise to improve detection limits over classical gamma-ray spectroscopy analytical techniques.

The Multiple Isotope Comparison Analysis (MICA) algorithm was developed previously to demonstrate the concept for this technique for β–γ coincidence spectra utilized for radioxenon analysis. These spectra are difficult to analyze by the classical peak analysis technique due to spectral interferences among the radioxenon isotopes as well as the interferences between radon progeny and the radioxenon isotopes. The deconvolution algorithm unravels the interferences and utilizes the complete signal from each radionuclide. While the MICA algorithm has demonstrated its utility for the analysis of β–γ coincidence spectra, additional developments are necessary before this technique reaches a point where it may be applied in an operational environment. SDAT will incorporate the complete functionality of the MICA algorithm, will add in spectral weighting functions to reduce the analytical residual, and will include the ability to analyze high-resolution HPGe spectra with the deconvolution method.

A significant portion of this work will involve the development of calibration methods for both radioxenon and high-resolution HPGe systems. Proper calibrations of the detection systems are especially necessary for application of the spectral deconvolution spectroscopy algorithm. The detector response from each radionuclide of interest must be individually determined. The University of Texas TRIGA reactor will be utilized to irradiate a fission product generator for production of xenon isotopes for calibration. Fission products will also be generated for calibration and testing of the SDAT algorithm for HPGe spectra. Calibrations will be conducted through experimental measurements and will also be supported through Monte Carlo N-Particle Extended (MCNPX) modeling.

OBJECTIVE(S)

Background

Spectral deconvolution algorithims have been investigated for use on the analysis of β-γ coincidence spectra from the Automated Radioxenon Sampler/Analyzer (ARSA) (Biegalski and Biegalski, 2004). The ARSA was initially developed by Pacific Northwest National Laboratory (PNNL) for the purpose of monitoring atmospheric radioxenon levels for nuclear explosion monitoring (Bowyer, 1998). This system utilizes a β-γ coincidence spectroscopy system that acquires energy dispersive data on both the beta energy and photon energy axes. The initial software program written at the Center for Monitoring Research to analyze the ARSA data was *rms_xanalyze* (Biegalski, 2001). This algorithm is based on a Region of Interest (ROI) approach similar to most high-resolution gamma spectroscopy algorithms, but applied in two dimensions. The main problem arising with the *rms_xanalyze* algorithm is that not all the physics of the problem are taken into account (such as ^{135}Xe contributions in the E_γ = 30 keV region), which can result in negative net counts or higher than necessary false positive detection rates for some radioxenons. To improve the data processing for such β-γ coincidence data, work has been underway on a new software algorithm that incorporated all known physics and reduced the uncertainties involved in the results.

The new software involves the deconvolution of a sample signal into the contributions from each isotope. Detector-specific responses for each possible isotope are used in the deconvolution. The responses can be generated using actual radioisotope sources counted on the detector, or they can be created using modeling techniques like the MCNPX code. The geometry and materials of the detector must be known to produce good response files. Because only five different isotopes are detected in xenon samples, deconvolving a sample is not highly complex. Deconvolution of nuclear spectroscopy data is not a new concept (Prettyman et al., 1995). What makes this research unique is that this methodology is applied to 3-D β-γ coincidence data.

Required Input Data

The detector response for a sample consists of a 255 x 255 matrix of numbers. Each entry represents the number of counts registered in a certain $E\gamma$, E_β bin. Each row represents a γ-channel bin and each column represents a β-channel bin. We will refer to this structure as a histogram. Other information contained in the sample file includes calibration information and other sample characteristics, e.g., the sample Xe gas volume from which the total sampled atmospheric volume is calculated.

