

DECONVOLUTION OF THREE-DIMENSIONAL BETA-GAMMA COINCIDENCE SPECTRA FROM XENON SAMPLING AND MEASUREMENT UNITS

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Sponsored by the Army Space and Missile Defense Command

Contract No. DTRA01-99-C-0031

ABSTRACT

Because radionuclides may be all that is released from nuclear weapons tests performed deep underground or underwater, environmental xenon sampling and measurement units are used in nuclear weapons test monitoring networks. Some environmental xenon sampling and measurement units, like the Automated Radionuclide 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 axes. Signals from four radionuclides (^{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. Deconvolving the 3-D sample spectra into the most probable combination of signals using non-negative least-squares results in a better ability to resolve spectral interferences. In addition, this method utilizes the entire signal from each radionuclide and consequently improves the signal to noise ratio. This paper describes the multiple isotope component analysis (MICA) algorithm developed for analysis of 3-D β - γ spectra from xenon sampling and measurement units, which comprises activity concentration and MDC calculations as well as nuclide identification methodology.

OBJECTIVE

There are two acceptable noble gas monitoring measurement modes for Comprehensive Nuclear-Test-Ban Treaty (CTBT) verification purposes as defined in CTBT/WGB/TL-11/5/Rev.8 (2000). These are 1) beta-gamma (β - γ) coincidence counting and 2) high-resolution γ -spectrometry. Two out of the three β - γ coincidence systems designed for this purpose acquire energy dispersive data on both the electron energy and photon energy axes: these systems are the ARSA (Bowyer et al., 1998) developed by PNNL and produced by EG&G Ortec, and the SAUNA (Weiss et al., 2000) developed by FOI in Stockholm, Sweden. The current software used by the CTBT organization in Vienna, Austria and the United States (US) National Data Center in Satellite Beach, Florida to analyze such data is *rms_xanalyze* (Dompierre et al., 2001; Donohoe et al., 2001). This algorithm was developed by the US Defense Threat Reduction Agency through a contract with Veridian. It is based on a Region of Interest (ROI) approach not dissimilar from most high-resolution gamma spectroscopy algorithms, but applied in two dimensions (Biegalski and Magyar, 2001). 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 $E_{\gamma} = 30$ 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 begun on a new software algorithm that takes into account all known physics and reduces the uncertainties involved in the results. This new software is called the Multiple Isotope Comparison Analysis (MICA) algorithm.

RESEARCH ACCOMPLISHED

MICA 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 Monte Carlo N-Particle Extended (MCNPX) code (LA-CP-02-408 and LA-CP-02-2607, 2002). 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 (Meng and Ramsden, 2000; Guttormsen et al., 1996; Prettyman et al., 1995). What makes MICA unique is that this methodology is applied to 3-D β - γ coincidence data.

Required Input Data

The format of the data sent by β - γ coincidence detectors in the International Monitoring System network is described in IDC3.4.1rev3, "Formats and Protocols for Messages" (Salzberg et al., 2001). 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. Therefore, we need the following detector response matrices with their associated activities:

$$\left[\begin{matrix} ^{131m}\text{Xe} \\ 55 \times 255 \end{matrix} \right], \left[\begin{matrix} ^{133m}\text{Xe} \\ 55 \times 255 \end{matrix} \right], \left[\begin{matrix} ^{133}\text{Xe} \\ 55 \times 255 \end{matrix} \right], \left[\begin{matrix} ^{135}\text{Xe} \\ 55 \times 255 \end{matrix} \right], \left[\begin{matrix} ^{214}\text{Pb} \\ 55 \times 255 \end{matrix} \right], \text{ and } \left[\begin{matrix} \text{DET BKG} \\ 55 \times 255 \end{matrix} \right].$$

These histograms should have the same size and calibration characteristics as the sample histogram in addition to good counting statistics; however, MICA is designed to be able to handle changes in the β or β -ECR. Changes in the RER or EER require new detector response histograms to be acquired. Unique weight matrices can be used with each, but these should be designed a priori, be the same size as the detector response matrices, and not change over time.

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. Weight matrices are also detector-specific since the calibration can differ between detectors. Calibration information should be supplied in the beginning part of the PHD message. With all of this information at hand, and knowing in which detector cell the sample was counted, the MICA algorithm can be applied to a sample for determining the atmospheric radioxenon concentrations.

MICA Concentration Calculation

First, it must be checked that the sample histogram has the same energy vs. channel calibration characteristics on both the \square and \square axes as the detector response histograms. If this is not the case, the sample histogram must be mapped into the energy vs. channel space of the detector response histograms. The details of this operation will not be described in this paper due to space considerations.

