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A New Algorithm for Radioisotope Identification of Shielded and Masked SNM/RDD Materials

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ABSTRACT

Detection and identification of shielded and masked nuclear materials is crucial to national security, but vast borders and high volumes of traffic impose stringent requirements for practical detection systems. Such tools must be be mobile, and hence low power, provide a low false alarm rate, and be sufficiently robust to be operable by non-technical personnel. Currently fielded systems have not achieved all of these requirements simultaneously. Transport modeling such as that done in GADRAS is able to predict observed spectra to a high degree of fidelity; our research is focusing on a radionuclide identification algorithm that inverts this modeling within the constraints imposed by a handheld device. Key components of this work include incorporation of uncertainty as a function of both the background radiation estimate and the hypothesized sources, dimensionality reduction, and nonnegative matrix factorization. We have partially evaluated performance of our algorithm on a third-party data collection made with two different sodium iodide detection devices. Initial results indicate, with caveats, that our algorithm performs as good as or better than the on-board identification algorithms. The system developed was based on a probabilistic approach with an improved approach to variance modeling relative to past work. This system was chosen based on technical innovation and system performance over algorithms developed at two competing research institutions. One key outcome of this probabilistic approach was the development of an intuitive measure of confidence which was indeed useful enough that a classification algorithm was developed based around alarming on high confidence targets. This paper will present and discuss results of this novel approach to accurately identifying shielded or masked radioisotopes with radiation detection systems.

BACKGROUND

Radioisotope identification algorithms based on gamma-ray spectra take an unknown gamma-ray spectrum recorded by a detector as input and attempt to determine which isotopes emitted the observed gamma photons. Approaches can generally be divided into two broad areas. The first focuses on specific regions of interest in the unknown spectrum where gamma photons from certain isotopes would be expected to be seen. This approach is exemplified by peak picking, in which the locations of peaks in the spectrum are used to identify the gamma-ray sources (see, for example Routti and Prussin [9]). In a noisy spectrum, it can be difficult to determine the exact location of the peaks, and many approaches to this problem have been explored [11]. Furthermore, when shielding is present, the photons emitted at the expected peak energy may be shifted down to lower energies, to the point where a peak is not identifiable.

In the second approach, the algorithm attempts to match the full spectrum with templates of spectra from known isotopes. To accomplish this, some measure of similarity must be chosen; the most common

choices are correlation coefficients and error measures. For correlation measures, the correlation coefficient is computed between an unknown spectrum and a library of reference spectra [17].

Many authors have proposed least square error approaches. Salmon [1] and McWilliams, *et al.* [2] used non-weighted least squares procedures to solve for the target strengths of isotopes in an unknown spectrum. The former author mentions that a weighted approach would be more robust but does not pursue the idea. Other researchers have pursued weighted least squares approaches [3, 4, 5, 6, 7, 8, 10]. In particular, Eckhoff [10] describes a nonnegative weighted least squares algorithm, where the weights are determined by variance estimates from the observed spectrum, the template spectrum, and the estimator. If the template set of known spectra is constructed without shielding, however, the usefulness of these approaches is reduced when shielding materials are present. These approaches either assumed simple background subtraction or that the background could be described as a combination of the template sets; no separate background contribution to the weighting was incorporated.

Gamma photons interact with matter via several different processes: Compton scattering, the photoelectric effect, and pair production. We will not give a detailed description of these effects here because they are well documented in the literature (e.g., Knoll [13] or Gilmore and Hemingway [12]). However, in general, the interaction of gamma photons with matter between the source and detector causes the photon to be detected at a different energy than that at which it was emitted. This effect complicates the process of radioisotope identification because the measured spectrum from a given isotope may be significantly different from that which was expected. Therefore it is highly desirable to develop radiation isotope identifier (RIID) algorithms which are robust to the effects of shielding.

