TESTING THE SPECTRAL DECONVOLUTION ALGORITHM TOOL (SDAT) WITH XE SPECTRA

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ABSTRACT

The Spectral Deconvolution Analysis Tool (SDAT) software was developed to improve counting statistics and detection limits for nuclear explosion radionuclide measurements. SDAT utilizes spectral deconvolution spectroscopy techniques and can analyze both β-γ coincidence spectra for radioxenon isotopes and high-resolution HPGe spectra from aerosol monitors.

The SDAT tool has been integrated into a standalone graphical user interface (GUI). This GUI may import spectra, analyze the data for Xe concentrations, and graphically display the results. This tool has been tested with data generated via MCNPX models as well as real data.

OBJECTIVES

Environmental xenon sampling and measurement units are used in nuclear weapons test monitoring networks because radioxenon isotopes may be all that is released from tests performed deep underground or underwater. Some environmental xenon sampling and measurement units, like the Automated Radioxenon Sampler-Analyzer (ARSA) developed by Pacific Northwest National Laboratory (PNNL) and the Swedish Automated Noble Gas Unit (SAUNA) developed by the Swedish National Defense Research Establishment (FOI), use β-γ coincidence detectors that are energy dispersive on both the β and γ energy axes. Signals from four radioxenon isotopes (^{131m}Xe, ^{133}Xe, ^{133m}Xe, ^{135}Xe) comprise a sample spectrum. Under poor operating conditions, a few radon daughters (^{214}Pb, ^{214}Bi) may interfere with the sample spectrum. Applying conventional region-of-interest (ROI) spectrum analysis algorithms to such 3-D spectra results in relatively high minimum detectable concentrations (MDCs) due to the subtractive process of determining net counts in the ROI.

To overcome this problem, a project team from The University of Texas in Austin is developing software to deconvolve the 3-D sample spectra into the most probable combination of signals using the non-negative least-squares method. This method can use the entire signal from each radionuclide and consequently may improve the signal to noise ratio. It has been postulated that the use of such an algorithm will result in a better ability to resolve spectral interferences and thus improve counting statistics and detection limits for nuclear explosion radionuclide measurements. The SDAT GUI, expands upon a previously developed algorithm, the Multiple Isotope Component Analysis (MICA) tool.

The SDAT tool was tested against the CORIANT code (Foltz Biegalski, 2001) on data produced from an automated radioxenon sampler/analyzer (ARSA) unit when it was stationed in Guangzhou, China. The CORIANT tool utilizes a region of interest approach to the spectral analysis while SDAT utilizes a spectral deconvolution approach. Samples were selected that contain clear radioxenon signals. While a true test of SDAT will be the analysis of samples with low count rates, this test was conducted as a first step to show that the two methods produce similar results.

RESEARCH ACCOMPLISHED

This occurred in three steps. The first step was the creation of the SDAT tool. The second step was the creation of standard library inputs for the SDAT code, which was accomplished via MCNP simulations. The third step was to analyze samples with SDAT and compare the results to those produced with CORIANT.

Step 1: Creation of SDAT GUI

The detector response for a sample consists of a 255x255 matrix of numbers. Each entry represents the number of counts registered in a certain E_γ, 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.

The sample histogram will be deconvolved into individual isotopic responses using the SDAT 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:

$$\left[^{131m}Xe \right]_{255x255}, \ \left[^{133m}Xe \right]_{255x255}, \ \left[^{133}Xe \right]_{255x255}, \text{ and } \left[^{135}Xe \right]_{255x255}$$

Each detector-response histogram can be generated using a detector modeling program like MCNP 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.

The SDAT code was developed to input the library and sample files. These files are text files. The current version of SDAT just reads the sample spectrum and does not read any header information that may be associated with Sample Pulse Height Data (SPHD) formatted files. The open sample window is shown in Figure 1.

Once the proper files have been selected, SDAT analyzes the data and displays the results. Figure 2 shows the results display. The sample is displayed as a 2-D plot in the top left corner. Below the sample plot are three tables. The top left table is the Axis Parameters table which allows users to modify the minimum and maximum x and y intercepts of all the plots in the *SDAT* window. By default, they are set at 0 and 250. The Parameter/Value Table is situated to the right of the Axis Parameters table and contains a summary of the parameters specified in the *Open Single* window, i.e., the sensitivity number, the sample file name, and the library file names for each radioxenon of interest. This table is located in the bottom left corner of the *SDAT* window. It contains the concentration coefficients and errors of each radioxenon of interest calculated with and without the use of data weighting via the weighting matrix described earlier. The resulting multiplier coefficients can be used to determine the activity of each radioxenon using equation 1.

$$C_i = \frac{M_i A_i}{V} \tag{1}$$

where

C_i = Sample activity concentration of radioxenon isotope I;

M_i = Multiplier coefficient of radioxenon isotope i as determined by SDAT;

A_i = Activity represented by library file of radioxenon isotope I; and

V = Air volume sampled.