As mentioned previously, we then have the following matrices as input to the deconvolution problem:

$$\begin{bmatrix} {}^{131m}\text{Xe} \end{bmatrix}_{55 \times 255}, \begin{bmatrix} {}^{133m}\text{Xe} \end{bmatrix}_{55 \times 255}, \begin{bmatrix} {}^{133}\text{Xe} \end{bmatrix}_{55 \times 255}, \begin{bmatrix} {}^{135}\text{Xe} \end{bmatrix}_{55 \times 255}, \begin{bmatrix} {}^{214}\text{Pb} \end{bmatrix}_{55 \times 255}, \text{ and } \begin{bmatrix} \text{DET BKG} \end{bmatrix}_{55 \times 255}.$$

These all have the same calibration characteristics and size. These matrices can be weighted so that noise is filtered out. This requires the following predefined detector-specific weight matrices:

$$\begin{bmatrix} W_{131m} \end{bmatrix}_{255 \times 255}, \begin{bmatrix} W_{133m} \end{bmatrix}_{255 \times 255}, \begin{bmatrix} W_{133} \end{bmatrix}_{255 \times 255}, \begin{bmatrix} W_{135} \end{bmatrix}_{255 \times 255}, \begin{bmatrix} W_{214} \end{bmatrix}_{255 \times 255}, \begin{bmatrix} W_{\square} \end{bmatrix}_{255 \times 255}.$$

Elements in the weight matrices are binary, i.e., either 0 or 1. The last weight matrix listed, $\begin{bmatrix} W_{\square} \end{bmatrix}_{255 \times 255}$, is used for weighting both the SAMPLEPHD and the DETBKPHD histograms. $\begin{bmatrix} W_{\square} \end{bmatrix}_{255 \times 255}$ is determined by combining the isotopic weight matrices listed above. Essentially, if element i,j in any of the isotopic weight matrices is 1, then this element in $\begin{bmatrix} W_{\square} \end{bmatrix}_{255 \times 255}$ is assigned 1. Otherwise, it is zero.

To weight the input matrices, they are array-multiplied (\cdot) with the appropriate weight matrix. Array multiplication entails multiplying two matrices element by element and is different than matrix multiplication (i.e., $\begin{bmatrix} A \end{bmatrix} \times \begin{bmatrix} B \end{bmatrix}$):

$$\begin{aligned} \begin{bmatrix} {}^{131m}\text{Xe} \end{bmatrix}_{55 \times 255} \cdot \begin{bmatrix} W_{131m} \end{bmatrix}_{255 \times 255} &= \begin{bmatrix} {}^{131m}\text{Xe} \end{bmatrix}_{55 \times 255}, \\ \begin{bmatrix} {}^{133m}\text{Xe} \end{bmatrix}_{55 \times 255} \cdot \begin{bmatrix} W_{133m} \end{bmatrix}_{255 \times 255} &= \begin{bmatrix} {}^{133m}\text{Xe} \end{bmatrix}_{55 \times 255}, \\ \begin{bmatrix} {}^{135}\text{Xe} \end{bmatrix}_{55 \times 255} \cdot \begin{bmatrix} W_{135} \end{bmatrix}_{255 \times 255} &= \begin{bmatrix} {}^{135}\text{Xe} \end{bmatrix}_{55 \times 255}, \\ \begin{bmatrix} {}^{214}\text{Pb} \end{bmatrix}_{55 \times 255} \cdot \begin{bmatrix} W_{214} \end{bmatrix}_{255 \times 255} &= \begin{bmatrix} {}^{214}\text{Pb} \end{bmatrix}_{55 \times 255}, \\ \begin{bmatrix} \text{DET BKG} \end{bmatrix}_{55 \times 255} \cdot \begin{bmatrix} W_{\square} \end{bmatrix}_{255 \times 255} &= \begin{bmatrix} \text{DET BKG} \end{bmatrix}_{55 \times 255}, \\ \begin{bmatrix} \text{SAMPLE} \end{bmatrix}_{55 \times 255} \cdot \begin{bmatrix} W_{\square} \end{bmatrix}_{255 \times 255} &= \begin{bmatrix} \text{SAMPLE} \end{bmatrix}_{55 \times 255}. \end{aligned}$$