The Multiple Isotope Material Basis Set (MIMBS) method for RIID works on the principle that the effects of any shielding material can be approximately modeled using the effective atomic number. MIMBS uses a small number of basis shielding materials and unshielded reference spectra and attempts to solve, using a least squares approach, simultaneously for the composition of the shielding and the source emitters [16].

ALGORITHM REVIEW

In previous work [25], the authors developed an algorithm for shielded radioisotope identification based on modeling the detected spectrum as a random variable

$$\boldsymbol{\kappa_u} \approx \mathbf{Lc} + \boldsymbol{\lambda}_b + g(\mathbf{Lc} + \boldsymbol{\lambda}_b) \tag{1}$$

where $\mathbf{c} = [c(1), c(2), ..., c(m), ..., c(N)]^{\mathrm{T}}$ are the unknown isotope strengths, and $\mathbf{L}$ is our library of expected spectra for different isotopes in different shielding conditions ($\mathbf{L}(n, m) = \lambda_j(n)$ is the expected count for spectrum m in channel n for $c(m) = 1$). $g(\mu)$ indicates a normally-distributed random variable with mean μ and variance σ^2 and let $n(\sigma^2) = n(0, \sigma^2)$, and vector arguments to the normal function are interpreted as the diagonal values of a covariance matrix with no non-zero off-diagonal values. $\boldsymbol{\lambda}_b = [\lambda_b(1), \lambda_b(2), ..., \lambda_b(i), ..., \lambda_b(N)]^{\mathrm{T}}$ is the background spectrum estimate. We project into a scaled space $\xi(n) = [k_u(n) + k_r(n)]^{-1/2}$ Given this model, we can estimate the pdf of observed channel counts

$$p(\mathbf{k}_u|\mathbf{k}_r, \mathbf{L}', \hat{\mathbf{c}}) = \frac{1}{(2\pi)^{N/2} \prod_{i=1}^{N} s(i)} exp(f(\mathbf{k}_u, \mathbf{k}_r), \hat{\mathbf{c}}) \tag{2}$$

where the primes indicate the scaled variables and

$$f(\mathbf{k}_u, \mathbf{k}_r, \hat{\mathbf{c}}) = -\frac{1}{2}(\mathbf{L}'\mathbf{c} - [\mathbf{k}'_u - \mathbf{k}'_r])^T(\mathbf{L}'\mathbf{c} - [\mathbf{k}'_u - \mathbf{k}'_r]) \tag{3}$$

which we note is a simple function of the distance between the expected target spectrum $\mathbf{L}\hat{\mathbf{c}}$ for isotope strength vector $\hat{\mathbf{c}}$ and the estimated target spectrum $\mathbf{k}_u - \mathbf{k}_r$ in the scaled space. As such, we can use this

distance as a measure of fit of the model to the data. More particularly, we have a simple measure of how likely a particular model component is to be present in the data, since we can calculate how much the pdf increases when we add that component to the model:

$$p(\mathbf{k}_u|\mathbf{k}_r, \mathbf{L}', \hat{\mathbf{c}}) - p(\mathbf{k}_u|\mathbf{k}_r, \mathbf{L}'_m, \hat{\mathbf{c}_m}) \tag{4}$$

where L'_j and $\hat{\mathbf{c}_m}$ are, respectively, the expected library and estimated target strengths with the isotope in question removed.

CROSS-DETECTOR TESTING

In previous work, we reported the above-described algorithm's successful performance in an independent test performed after the algorithm was trained on simulated Identifinder spectra and tested against both collected and simulated Identifinder spectra. In the new tests reported below, we were interested in evaluating cross-detector performance. The original Identifinder-trained algorithm was applied to real data collected by Applied Research Associates on a variety of sources shielded by a variety of different materials, but using a different manufacturer's handheld detector. The data was organized into three data sets, and the sources and shielding materials for each data set are listed in Table 1.