It is the sample histogram that will be deconvolved into individual isotopic responses using the MICA concept to determine atmospheric activity concentrations for each radioxenon of interest. To do this, however, we must have calibrated histograms of all the possible individual signals that can make up a sample histogram. These histograms should have the same size and calibration characteristics as the sample histogram in addition to good counting statistics. Therefore, we need the following detector response matrices with their associated activities:

$$\left[^{131m}Xe\right]_{255x255},\ \left[^{133m}Xe\right]_{255x255},\ \left[^{133}Xe\right]_{255x255},\ \left[^{135}Xe\right]_{255x255},\ \left[^{214}Pb\right]_{255x255},\ \text{and}$$
$$\left[DETBKG\right]_{255x255}. \quad (1)$$

Each detector response histogram can be generated using a detector modeling program like MCNPX or acquired by counting a calibration source on the detector. Since the ARSA has four beta detector cells, the above detector response histograms would need to be generated or acquired for each beta cell. In addition, the energy, resolution, and efficiency calibrations have to be determined for each beta cell. With all of this information at hand, and knowing in which detector cell the sample was counted, the following algorithm can be applied to a sample for determining the atmospheric radioxenon concentrations.

Concentration Calculation

First, the detector response histograms and sample histogram are vectorized, i.e., the 255 x 255 matrices are converted into column vectors of dimension 65025x1. This is done by appending each histogram row onto one another, and then transposing the resulting row matrix, i.e.,

$$\begin{aligned}
&\left[^{131m}Xe\right]_{255\,x255} \rightarrow \left[^{131m}Xe\right]_{65025\,x1}, \\
&\left[^{133m}Xe\right]_{255\,x255} \rightarrow \left[^{133m}Xe\right]_{65025\,x1}, \\
&\left[^{133}Xe\right]_{255\,x255} \rightarrow \left[^{133}Xe\right]_{65025\,x1}, \\
&\left[^{135}Xe\right]_{255\,x255} \rightarrow \left[^{135}Xe\right]_{65025\,x1}, \\
&\left[^{214}Pb\right]_{255\,x255} \rightarrow \left[^{214}Pb\right]_{65025\,x1}, \\
&\left[DETBKG\right]_{255\,x255} \rightarrow \left[DETBKG\right]_{65025\,x1}, \\
&\left[SAMPLE\right]_{255\,x255} \rightarrow \left[SAMPLE\right]_{65025\,x1}.
\end{aligned} \tag{2}$$

The separate vectorized detector response histograms are then assembled into a response matrix like so:

$$\left[\left[^{131m}Xe\right]_{65025\,x1}\ \left[^{133m}Xe\right]_{62025\,x1}\ \left[^{133}Xe\right]_{62025\,x1}\ \left[^{135}Xe\right]_{62025\,x1}\ \left[^{214}Pb\right]_{62025\,x1}\ \left[DETBKG\right]_{62025\,x1}\right]_{62025\,x6} \tag{3}$$

This results in a two-dimensional matrix with 65,025 rows and six columns, i.e.,

$$\left[\mathrm{Response}\right]_{65025x6} = \begin{bmatrix}
X & X & X & X & P & D \\
e & e & e & e & b & E \\
1 & 1 & 1 & 1 & 2 & T \\
3 & 3 & 3 & 3 & 1 & B \\
1 & 3 & 3 & 5 & 4 & K \\
m & m & & & & G
\end{bmatrix}_{65025x6} \tag{4}$$

The response matrix is related to the vectorized sample histogram by the following equation:

$$\left[\mathrm{Response}\right]_{65025x6}\ \left[Coefficients\right]_{6x1} = \left[Sample\right]_{65025x1} \tag{5}$$

The coefficient matrix holds the multipliers needed for multiplying the activities associated with each of the detector response histogram to obtain the sample activities for each isotope in the sample. This is a classic over-determined system of equations. The non-negative least squares solution was chosen to solve for $[\mathrm{Coefficients}]_{6x1}$ in this problem (Lawson and Hanson, 1974). This method was chosen over the standard least squares solution since all the components of the $[\mathrm{Coefficients}]_{6x1}$ matrix should be greater than or equal to zero.

This same analysis methodology may be applied to HPGe detector spectra as well. However, the library for the HPGe spectra needs to be much larger than for the ARSA spectra since there are significantly more component possibilities. Generation of the HPGe library is possible through individual measurement of each radionuclide of interest or through modeling of the detector with MCNPX.

RESEARCH ACCOMPLISHED

The research accomplished to date falls into two categories. The first category is the writing of the SDAT tool for the spectral analysis. The second category is the detector model generation in MCNPX that will be used to create simulated detector spectra.