The weighted detector response histograms and sample histogram are then vectorized, i.e., the 255×255 matrices are converted into column vectors of dimension 65025×1 . This is done by appending each histogram row onto one another, and then transposing the resulting row matrix, i.e.,

$$\begin{aligned} \begin{bmatrix} {}^{131m}\text{Xe} \end{bmatrix}_{55 \times 255} \square \begin{bmatrix} {}^{131m}\text{Xe} \end{bmatrix}_{5025 \times 1}, \\ \begin{bmatrix} {}^{133m}\text{Xe} \end{bmatrix}_{55 \times 255} \square \begin{bmatrix} {}^{133m}\text{Xe} \end{bmatrix}_{5025 \times 1}, \\ \begin{bmatrix} {}^{133}\text{Xe} \end{bmatrix}_{55 \times 255} \square \begin{bmatrix} {}^{133}\text{Xe} \end{bmatrix}_{5025 \times 1}, \\ \begin{bmatrix} {}^{135}\text{Xe} \end{bmatrix}_{55 \times 255} \square \begin{bmatrix} {}^{135}\text{Xe} \end{bmatrix}_{5025 \times 1}, \\ \begin{bmatrix} {}^{214}\text{Pb} \end{bmatrix}_{55 \times 255} \square \begin{bmatrix} {}^{214}\text{Pb} \end{bmatrix}_{5025 \times 1}, \\ \begin{bmatrix} \text{DET BKG} \end{bmatrix}_{55 \times 255} \square \begin{bmatrix} \text{DET BKG} \end{bmatrix}_{5025 \times 1}, \\ \begin{bmatrix} \text{SAMPLE} \end{bmatrix}_{55 \times 255} \square \begin{bmatrix} \text{SAMPLE} \end{bmatrix}_{5025 \times 1}. \end{aligned}$$

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

$$\begin{bmatrix} \begin{bmatrix} {}^{131m}\text{Xe} \end{bmatrix}_{5025 \times 1} \begin{bmatrix} {}^{133m}\text{Xe} \end{bmatrix}_{5025 \times 1} \begin{bmatrix} {}^{133}\text{Xe} \end{bmatrix}_{5025 \times 1} \begin{bmatrix} {}^{135}\text{Xe} \end{bmatrix}_{5025 \times 1} \begin{bmatrix} {}^{214}\text{Pb} \end{bmatrix}_{5025 \times 1} \begin{bmatrix} \text{DET BKG} \end{bmatrix}_{5025 \times 1} \end{bmatrix}_{65025 \times 6}$$

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

$$[Response]_{6 \times 5025 \times 6} = \begin{bmatrix} X & X & X & X & P & D \\ e & e & e & e & b & E \\ 1 & 1 & 1 & 1 & 2 & T \\ 3 & 3 & 3 & 3 & 1 & B \\ 1 & 3 & 3 & 5 & 4 & K \\ m & m & & & & G \end{bmatrix}_{6 \times 5025 \times 6}$$

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

$$[Response]_{6 \times 5025 \times 6} [Coefficients]_{6 \times 1} = [Sample]_{6 \times 5025 \times 1}$$

The coefficient matrix holds the multipliers, λ , 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 is used to solve for $[Coefficients]_{6 \times 1}$ in this problem. This method was chosen over the standard least squares solution since all the components of the $[Coefficients]_{6 \times 1}$ matrix should be greater than or equal to zero.

Once the response coefficients, λ , are known, the activity concentration of the radionuclides without parents (^{131m}Xe , ^{133m}Xe , and ^{135}Xe) may be calculated as:

$$Q_{Sample_i} = \frac{A_{std} \lambda_{Sample_i} \left(\frac{(1 - e^{-\lambda_i t_{a_{std}}})}{(1 - e^{-\lambda_i t_{a_{Sample}}})} \right) \lambda_{GB_i} \left(\frac{(1 - e^{-\lambda_i t_{a_{std}}})}{(1 - e^{-\lambda_i t_{a_{GB}}})} \right) e^{-\lambda_i t_{d_{(GB \setminus Sample)}}}}{V_{Xe} \left(e^{-\lambda_i t_{d_{Sample}}} \right) \left(1 - e^{-\lambda_i t_{c_{Sample}}} \right)}$$

where Q_{Sample_i} is the activity concentration of radioisotope i in the sample, A_{std_i} is the activity of radioisotope i in the calibration standard used to obtain the detector response matrix for that radioisotope, λ is the concentration of elemental Xe in air (i.e., $0.087 \text{ cm}^3/\text{m}^3$), V_{Xe} is the sample volume of elemental Xe in cm^3 , λ_{Sample_i} is the response coefficient of radioisotope i in the sample, λ_{GB_i} is the response coefficient of radioisotope i in the gas background, λ_i is the decay constant of radioisotope i , $t_{a_{std}}$ is the acquisition time of the detector response matrix, $t_{a_{Sample}}$ is the acquisition time of the gas background histogram, $t_{d_{(GB \setminus Sample)}}$ is the difference in time between the start of the gas background histogram acquisition and the start of the sample histogram acquisition, $t_{d_{Sample}}$ is the decay time between the end of the sample collection and the start of the sample histogram acquisition, and $t_{c_{Sample}}$ is the collection time of the sample histogram.