Set	Isotopes	Shields
A	Co57, Co60, Ga67, Tc99m, In111, I131, Ba133, Xe133, Cs137, Tl201, U238, Am241, AmBe	Unshielded , 1-6 cm steel, 0.5" steel box, 0.5"-1.0" aluminum box, 0.25"-0.5" Pb Box, 5.0" Water, Poly., Pig
B	Cs137, Eu152, U235, U238, Pu239, Pu240	2"-4" Pb bricks + clamshell, 2-5 cm Steel Cylinder, 0.5"-1.0" Pb Box, 2" Pb Bricks, 0.25"-1.0" DU, Foil
C	Ga67, Tc99m, I131, Cs137, Eu152, Tl201, U233, U235, U238, Pu239	1.0" Al Box, 2-5 cm Steel, 2" Pb Pig, 0.25" DU, 0.5" DU, 0.5"-1.0" Pb Box, 2.0" Pb Bricks

Table 1: Isotopes and shielding materials used in the three data sets.

The on-board identification algorithm was used at the time of collection, and we compare the performance of our algorithm (labeled APL) to that of the on-board algorithm (labeled DetectorB). The classification results are given in Table 2.

Algorithm/Data set	Correct	Miss	False Alarms
APL / A	71	41	51
DetectorB / A	56	56	65
APL / B	7	42	29
DetectorB / B	18	31	26
APL / C	24	62	23
DetectorB / C	8	78	24

Table 2: Identification performance.

As can be seen in the results, these were challenging data sets for both algorithms. Our algorithm performed better on sets A and C, with more correct classifications and fewer misses and false alarms than the DetectorB. On data set B, however, the DetectorB had better performance. On this data set, many of the errors made by our algorithm involved identifying the wrong Uranium or Plutonium isotope. Additionally, the training data used for our algorithm was based on a simulated version of a detector from a different manufacturer; we believe that this mismatch between the training and testing data could be responsible for some errors.

PROPORTIONAL-WIDTH SMOOTHING

Data reduction is important as it enables more efficient processing suitable for handheld devices. More critically, it can also improve classification performance through the decrease in the number of parameters that need to be estimated. Handheld detectors have energy resolution that differs as a function of energy, and thus the maximal preservation of information for a given dimensionality must reflect this resolution. A common method of reducing data is smoothing and subsampling. Proportional width smoothing, where the smoothing width is proportional to the center energy, has been used with success in many applications, including automatic classification of speech signals (motivated by the human ears differing sensitivity to frequency change as a function of frequency). We performed proportional width smoothing of the isotope database using triangular filters; the filter bank is illustrated in Figure 1 below. An example of this smoothing applied to gamma ray spectral data is shown in Figure 2. The plot at right shows the original spectrum of Ra226 shielded by 0.953 cm of lead. The plot at left shows the smoothed spectrum. The number of spectral coefficients has been reduced by more than an order of magnitude, from 694 to 44. Nonetheless, all of the major spectral peaks, and most of the minor ones, are preserved in the smoothed spectrum.

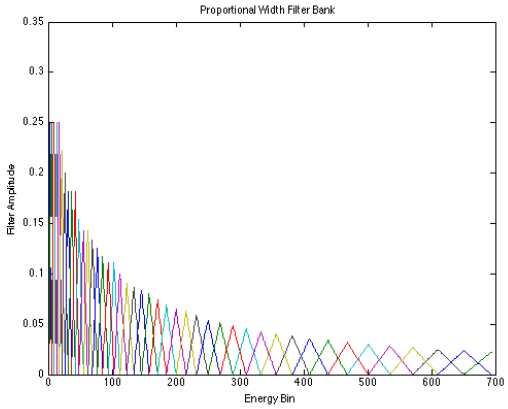

Figure 1: Proportional-width filter bank using overlapping triangular filters for data reduction.