The activity concentration uncertainties of each radioxenon can be determined using equation 2.

$$\sigma_{C_i} = \frac{1}{V} \sqrt{\left(A_i \sigma_{M_i}\right)^2 + \left(M_i \sigma_{A_i}\right)^2 + \left(\frac{M_i A_i \sigma_V}{V}\right)^2} \tag{2}$$

where σ_x is the uncertainty of variable x. All other variables have been defined above.

The weighted result and unweighted result plots are located at the top middle and top right corner of the *SDAT* window. The plots are calculated as shown in equation 3.

$$\left(\sum_i C_i [lib]_i\right) \cdot [\xi], \tag{3}$$

where C_i are the concentration coefficients for radioxenon isotope i, $[lib]_i$ are the library files for radioxenon isotope i, and $[\xi]$ is the weighting matrix for the weighted result plot and the identity matrix for

the unweighted result plot. These plots may be compared with one another and the original sample plot to see how well the SDAT calculated results model the sample data.

The Weighted Residual and Unweighted Residual Plots are located directly below the Weighted Result and Unweighted Result Plots. The plots are calculated shown in equation 4.

$$\left| [Sample] - \left(\left(\sum_i C_i [lib]_i \right) \cdot [\xi] \right) \right|, \qquad (4)$$

where [*Sample*] is the original sample data and all other symbols have been previously defined. These plots show the differences between the original sample data and the calculated models of the sample as shown in the weighted result and unweighted result plots. SDAT also allows for sample re-analysis may with different input data. Results may also be saved.

Over time, the energy vs. channel calibration of both beta and gamma detectors can change. If the changes are significant, the SDAT program will return large errors because the sample signal will no longer match those in the library files. In such cases, the sample spectra should be rescaled to match the energy calibration properties of the original library files. Alternatively, the library files can be rescaled to match the sample calibration properties and saved. This program can also be used to compress or expand the data as desired. Figure 3 shows the SDAT rescaling window.

Step 2: Creation of Xe Library Files via MCNP

Library files were created for ^{131m}Xe, ^{133m}Xe, ^{133}Xe, and ^{135}Xe via a combination of MCNPX and MATLAB post processing. This is the same method as reported in Haas et al. (2007). The library files were created for each of the four cells in the β−γ coincidence detector system utilized in the ARSA system. The library files were then adjusted to match the energy calibration of the samples collected in the field. Figure 4 shows a comparison between the synthetically generated ^{133}Xe library file and a sample with ^{133}Xe collected by the ARSA in Guangzhou, China.

Step 3: Comparison of SDAT with CORIANT Results

As an initial quality control check, five spectra were analyzed wit SDAT. These samples collected with an ARSA unit in Guangzhou, China were initially analyzed by CORIANT at the Center for Monitoring Research in Arlington, VA. Figure 5 illustrates the comparison that was made between the SDAT results and the CORIANT results. The ^{133}Xe results compare very well. Since these results are from samples collected in the field, the correct quantity of ^{133}Xe in the sample is not known. This just serves as a check to make sure that the SDAT tool produces acceptable results for samples with strong Xe signals.

The analysis of ^{131m}Xe, ^{133m}Xe, and ^{135}Xe were also performed. These results did not compare as well as the ^{133}Xe results shown in figure 5. This issues with the ^{131m}Xe and ^{133m}Xe was primarily a detection issue. CORIANT reported ^{131m}Xe and ^{133m}Xe present in all of the samples analyzed. However, SDAT calculated ^{131m}Xe to be present in four out of the five samples and did not calculate ^{133m}Xe to be present in any of the samples. Discrepancies in these results are largely attributed to having correct β channel calibrations, which are questionable for both the CORIANT and the SDAT analysis. Visual inspection of the data does not show any clear ^{131m}Xe or ^{133m}Xe signals.

Another issue raised by this comparison is the error calculation performed in SDAT. The error bars produced for this comparison appear to be smaller than expected. While the error calculations may be correct, efforts should be made to review the error calculation in SDAT.

CONCLUSIONS AND RECOMMENDATIONS

The first version of the SDAT code was completed. Samples were analyzed with the code using MCNPX/MATLAB generated library files. Results from this comparison agreed with CORIANT results from the Center for Monitoring Research. Future work for the SDAT code will include an incorporation of activity calculations as well as an ability to read SPHD file headers.