Assuming that the uncertainties of the decay constants and the times are negligible, the uncertainty of the activity concentration for the radionuclides without parents (^{131m}Xe , ^{133m}Xe , and ^{135}Xe) may be calculated as:

$$\sigma_{Q_{Sample_i}} = Q_{Sample_i} \sqrt{\left(\frac{\lambda_{std_i}}{A_{std_i}} \right)^2 + \left(\frac{\lambda_{Xe}}{V_{Xe}} \right)^2 + \left(\frac{\lambda_{Sample_i}}{\lambda_{Sample_i}} \right)^2 + \left(\frac{\lambda_{Sample_i} \left(\frac{(1 - e^{-\lambda_i t_{a_{std}}})}{(1 - e^{-\lambda_i t_{a_{Sample}}})} \right) \lambda_{GB_i} \left(\frac{(1 - e^{-\lambda_i t_{a_{std}}})}{(1 - e^{-\lambda_i t_{a_{GB}}})} \right) e^{-\lambda_i t_{d_{(GB \setminus Sample)}}}}{\lambda_{Sample_i} \left(\frac{(1 - e^{-\lambda_i t_{a_{std}}})}{(1 - e^{-\lambda_i t_{a_{Sample}}})} \right) \lambda_{GB_i} \left(\frac{(1 - e^{-\lambda_i t_{a_{std}}})}{(1 - e^{-\lambda_i t_{a_{GB}}})} \right) e^{-\lambda_i t_{d_{(GB \setminus Sample)}}}} \right)^2}$$

^{133}Xe poses a particular complication due to the possibility of a parent-daughter relationship existing in the sample. In cases where ^{133m}Xe is present, our sample histogram indicates ^{133}Xe decays representing ^{133m}Xe that was collected in the atmosphere. Therefore, for ^{133}Xe the following must be utilized to calculate the atmospheric aerosol concentration:

$$Q_{Xe133}^{Sample} = \frac{A \square C \square D \square E}{\frac{F}{\square_{Xe133}^2} \left(\square e^{\square_{Xe133} t_c} \right) \left(e^{\square_{Xe133} t_d} \right) \left(\square e^{\square_{Xe133} t_a} \right)}$$

where

$$A = \frac{A_{oXe133}^{std}}{\square_{Xe133}} \left(\square e^{\square_{Xe133} t_a^{std}} \right) \square_{Xe133}^{sample}$$

$$C = \frac{\square_{Xe133}^{GB} A_{oXe133}^{GB} e^{\square_{Xe133} t_a^{GB}}}{\square_{Xe133}} + \frac{A_{oXe133}^{GB} \square_{Xe133}}{\square_{Xe133m} \square_{Xe133}} \left[\frac{e^{\square_{Xe133m} t_d^{GBS}}}{\square_{Xe133m}} \left(\square e^{\square_{Xe133m} t_a^{GB}} \right) \right] \frac{e^{\square_{Xe133} t_d^{GBS}}}{\square_{Xe133}} \left(\square e^{\square_{Xe133} t_a^{GB}} \right) \square_{Xe133}^{GB}$$

$$A_{oXe133}^{GB} = \frac{A_{oXe133}^{std} \left(\square e^{\square_{Xe133} t_a^{std}} \right) \square_{Xe133}^{GB} \square_{Xe133} A_{oXe133m}^{GB} \frac{\square_{Xe133}}{\square_{Xe133m} \square_{Xe133}} \left(e^{\square_{Xe133m} t_d^{GB}} \square e^{\square_{Xe133} t_a^{GB}} \right)}{\left(\square e^{\square_{Xe133} t_a^{GB}} \right)}$$

$$A_{oXe133m}^{GB} = \frac{A_{oXe133m}^{std} \left(\square e^{\square_{Xe133m} t_a^{std}} \right)}{\left(\square e^{\square_{Xe133m} t_a^{GB}} \right) \square_{Xe133m}^{GB}}$$

$$D = \frac{Q_{Xe133m} F}{\square_{Xe133m} \square_{Xe133}} \left[\frac{\square_{Xe133m}}{\square_{Xe133m}} \left(\square e^{\square_{Xe133m} t_c} \right) \square_{Xe133} \left(\square e^{\square_{Xe133m} t_c} \right) \right] \left(e^{\square_{Xe133} t_d} \right) \left(\square e^{\square_{Xe133} t_a} \right)$$

$$E = \frac{\square_{Xe133} Q_{Xe133m} F}{\square_{Xe133m} \left(\square_{Xe133} \square_{Xe133m} \right)} \left(\square e^{\square_{Xe133m} t_c} \right) \left[\frac{e^{\square_{Xe133m} t_d}}{\square_{Xe133m}} \left(\square e^{\square_{Xe133m} t_a} \right) \right] \frac{e^{\square_{Xe133} t_d}}{\square_{Xe133}} \left(\square e^{\square_{Xe133} t_a} \right) \square_{Xe133}$$