Isotope Identification in reduced data space

The advantage of data reduction becomes apparent in automatic classification, as the results given in Table 3 indicate. Isotope classification performance using the non-negative least squares algorithm (**lsqnn** in Matlab) increased slightly (from score of 67.31% to max of 67.96%) over our baseline results [25] over a range of reduced data cardinalities, up to a nearly order-of-magnitude reduction (from 694 to 88 coefficients). Further reduction to only 44 coefficients saw a small reduction of performance (65.55%). While these performance

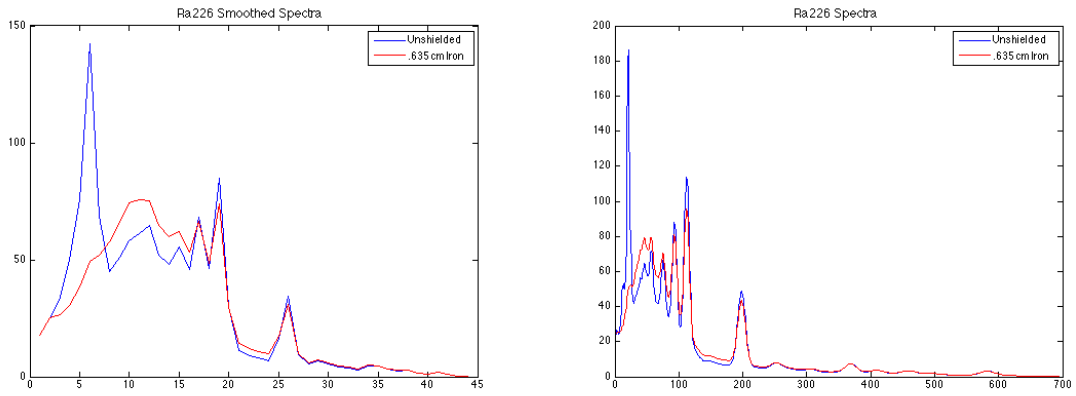

Figure 2: left: gamma ray spectrum of Ra226 shielded by .953 cm of lead; right: same isotope spectrum after data reduction using filter bank of Figure 1.

gains are small, they are significant because of the reduction in overall computation that results from being able to operate in this greatly reduced data space. Handheld detectors may not be able to process the full band data using advanced algorithms, but may be able to do so on lower cardinality data. Furthermore, classifier training and performance is more robust when fewer parameters, corresponding to few input variables, need to be estimated.

LSQNONNEG	44 bands	88 bands	130 bands	172 bands	214 bands	694 bands (all)
Score (% of possible)	65.55%	67.73%	67.89%	67.96%	67.55%	67.31%

Table 3: LSQNONNEG results

We have also explored an alternative to our current approach of using nonnegative least squares to estimate isotope strengths. Specifically, we have run initial experiments using regression under a Poisson data model; the results are shown in Table 4. A slight increase in performance on the reduced data was obtained using iterations derived under a Poisson assumption. Interestingly, performance dropped on the full data set when Poisson regression was used. One possible reason for this effect is that the scoring algorithm we use assumes Gaussian-distributed data. For large counts, the Poisson distribution approaches a Gaussian. The smoothing we use in data reduction has the added effect of producing a large-count Poisson random variable for energy bins with significant counts, so the Gaussian assumption is reasonable. The same does not hold true for the unsmoothed data, however. If this hypothesis is valid, then the solution is to modify the scoring algorithm to likewise employ Poisson statistics.

MATRIX TRANSPORT MODELING

We can define a partitioning of a portion of the gamma-ray energy spectrum into N_I regions by choosing a set of boundaries $s_I(i), i = 0, 1, ...N_I$ and $s_I(i) < s_I(i+1)$. Using this notation, we can represent the spectrum of a radioactive source as a length-N_I vector $\mathbf{x}$, where $x(i)$ is the probability that the source will emit a photon with energy between $s_I(i)$ and $s_I(i+1)$ over some reference time period. Given a second

POISSON	88 bands	172 bands	694 bands (all)	LSQNONNEG
Score (% of possible)	68.42%	68.25%	65.72%	67.31%