SDAT Development

The SDAT development has been initiated in a Matlab environment. Spectral files and library files are stored as text files. SDAT reads these into the code as matrices. The deconvolution is performed via the Matlab least squares routine. Testing has been performed with both the standard least squares routing and the non-negative least squares routine that forces the solution to yield positive results. Both methods have produced the same results for our test spectra.

A spectral gain shifting algorithm has also been written in Matlab for SDAT. This algorithm is necessary to match the gain of the sample spectra with the gain of the library spectra. It may be used for either β–γ coincidence spectra or for HPGe γ spectra. For the β–γ coincidence spectra, the β energy calibration is treated as a second-order polynomial and the γ energy calibration is treated as a first-order polynomial. For the HPGe γ spectra, the energy calibration is treated as a fourth-order polynomial. The code takes the original energy calibrations and then determines transfer coefficients by taking into account the new energy calibration and the number of channels in the spectrum. If the code performs a negative gain shift (condenses data into a fewer number of channels), then the channels at the end of the spectrum are filled with zeros.

Work has also begun on a spectral weighting function algorithm. This algorithm weights parts of the spectrum according to their importance during the deconvolution. This algorithm helps reduce the residual of the deconvolution process and potentially improves the method detection limits. Currently the weighting process is binary: important regions of the spectrum are given a weight of one and unimportant regions of the spectrum are given weights of zero. A quality control routine is being worked on that will alert users if an unanticipated signal is being down weighted.

MCNPX Model Development

MCNPX has been chosen as the code to be utilized for generating the detector models. The reason for choosing MCNPX over MCNP is due the MCNPX's new mesh tallies, coincidence tallies, and plotting capabilities. New detector models have been generated that use the macrobody feature available in MCNPX.

Figure 1 illustrates the photon flux modeled in β–γ coincidence detector model with ^{133}Xe in the top cell. The model simulates the ARSA detector currently being utilized in the Provisional Technical Secretariat Noble Gas Experiment. The detector has four gas cells surrounded by two NaI(Tl) crystals. The beta flux from ^{133}Xe generated from this model is shown in Figure 2. Its first goal will be to qualitatively compare the data generated via the MCNPX model to data collected from the ARSA in China (shown in Figure 3). Initial testing was conducted with the coincidence tallies available in MCNPX, but they do not appear useful for this problem. A post-MCNPX algorithm will be generated to generate the coincidence spectra.

A MCNPX model has also been developed for a HPGe detector. This model is based off of a TennElec closed-end coaxial cylindrical detector with a crystal of diameter 59.5 mm and length of 59 mm. Figure 4 shows an example of this detector model. The model has been used to generate gamma-ray spectra and detector efficiency curves. Initial comparisons show a good correlation to data obtained directly from the detector.

CONCLUSION(S) AND RECOMMENDATION(S)

The work reviewed above had just begun. The SDAT software is still in its infancy state and a significant level of testing is required to optimize the spectral deconvolution algorithm. Data for the SDAT calibration libraries will be generated both through MCNPX models and through experiment. MCNPX models have been developed for both the $\beta-\gamma$ coincidence detector for noble gas detection and the HPGe detector for γ-ray spectroscopy. Work on obtaining fission product spectra through real measurements has not started.

REFERENCE(S)

K. M. F Biegalski and S. Biegalski (2001). Determining detection limits and minimum detectable concentrations for noble gas detectors utilizing beta-gamma coincidence systems, *J. Radioanalytical Nucl. Chem.*, 248: (3), 673.

K.M. Foltz Biegalski (2001). Automatic Analysis Algorithm for Radionuclide Pulse Height Data from Beta-Gamma Coincidence Systems, *J. Radioanalytical Nucl. Chem.* 248: (3), 663.

T.W. Bowyer (1998). Xe Radionuclides, Atmospheric: Monitoring in Encyclopedia of Environmental Analysis and Remediation, editor: R. A. Meyers, John Wiley & Sons, Inc.

G. Knoll (1989) *Radiation Detection and Measurement, 2nd Edition*, John Wiley & Sons, New York.

C.L. Lawson and R.J. Hanson (1974). Solving Least Squares Problems, Englewood Cliffs, N.J., Prentice-Hall, 1974.