$$F = \frac{\square}{V_{Xe}}, Q_{Xe133}^{Sample} \text{ is the concentration of } ^{133}\text{Xe} \text{ in the sampled air, } Q_{Xe133m} \text{ is the concentration of } ^{133m}\text{Xe} \text{ in the}$$

sampled air, A_{oXe133}^{std} is the activity of ^{133}Xe in the standard used to create the detector response matrix, $A_{oXe133m}^{std}$ is the activity of ^{133m}Xe in the standard used to create the detector response matrix, A_{oXe133}^{GB} is the activity of ^{133}Xe in the gas background at the start of the gas background histogram acquisition, $A_{oXe133m}^{GB}$ is the activity of ^{133m}Xe in the gas background at the start of the gas background histogram acquisition, \square_{Xe133}^{sample} is the detector response coefficient for ^{133}Xe in the sample histogram, \square_{Xe133}^{GB} is the detector response coefficient for ^{133}Xe in the gas background histogram, \square_{Xe133m}^{GB} is the detector response coefficient for ^{133m}Xe in the gas background histogram, \square_{Xe133} is the decay constant for ^{133}Xe , \square_{Xe133m} is the decay constant for ^{133m}Xe , t_a is the acquisition time of the sample histogram, t_a^{std} is the acquisition time of the detector response matrix, t_a^{GB} is the acquisition time of the gas background histogram, t_d is the decay time between sample collection and the start of the sample histogram acquisition, t_d^{GBS} is the decay time between the start of the sample background acquisition and the start of the sample acquisition, and t_c is the sample collection time. All other variables have been previously defined. The uncertainty of the ^{133}Xe atmospheric concentration is also very complicated and is not addressed in this paper due to space considerations.

Detection Limit Calculation

To determine an MDC, the nuclide-specific detection limit (L_D) must be calculated. This section includes the derivation of L_D for each radioxenon of interest according to the MICA algorithm requirements. The MDC calculation is based on the L_D derivation as described in Foltz Biegalski and Biegalski (2001). The nuclide-specific L_D is utilized to calculate an MDC as shown below.

$$MDC = \frac{L_D}{\frac{\epsilon_{\gamma} \cdot \epsilon_{\beta} \cdot I_{\gamma} \cdot I_{\beta} \cdot F}{\lambda} \left(1 - e^{-\lambda t_c}\right) \left(1 - e^{-\lambda t_d}\right) \left(1 - e^{-\lambda t_r}\right)}$$

where ϵ_{γ} is the energy-dependent detection system photon efficiency, ϵ_{β} is the energy-dependent detection system electron efficiency, I_{γ} is the photon intensity, I_{β} is the intensity of the β particle or conversion electron in coincidence with the photon, F is the sampling flow rate, t_c is the sample collection time, t_d is the sample decay time, t_r is the sample acquisition real time, and λ is the nuclide decay constant.

The L_D is the true net signal level that may be *a priori* expected to lead to detection (Currie, 1968). The detection limit is considered the minimum level at which a signal can be reliably quantified. It is a function of the critical level (L_C): the level at which a net signal may not be reliably detected. Both the L_D and L_C may be calculated for nuclides not found in a spectrum, as well as for those found (Canberra, 1995). The detection limit is defined as

$$L_D = L_C + k \sigma_D$$

where $L_C = k \sigma_o$, k_{β} and k_{γ} are abscissas of the Normal distribution, σ_D is the standard deviation of the net signal when its real value is equal to L_D , and σ_o is the standard deviation of the observed net signal. Nominally, the acceptable probabilities for errors of the first and second kind are set equal, i.e., $\alpha = \beta$ and $k_{\beta} = k_{\gamma} = k$. For a 95% confidence level, $\alpha = \beta = 0.05$ and $k = 1.645$ (Currie, 1968).

In the case of β - γ coincidence detectors, the observed gross signal (G) is a function of the counts from the nuclide of interest (S), the counts from interference radionuclides (I), and the counts from nuclides adsorbed onto the walls of the detection chamber from the previous sample (M). There is no interference from Compton for this analytical method (i.e., MICA) since the Compton counts are part of the nuclide signal. In addition, there are no radioxenon isotopes in the detector background signal that need to be counted as interferences. The observed net signal is then

$$S = G - I - M$$

and the signal variance is

$$\sigma_S^2 = \sigma_G^2 + \sigma_I^2 + \sigma_M^2$$

where σ_G^2 , σ_I^2 , and σ_M^2 are the variances of G , I , and M , respectively. It is assumed that background interferences are included in M .