Table 4: POISSON results

partioning of a region of the energy spectrum defined by $s_M(j), j = 0, 1, ...N_M$ (which is not necessarily the same as that defined by $\mathbf{s}_I$), we can represent the expected spectrum of gamma rays which pass through a material with label η as $\mathbf{y} = \mathbf{A}_\eta \mathbf{x}$, where $A_\eta(j, i)$ is the probability that a photon emitted from the source with energy in partition i will pass through material η with energy in partion j. Such a matrix is referred to as a transition matrix or response matrix. Similarly, we can represent the response of a real detector (with energy partioning defined by $s_O(k), k = 0, 1, ...N_O$) to an entering gamma-ray spectrum $\mathbf{y}$ as $\mathbf{z} = \mathbf{B}\mathbf{y}$ where $B(k, j)$ is the probability that a photon entering the detector with energy in partition j will result in a detection η with energy in partion k. (We note that, for a real detector with real time-gating, this formulation neglects cases where multiple photons enter the detector close enough in time that they fail to be resolved as separate events.) The entirety of the mapping from emitted to detected spectrum can then be modeled as $\mathbf{z} = \mathbf{B}\mathbf{A}\mathbf{x}$.

Estimation of these matrices for specific detectors and shielding conditions was done using software developed at Sandia National Laboratories for this research. The software, which uses the transport modeling engine underlying GADRAS, enables the user to specify arbitrary partitions $\mathbf{s_I}, \mathbf{s_M}$ and $\mathbf{s_O}$ and to select detector and shielding conditions with the same flexibility as in GADRAS. An example transfer matrix, for Identifinder-NG, is shown graphically in Figure 3.

It has been noted that, for many tasks including source isotope identification, deconvolution of the transition matrix would be of great value in simplifying the search algorithm. This has been studied in situations where there is no intervening material η (i.e. where $\mathbf{A}$ is the identity matrix), in which case the goal has been stated as restoring counts in the Compton continuum onto their corresponding photo-peaks. However, for detectors of practical use the matrix is ill-conditioned and direct inversion is unstable [24]. The well-known Shepp and Vardi ML-EM algorithm provides an iterative method for estimating $\mathbf{y}$ which uses only the forward transfer equation and then updates based on the error in the match of the predicted to the observed data. However, the instability in the direct inversion is related to the desired resolution of the estimate of the emitted spectrum $\mathbf{x}$; as we presented in the previous section, a proportional-width smoothed filtering can allow a reduced-dimensionality estimate of the input and thus of the transfer matrix. As a result, we can use pseudoinverse methods to use the high-dimensional detector output to create a low-dimensional detector input estimate which can then be searched over shielding space using much simpler methods. An example is shown in Figure 4.

NON-NEGATIVE MATRIX FACTORIZATION

The effects of shielding on gamma ray spectra are two-fold: an overall attenuation of emissions, and a shift of the emitted gamma rays to lower energies. This energy shift can be represented by a transform matrix that maps the input energy to the output energy of the emitted gamma rays. In future work, we will investigate methods to compensate for shielding and detector variability by combining modern signal processing methods such as non-negative matrix factorization (NMF) [21] with GADRAS-based modeling of transport and detector physics to. In an ongoing collaboration with APL-UW, Sandia National Laboratory is modifying GADRAS to work in a batch mode and to model the shielding and detector physics separately. We will