The highest priority work will entail the creation of radioxenon isotopes via The University of Texas at Austin TRIGA reactor. These spectra will be important to use as library files for SDAT. They will also allow for advanced testing of SDAT with low counting statistics samples. Since the xenon concentrations may be experimentally controlled, the accuracy and precision of SDAT may be determined.

REFERENCES

K.M. Foltz Biegalski (2001). *J. Radioanalytical Nucl. Chem.* 248: (3), pp. 663–672.

D. A. Haas, S. R. Biegalski, and K. M. Foltz Biegalski (2007) Modeling β-γ coincidence spectra of ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe Signals, *Journal of Radioanalytical and Nuclear Chemistry* (in press).
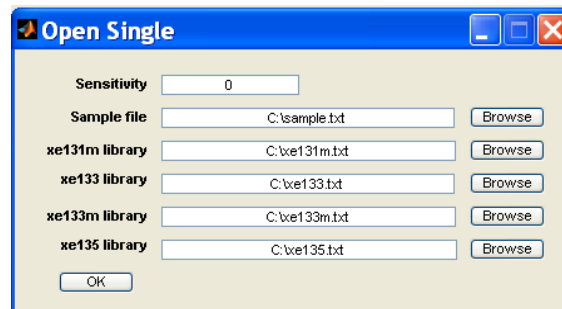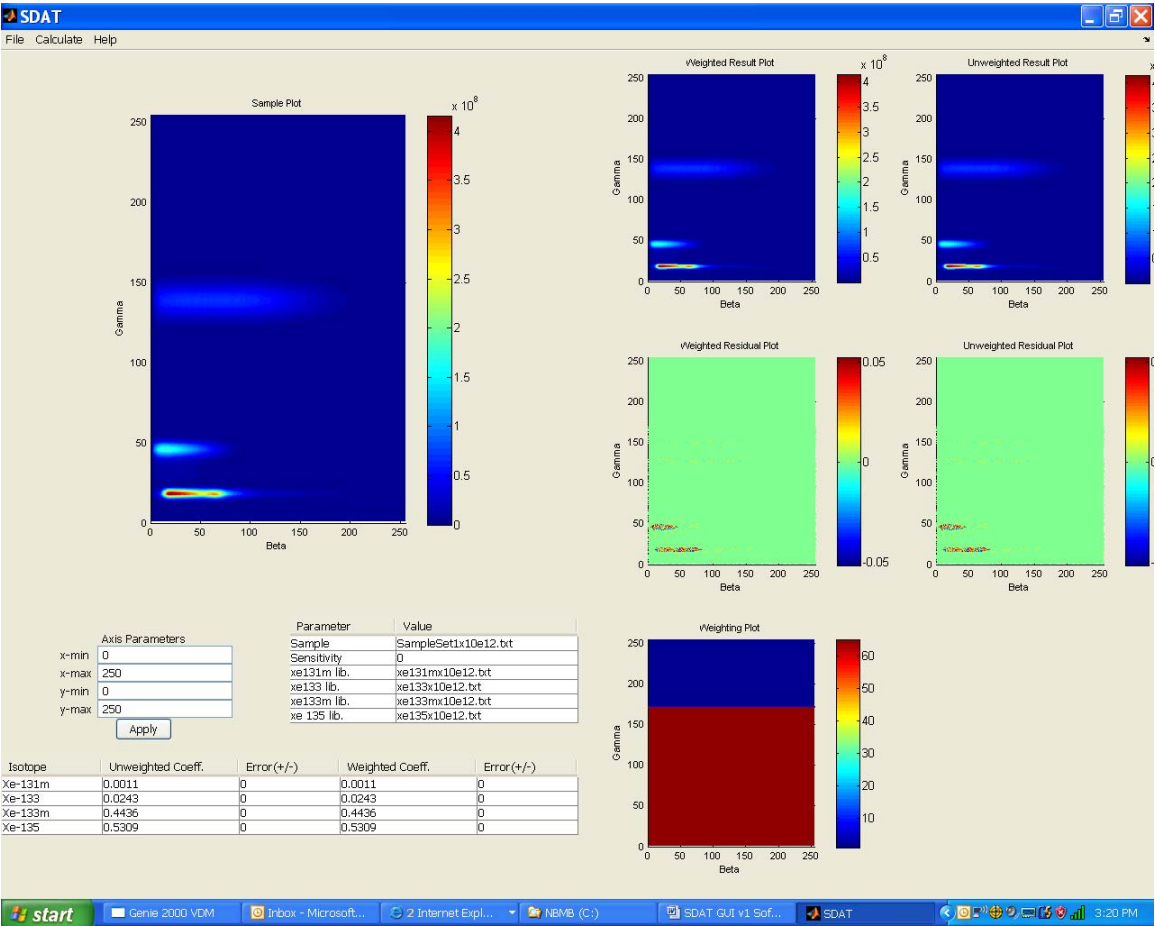
Figure 1. SDAT GUI open single window.