Because the gross signal may be assumed Poisson distributed (Currie, 1968), the variance of the gross signal can be approximated by the "true" mean gross signal, μ_G . Therefore,

$$\sigma_S^2 = \mu_G + \sigma_I^2 + \sigma_M^2$$

or

$$\sigma_S^2 = \mu_S + \sigma_I + \mu_M + \sigma_I^2 + \sigma_M^2$$

where μ_S , μ_I , μ_M , and μ_B are the "true" mean net counts for S , I , and M , respectively. If the net signal is null (i.e., $\mu_S = 0$), then the variance of the net signal becomes

$$\sigma_o^2 = \mu_I + \mu_M + \sigma_I^2 + \sigma_M^2$$

When the true net signal is equal to the detection limit (i.e., $\mu_S = L_D$), then the net signal variance becomes

$$\sigma_D^2 = L_D + \mu_I + \mu_M + \sigma_I^2 + \sigma_M^2.$$

Inserting the definitions of L_D and L_C and setting $k_{\beta} = k_{\gamma} = k$ yields

$$L_D = L_C + k \sqrt{L_D + \mu_I + \mu_M + \sigma_I^2 + \sigma_M^2}$$

and

$$L_C = k \sigma_o = k \sqrt{\mu_I + \mu_M + \sigma_I^2 + \sigma_M^2}.$$

Solving for L_D yields

$$L_D = k^2 + 2L_C.$$

Xenon-133

In a β - γ coincidence detector, the primary ^{133}Xe signal is the β -line at 81 keV in coincidence with a γ particle of maximum energy equal to 346.4 keV. Interference with this signal occurs from radon daughters, primarily ^{214}Pb . The ^{133}Xe signal in the 30 keV X-ray region (from coincidence with beta particles) has interference from $^{131\text{m}}\text{Xe}$,

^{133m}Xe , and a small amount from ^{135}Xe . The detection limit for ^{133}Xe is calculated by first determining the critical level, L_C :

$$L_C^{Xe-133} = k\sqrt{\sigma_I^{Xe\text{[133]}^2} + \sigma_M^{Xe\text{[133]}^2} + (\sigma_I^{Xe\text{[133]}})^2 + (\sigma_M^{Xe\text{[133]}})^2}$$

The ^{133}Xe interference term is simply the difference between the gross number of ^{133}Xe counts in the sample histogram and the number of counts attributed to ^{133}Xe by the spectral deconvolution fit of the sample, or

$$\sigma_I^{Xe\text{[133]}} = \sigma_G^{Xe\text{[133]}} - \sigma_{G\text{ fit}}^{Xe\text{[133]}}$$

where $\sigma_G^{Xe\text{[133]}}$ is the gross counts in the sample spectrum in the channels that have counts attributed to ^{133}Xe and $\sigma_{G\text{ fit}}^{Xe\text{[133]}}$ is the gross counts attributed to ^{133}Xe by the spectral deconvolution fit. Following from the Poisson distribution assumption, the following equality can be assumed:

$$(\sigma_I^{Xe\text{[133]}})^2 = \sigma_I^{Xe\text{[133]}}$$

The memory effect, M , is caused by the adsorption of a small fraction of atoms from the previous sample on the detector cell wall. This fraction depends on the amount of atoms in the previous sample as well as the chemical characteristics of the element. For the Automated Radioxenon Sampler-Analyzer (ARSA), the residual count rate from the memory effect can range up to 10% of the previous sample count rate (Reeder, Bowyer, Abel, 1997). The expressions for the signal and variance due to the memory effect are derived as

$$\sigma_M^{Xe133} = \sigma_{Xe133} \sigma_{Xe133} \left[\frac{A_o^{Xe133} e^{-\lambda_{Xe133} t_d}}{\sigma_{Xe133}} \left(1 - e^{-\lambda_{Xe133} t_d} \right) + \frac{A_o^{Xe133m} \sigma_{Xe133}}{\sigma_{Xe133m} \sigma_{Xe133}} e^{-\lambda_{Xe133m} t_d} \left(1 - e^{-\lambda_{Xe133m} t_d} \right) \right] e^{-\lambda_{Xe133} t_d} \left(1 - e^{-\lambda_{Xe133} t_d} \right)$$