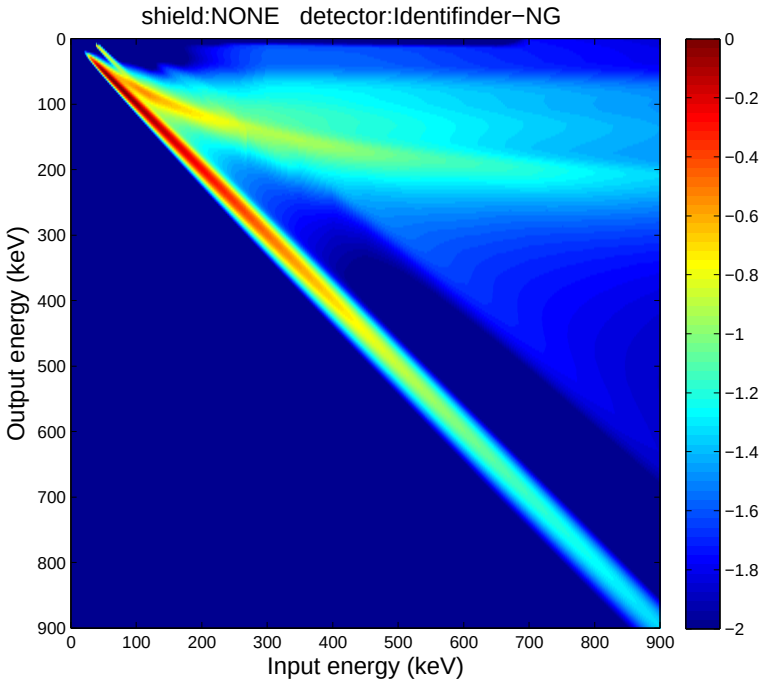

Figure 3: Log values of the estimated transfer matrix for the Identifinder-NG. One vertical column of this matrix represents the probability of detection at different energies, given a particular energy for a photon that enters the detector.

use this tool to build a database of shielding transfer functions and shielded and unshielded isotope spectra for a wide range of isotopes and shielding materials/thicknesses. A large database that includes the many possible sources found in operational environments will also improve performance in the presence of clutter. Shielding imposes an additional source of data variation (beyond that due to source variation), both by reducing peaks in the spectrum and by shifting emitted gamma rays to lower energies. New high-resolution (1% to 3% FWHM) gamma-ray spectroscopy systems will be particularly sensitive to shielding-induced spectral variation. Sparse NMF [22] will be used on observed gamma-ray spectra to separate shielding effects from hypothesized source spectra, yielding an estimate of the source spectrum which will be used for isotope identification. The details of this factorization are described next.

In the general case, we observe a spectrum $\boldsymbol{\kappa}$ composed of a background spectrum $\boldsymbol{\lambda}_b$ and a sum of N isotope spectra $\boldsymbol{\lambda}_m$ with strengths $c(m)$, which are subject to the effects of shielding and the detector in use, represented by the matrices $\mathbf{A}$ and $\mathbf{B}$, respectively. The matrix $\mathbf{A}$ models the shift of energy by the shielding to lower energies, and hence is non-diagonal. The observed spectrum can then be expressed succinctly in the form $\boldsymbol{\kappa} = \boldsymbol{\lambda_b} + \sum_m \mathbf{BA}c(m)\boldsymbol{\lambda_m}$. The challenge is to estimate the strengths $c(m)$ of an unknown number of isotopes, given an observed gamma ray spectrum, a library of known isotope spectra $\boldsymbol{\lambda}_m$, an estimate of the background $\boldsymbol{\lambda}_b$, and the detector characteristics given by $\mathbf{B}$. The shielding transform A is unknown. Sparse non-negative matrix factorization will be useful in order to prevent over-fitting with multiple small components, as standard least squares would do.

The standard problem in non-negative matrix factorization is to find non-negative matrices $\mathbf{W}$ and $\mathbf{H}$ that minimize $||\mathbf{V} - \mathbf{WH}||^2$ for some observed data matrix $\mathbf{V}$. Sparse NMF adds a sparsity constraint such as the L_1 norm on the matrix $\mathbf{H}$. The solution to this problem is iterative and typically involves alternating