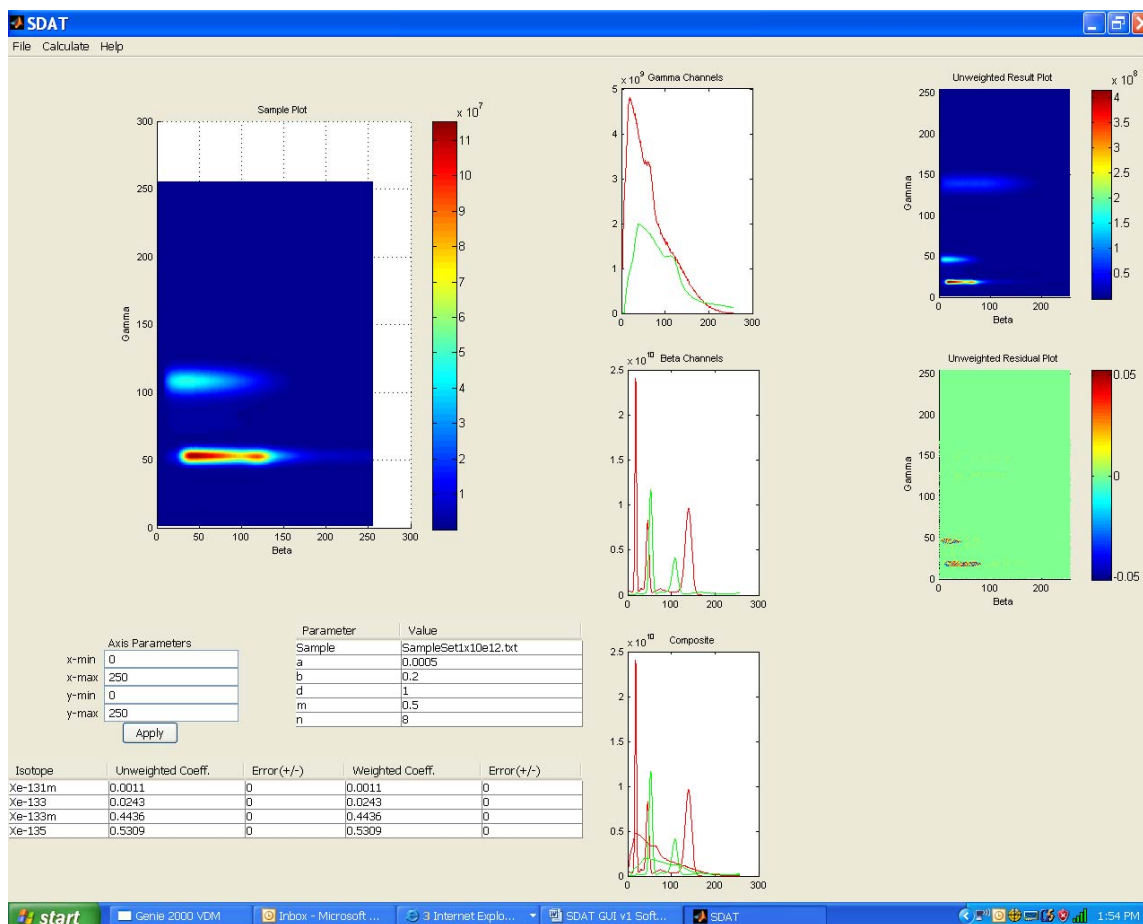**Figure 2. SDAT GUI sample analysis results display.**
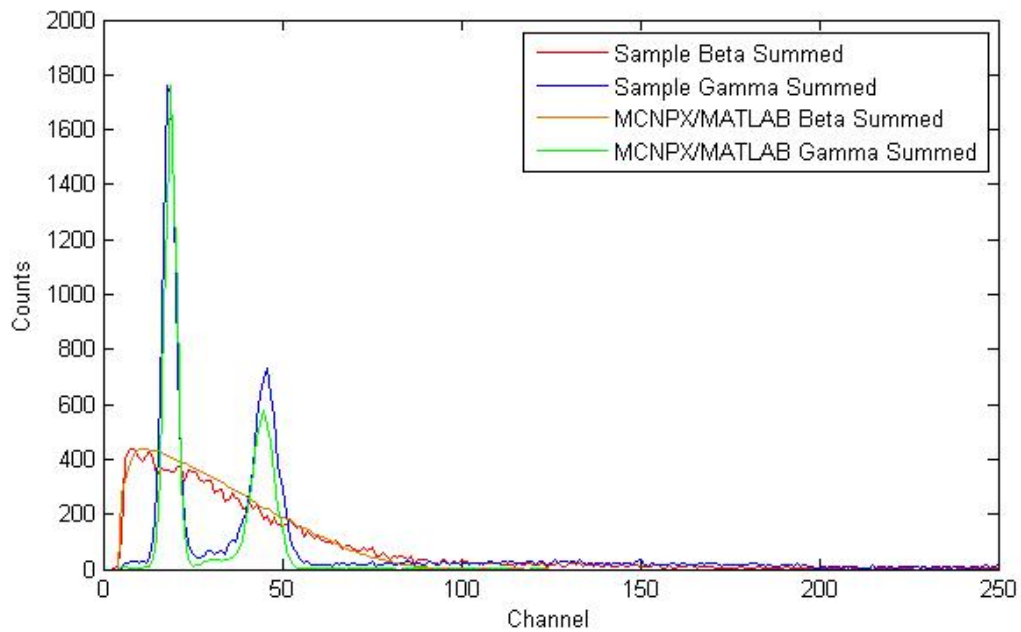
Figure 3. Display after rescaling a sample.