where $A_o^{Xe133} = \frac{\sigma_{GB}^{Xe133}}{\sigma_{Xe133} \sigma_{Xe133}} \frac{A_o^{Xe133m}}{\sigma_{Xe133m} \sigma_{Xe133}} \left(\frac{\sigma_{Xe133}}{\sigma_{Xe133m} \sigma_{Xe133}} \left(e^{-\lambda_{Xe133m} t_{GB}} - e^{-\lambda_{Xe133} t_{GB}} \right) \right)$, the ^{133}Xe activity at the start of the gas background count; $A_o^{Xe133m} = \frac{\sigma_{GB}^{Xe133m}}{\sigma_{Xe133} \sigma_{Xe133}} \left(1 - e^{-\lambda_{Xe133m} t_{GB}} \right)$, the ^{133m}Xe activity at the start of the GB count; σ_M^{Xe133} is the signal (gross counts) from ^{133}Xe in the sample count from the cell memory effect; σ_{GB}^{Xe133} is the signal (gross counts) from ^{133}Xe in the gas background count; σ_{GB}^{Xe133m} is the signal (gross counts) from ^{133m}Xe in the gas background count; σ_{Xe133} is the total detector efficiency for ^{133}Xe ; σ_{Xe133m} is the total detector efficiency for ^{133m}Xe ; σ_{Xe133} is the β - γ coincidence yield for ^{133}Xe ; and t_{GB} is the acquisition time for the gas background count. The gas background count occurs after the detector cell has been purged of the previous sample and before the next sample acquisition. All other parameters have been previously defined.

Xenon-135

A similar derivation can be performed for ^{135}Xe . This radioxenon has a primary β line at 249.8 keV in coincidence with a beta particle of $E_{\text{max}} = 908.2$ keV. Interference with this signal occurs primarily from ^{214}Pb . The detection limit for ^{135}Xe is calculated by first determining the critical level, L_C :

$$L_C^{Xe-135} = k\sqrt{\sigma_I^{Xe\text{[135]}^2} + \sigma_M^{Xe\text{[135]}^2} + (\sigma_I^{Xe\text{[135]}})^2 + (\sigma_M^{Xe\text{[135]}})^2}$$

Expression for the interference signal is:

$$\sigma_I^{Xe\text{[135]}} = \sigma_G^{Xe\text{[135]}} - \sigma_{G\text{ fit}}^{Xe\text{[135]}}$$

where \bar{N}_G^{Xe135} is the gross counts in the sample spectrum in the channels that have counts attributed to ^{135}Xe and $\bar{N}_{G\text{ fit}}^{Xe135}$ is the gross counts attributed to ^{135}Xe by the spectral deconvolution fit. As before, the variance of the interference signal is assumed equivalent to \bar{N}_I , i.e.,

$$\left(\bar{N}_I^{Xe135}\right)^2 = \bar{N}_I^{Xe135}.$$

Expressions for the signal and the variance from the memory effect are derived as

$$\bar{N}_M^{Xe135} = \frac{\left(\bar{N}_{GB\text{ fit}}^{Xe135}\right)}{\left(1 - e^{-\lambda_{Xe135} t_{GB}}\right)} \cdot e^{-\lambda_{Xe135} t_{GB}} \cdot \left(1 - e^{-\lambda_{Xe135} t_r}\right)$$

and

$$\left(\bar{N}_M^{Xe135}\right)^2 = \frac{\left[e^{-\lambda_{Xe135} t_{GB}} \cdot \left(1 - e^{-\lambda_{Xe135} t_r}\right)\right]^2}{\left(1 - e^{-\lambda_{Xe135} t_{GB}}\right)^2} \cdot \left(\bar{N}_{GB\text{ fit}}^{Xe135}\right)^2$$

where $\left(\bar{N}_{GB\text{ fit}}^{Xe135}\right)$ is the ^{135}Xe counts in the GB count and λ_{Xe-135} is the decay constant for ^{135}Xe . The other parameters have been previously defined.

Xenon-131m

The ^{131m}Xe signal is produced between the coincidence of one of several ~30 keV X-rays in coincidence with a conversion electron at 129.4 keV. Interference with this signature occurs from ^{133}Xe . The detection limit for ^{131m}Xe is calculated by first determining the critical level, L_C :

$$L_C^{Xe-131m} = k\sqrt{\bar{N}_I^{Xe131m} + \bar{N}_M^{Xe131m} + \left(\bar{N}_I^{Xe131m}\right)^2 + \left(\bar{N}_M^{Xe131m}\right)^2}.$$

Expressions for the interference signal and variance are similar to those of the other xenon radioisotopes. The expression for the interference signal is:

$$\bar{N}_I^{Xe131m} = \bar{N}_G^{Xe131m} \bar{N}_{G\text{ fit}}^{Xe131m}$$

where \bar{N}_G^{Xe131m} is the gross count in the sample spectrum in the channels that have counts attributed to ^{131m}Xe and $\bar{N}_{G\text{ fit}}^{Xe131m}$ is the gross counts attributed to ^{131m}Xe by the spectral deconvolution fit. Following from the Poisson distribution assumption, the following equality is assumed:

$$\left(\bar{N}_I^{Xe131m}\right)^2 = \bar{N}_I^{Xe131m}.$$

The signal and variance of the memory effect for ^{131m}Xe are derived as

$$\bar{N}_M^{Xe131m} = \frac{\left(\bar{N}_{GB\text{ fit}}^{Xe131m}\right)}{\left(1 - e^{-\lambda_{Xe131m} t_{GB}}\right)} \cdot e^{-\lambda_{Xe131m} t_{GB}} \cdot \left(1 - e^{-\lambda_{Xe131m} t_r}\right)$$