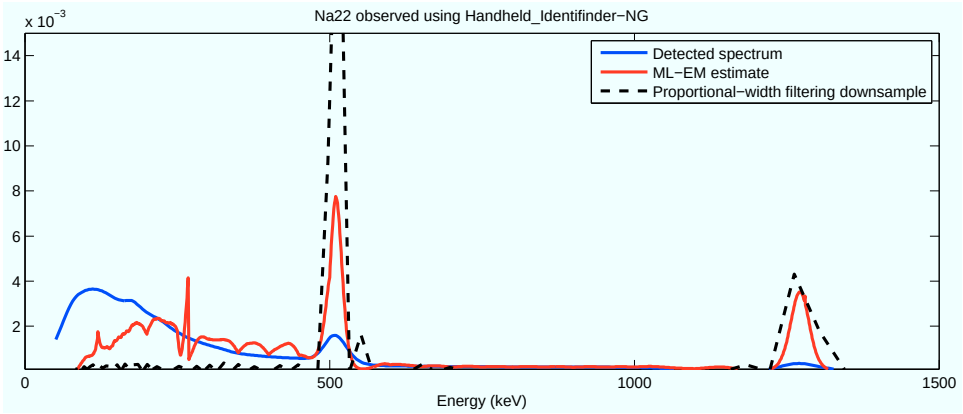

Figure 4: Removal of the effects of detector mapping and subsequent removal of (unknown) shielding effects. The (simulated) observed spectrum is shown in blue. For a known detector response matrix, the ML-EM estimate of the emitted spectrum is shown in red. The proposed method, shown as a black dotted line, has fewer artifacts and higher counts at the true source peaks, but at a tradeoff of lower resolution at high energies.

projections to solve for $\mathbf{W}$ and $\mathbf{H}$. We need to solve a more general problem. Equating the NMF problem with our gamma ray spectrum formulation $\boldsymbol{\kappa} = \boldsymbol{\lambda}_b + \sum_m \mathbf{BA}c(m)\boldsymbol{\lambda}_m$, we identify the observed spectrum $\boldsymbol{\kappa}$ with the data matrix $\mathbf{V}$, the vector of unknown isotope strengths $\mathbf{c}$ with the unknown matrix $\mathbf{H}$, and the unknown shielding transform $\mathbf{A}$ with the matrix $\mathbf{W}$. Our problem is more general, however, in that we also have the known detector operator $\mathbf{B}$, and the known isotope spectra $\boldsymbol{\lambda}_m$ – just their strengths $\mathbf{c}$ are unknown. Denoting this library of isotope spectra by $\mathbf{L}$, absorbing the background spectrum into the observed spectrum by defining $\bar{\boldsymbol{\kappa}} = \boldsymbol{\kappa} - \boldsymbol{\lambda}_b$ and adding a sparsity constraint to the isotope strength vector, we have the general sparse NMF problem for gamma ray spectroscopy:

$$\hat{\mathbf{A}}, \hat{\mathbf{c}} = \arg\min_{\mathbf{A},\mathbf{c}} ||\bar{\boldsymbol{\kappa}} - \mathbf{BALc}||^2 + \gamma|\mathbf{c}|_1. \tag{5}$$

Implicit in this formulation is a Gaussian assumption on the residual, imposed by the use of squared error as the optimization criterion (NMF was originally implemented under this Gaussian assumption). This shortcoming may be avoided by performing sparse NMF using Poisson statistics. Recent work has investigated NMF for other distributions, including Poisson [23], which is more appropriate for gamma-ray spectroscopy. This approach would use (and yield) the correct statistics, and also provide an efficient parameterization for the data through maximum likelihood optimization, leading to improved classification performance.

In all cases, a measure of the goodness of fit is provided by the model error at end of the NMF routine, which will be used to assign a confidence to the isotope identification. For testing, performance estimates can be obtained through a combination of predictor output probabilities (classification error) and modeling error. The heavy computational load would be off-line during classifier design; testing should be very efficient.

SUMMARY

The work presented in this paper represents updates in a number of areas related to a previously successful radioisotope detection algorithm. We showed new results on a mismatched cross-detector training/testing

experiment. We showed a physics-justified approach to reducing the resolution of the spectral estimate, and demonstrated that it could be achieved without degradation of the algorithm performance. We used this result to suggest a straightforward method of removing detector effects and focusing on shielding effects as a method of increasing future algorithm performance. We introduced non-negative matrix factorization as a method for analyzing the shielding effects in this environment.

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