T.H. Prettyman, R.A.Cole, R.J. Estep, G.A.Sheppard (1995). A Maximum-Likelihood Reconstruction Algorithm for Tomographic Gamma-Ray Nondestructive Assay, *Nucl.Inst .Meth. Phys. Res. A* 356: 470.

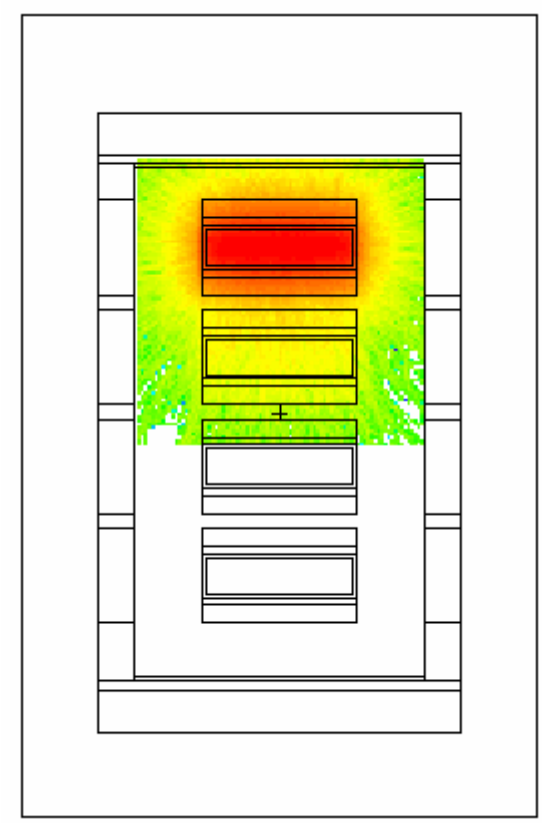

Figure 1. Map of photon flux from ^{133}Xe source in top chamber of β–γ coincidence detector MCNPX model.

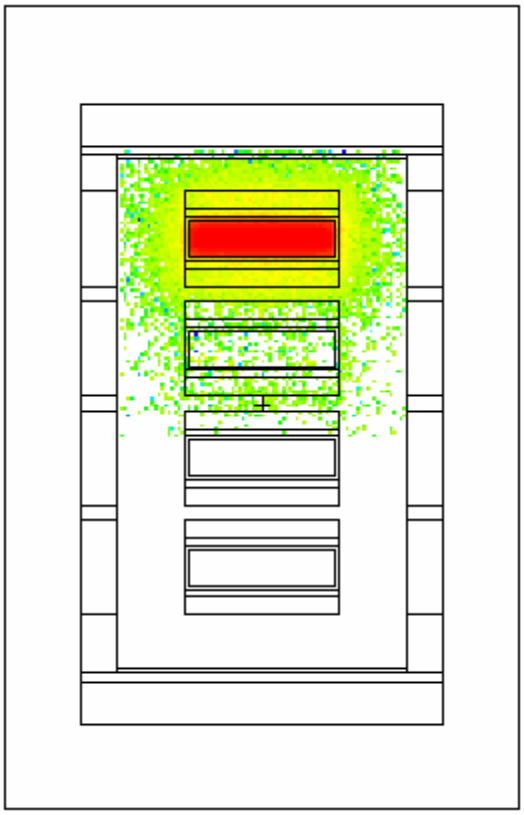

Figure 2. Map of beta flux from ^{133}Xe source in top chamber of β–γ coincidence detector MCNPX model.