and

$$\left(\bar{N}_M^{Xe131m}\right)^2 = \frac{\left[e^{-\lambda_{Xe131m} t_{GB}} \cdot \left(1 - e^{-\lambda_{Xe131m} t_r}\right)\right]^2}{\left(1 - e^{-\lambda_{Xe131m} t_{GB}}\right)^2} \cdot \left(\bar{N}_{GB\text{ fit}}^{Xe131m}\right)^2$$

where $\left(\bar{N}_{GB\text{ fit}}^{Xe131m}\right)$ is the ^{131m}Xe counts calculated for the gas background count and $\lambda_{Xe-131m}$ is the decay constant for ^{131m}Xe .

Xenon-133m

^{133m}Xe is detected through one of several ~30 keV X-rays in coincidence with a conversion electron at 198 keV. Interference with this signature occurs from ^{133}Xe . The detection limit for ^{133m}Xe is calculated by first determining the critical level, L_C :

$$L_C^{Xe-133m} = k\sqrt{\bar{N}_I^{Xe133m} + \bar{N}_M^{Xe133m} + \left(\bar{N}_I^{Xe133m}\right)^2 + \left(\bar{N}_M^{Xe133m}\right)^2}.$$

The interference signal and variance are derived as

$$\bar{N}_I^{Xe133m} = \bar{N}_G^{Xe133m} \bar{N}_{G\text{ fit}}^{Xe133m}$$

where $\mu_G^{Xe-133m}$ is the gross counts in the sample spectrum in the channels that have counts attributed to ^{133m}Xe and $\mu_{G\text{ fit}}^{Xe-133m}$ is the gross counts attributed to ^{133m}Xe by the spectral deconvolution fit. Following from the Poisson distribution assumption, the following equality can be assumed:

$$\left(\mu_G^{Xe-133m}\right)^2 = \mu_G^{Xe-133m}.$$

The signal and variance of the memory effect for ^{133m}Xe are derived as

$$\mu_M^{Xe-131m} = \frac{\left(\mu_{GB\text{ fit}}^{Xe-133m}\right)}{\left(1 - e^{-\lambda_{Xe-131m} t_{GB}}\right)} \cdot e^{-\lambda_{Xe-131m} t_{GB}} \cdot \left(1 - e^{-\lambda_{Xe-131m} t_r}\right)$$

and

$$\left(\mu_M^{Xe-131m}\right)^2 = \frac{e^{-2\lambda_{Xe-131m} t_{GB}} \cdot \left(1 - e^{-\lambda_{Xe-131m} t_r}\right)^2}{\left(1 - e^{-\lambda_{Xe-131m} t_{GB}}\right)^2} \cdot \mu_{GB\text{ fit}}^{Xe-133m}$$

where $\left(\mu_{GB\text{ fit}}^{Xe-133m}\right)$ is the ^{133m}Xe counts calculated for the gas background count and $\lambda_{Xe-133m}$ is the decay constant for ^{133m}Xe .

Nuclide Identification

The counts attributed to each radionuclide are compared against L_c to determine if the radionuclide is present. The counts for each radionuclide are determined from:

$$Counts_i = \sum_{j=1}^n \sum_{k=1}^m \mu_i(j, k)$$

where $Counts_i$ is a scalar combined by adding up all the counts in the detector response matrix, μ_i is the detector response coefficient for radionuclide i , and $\mu_i(j, k)$ is the detector response matrix for radionuclide i . If $Counts_i$ is greater than L_c for radionuclide i , it is considered a positive detection and the radionuclide is identified as present in the sample.

CONCLUSIONS AND RECOMMENDATIONS

This method allows for an increase in the utilized signal size by incorporating the entire detector response in the analysis algorithm. This equates to improved detection limits. However, such improvements are dependent on other radionuclide signals present in the sample spectrum and the quality of the detector response histograms used in the analysis. From applying the MICA algorithm to simulated and real sample data using simulated detector response histograms, it was found that the algorithm works very well. However, some error was introduced into the real sample data analysis by non-ideal simulated detector response histograms.

The work incorporated in this paper demonstrates the feasibility of using the spectral deconvolution concept for analysis of 3-D coincidence data. The next phase of this work will be to improve the detector response histograms utilized in the analysis and to allow for energy calibration matching between the sample histogram and the detector response histograms. The detector response histograms will be improved by a combination of MCNPX modeling and direct radioxenon calibration measurements on a coincidence detector. The algorithm will then be tested with a large quantity of real sample data.

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