Figure 4. Comparison of β and γ compressed spectra from MCNPX/MATLAB generated ^{133}Xe library file and from sample collected in Guangzhou, China.
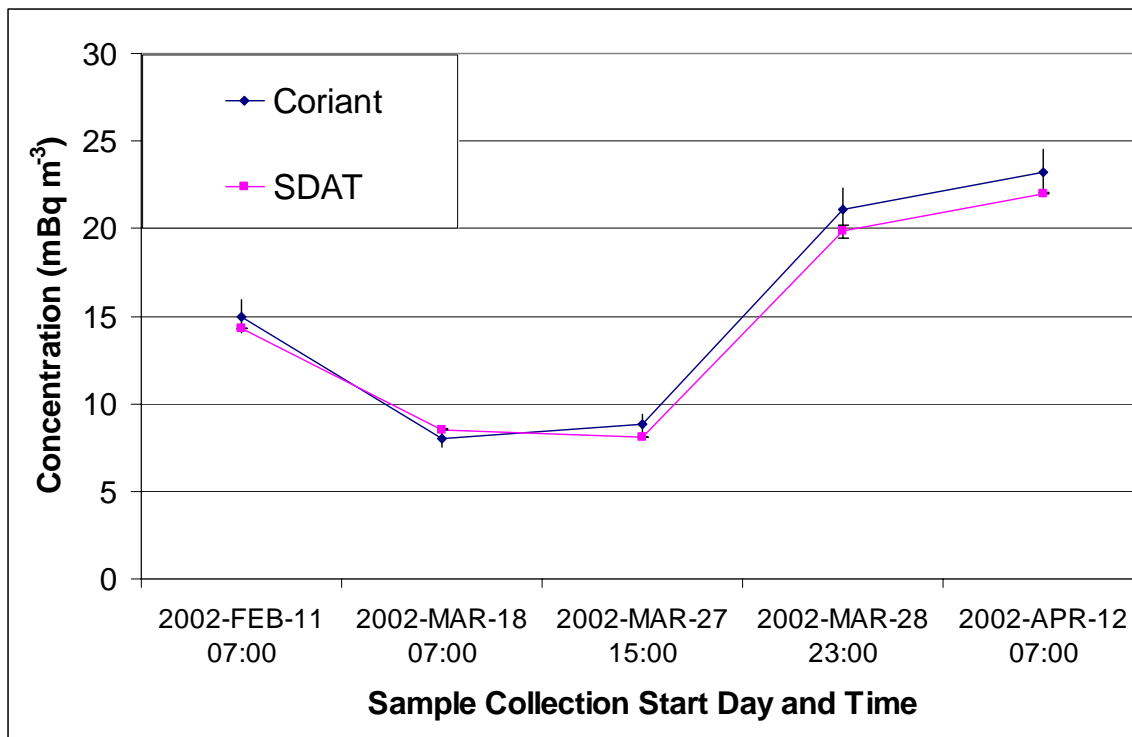
Figure 5. **Comparison of results between SDAT and CORIANT for five samples from the ARSA in Guangzhou, China.**