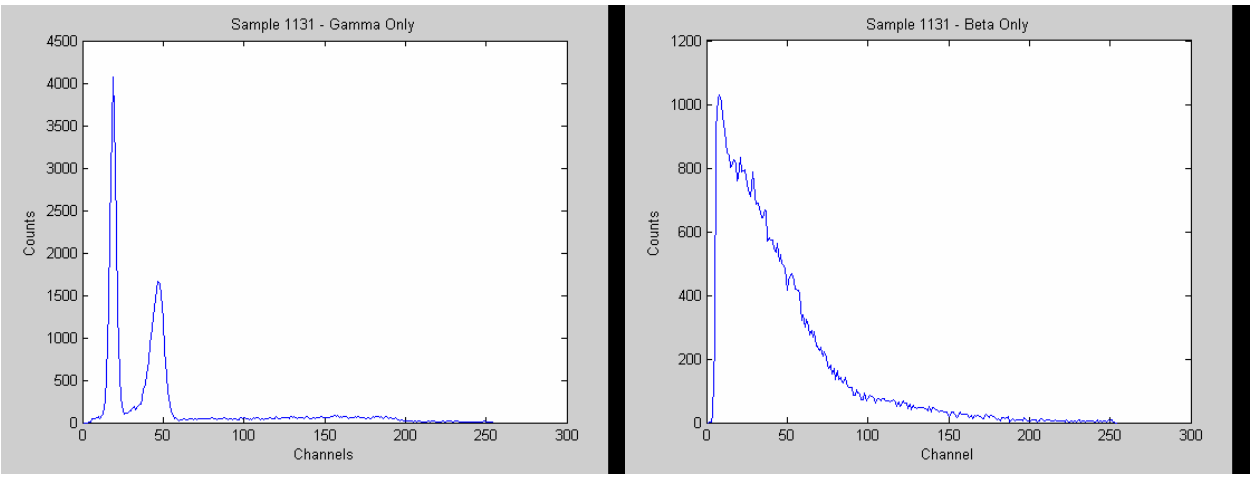

Figure 3. β and γ spectra generated from ARSA in China.

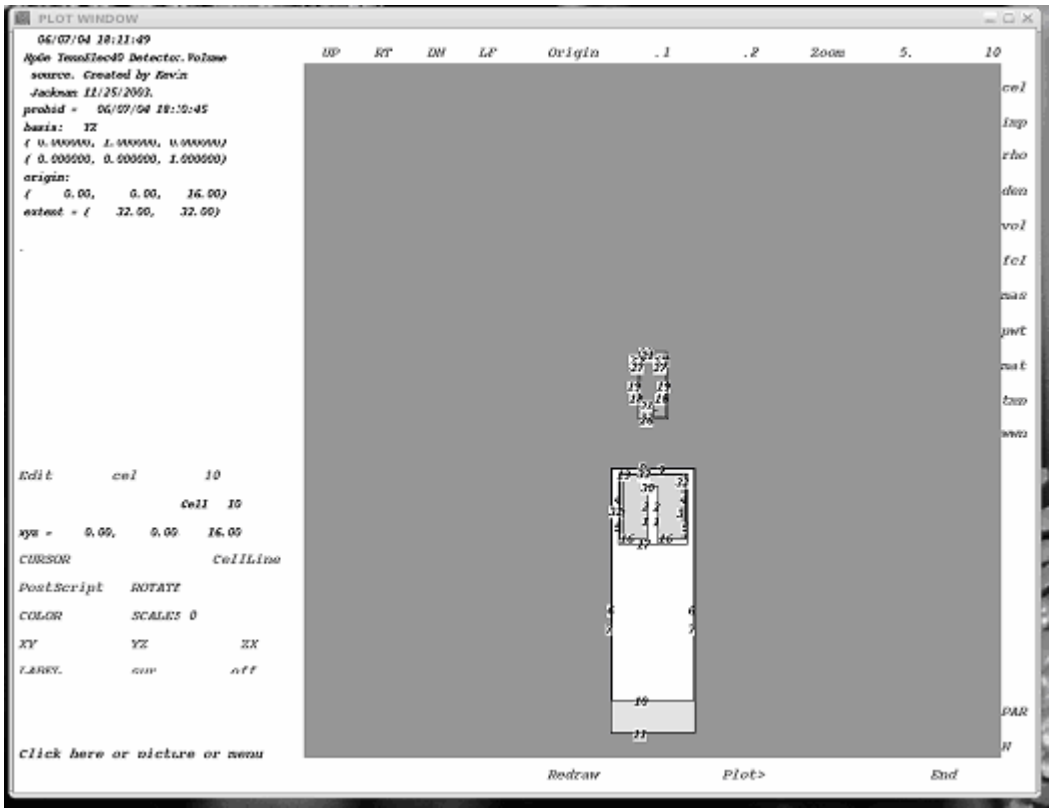

Figure 4. HPGe detector model.