Bayesian unmixing algorithms for identification of gamma sources using organic scintillators

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***Abstract*— One major problem with nuclear security measurements involves source identification in the presence of low signal-to-background ratio. This scenario is common to several applications, ranging from radiation identification at portal monitors to radiation source search with unmanned vehicles. In this context of identification of a large variety of sources, including natural and medical sources, sensitive sources of particular interest, but also potentially new/unknown sources for which no reference measurement is available, statistical methods are particularly appealing for their ability to capture the random nature of the measurements. Among them, Bayesian methods form a generic framework allowing for uncertainty quantification and propagation, which is of prime interest for detection (of known and unknown sources), classification, and quantification of smuggled nuclear and radiological materials. We demonstrate the use of Bayesian models for the identification of mixed gamma sources, measured with organic scintillators within short acquisition times. We also compare the estimation performance using two different materials: liquid EJ-309 and stilbene crystal.**

Keywords—source identification, statistical inference, unmixing

I. Introduction

One major challenge with nuclear security measurements involves the installation of radiation portal monitors (RPMs) at border crossings. RPMs typically contain ^{3}He proportional counters embedded in polyethylene for neutron detection, and slabs of polyvinyl-toluene (PVT) scintillator for gamma-ray detection. In the presence of natural background radiation, customs and border protection agents screen inbound vehicles and cargo container for suspicious levels of radiation relative to background and flag these for a more thorough secondary inspection. Although organic scintillators do not exhibit photopeaks, they are relatively inexpensive and easy to operate. They could benefit from statistical computational methods and in particular Bayesian approaches to detect, classify, and quantify smuggled nuclear and radiological materials and therefore reduce the need of time-consuming secondary inspections. In this work, we apply Bayesian models for the identification of the components of an unknown mixed radioactive source and compare the identification performance achieved by two different materials: liquid EJ-309 and stilbene crystal.

II. Materials and Methods

A. Experimental Methods

An RPM typically consists of a set of PVT, organic scintillation detectors. We measured several single sources and their combinations using two types of organic scintillation detectors, i.e., 7.62 cm diameter by 7.62 cm length EJ-309 and 5.08 cm diameter by 5.08 cm length stilbene crystals. The stilbene exhibits a better energy resolution, compared to the EJ-309 [1], because of the combined effect of the different composition and a more efficient light transport in a smaller detector cell [2]. Short-time acquisitions were performed, to resemble a realistic 3-s data acquisition time window of an RPM inspection. Sources tested included 51 g HEU (89.9% 235U), 6.6 g WGPu (93% ^{239}Pu), 185 kBq ^{57}Co, 518 kBq ^{133}Ba, 592 kBq ^{137}Cs, and 1740 kBq ^{241}Am. Medical isotopes, especially ^{99m}Tc, constitute a growing source of RPM nuisance

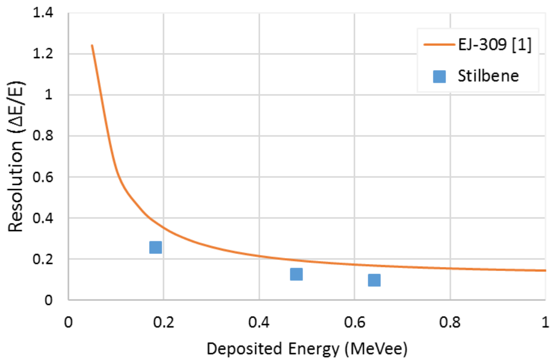

Figure 1. Measured energy resolution for EJ-309 [1] and stilbene detector. 1SD error bars are comparable to the measured stilbene data points.

alarms. Measurements associated with 260 kBq liquid vials of ^{67}Ga, ^{123}I, ^{131}I, ^{111}In, ^{201}Tl, and ^{99m}Tc were acquired at the University of Michigan C.S. Mott Children's Hospital with both EJ-309 and stilbene detectors [3]. Pulse height distributions (PHDs) associated with 30-minute measurement times were acquired for each individual medical isotope, and a down-selection process was used to create individual short measurement time PHDs with total counts similar to the radionuclide source measurements. Each source was also acquired for a longer period and, in this case, the PHD was used to compile a library of known nuclides.

B. Bayesian Algorithms

We use Bayesian estimators relying on a Poisson noise model to identify the potential radioactive constituents of an unknown mixture of radionuclides. We consider an observed response **y**, and an unknown vector of mixing coefficients **x,** associates with the different sources in the available gamma library. Since limited information is assumed to be available about the actual mixing coefficients of the observed spectrum, we define a weakly informative prior distribution **f(x)**, which only ensures that the unknown coefficients should be non-negative, i.e., $x \geqslant 0$.

Using the Bayes rule, the posterior distribution of x given y denoted **f(x|y)** satisfies **f(x | y)=f(y|x)f(x)/f(y)**. To solve for the mixing contribution coefficients x, we can maximize **f(x|y)** with respect to x, yielding a maximum a posteriori (MAP) technique, which provides us with the most likely solution (a posteriori). This can be solved using convex optimization tools [4]. Alternatively, we can compute the expectation of **x**|**y**, i.e. the posterior mean, which will minimize on average the squared error between the estimated mixing coefficients and their actual values. This estimator is known as the minimum mean squared error (MMSE) estimator. Given the complex shape of f(x|y), the expectation of x|y cannot be computed analytically and a Markov chain Monte Carlo (MCMC) method similar to that proposed in Altmann et al. [5] is used to approximate it numerically.

III. RESULTS

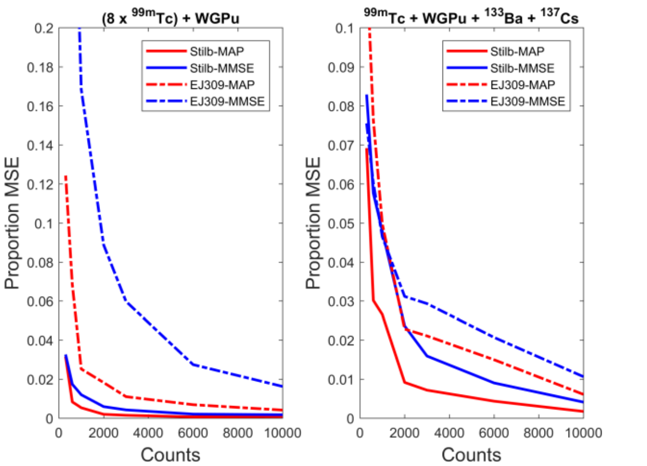

Figure 2. Mean square errors (MSEs) of the composition of two mixtures acquired using the stilbene and EJ-309 detectors and using the MAP and MMSE estimators.

Figure 2 shows the mean square errors in estimating the composition of two mixtures: ^{99m}Tc + WGPu (ratio 8:1) and ^{99m}Tc + WGPu + ^{133}Ba + ^{137}Cs (ratio 1:1:1:1) using both detectors and both methods. One may notice that the stilbene consistently better predicts the composition, compared to the EJ-309. Figure 3 shows two examples of source quantification results for the MAP and MMSE estimators, for the stilbene detector. One may notice that the MAP and MMSE methods provide less reliable estimates when four sources are present, while it accurately classifies the sources when a mixture of two radionuclide is presented to the algorithm. In these two tests, no additional information constraining the maximum number of sources was added. By constraining the number of source, an improvement in the quantification results may be expected [6].

The MMSE estimator, computed with associated confidence regions (Fig. 4), generally provides more robust results, affected by a lower mean squared error,

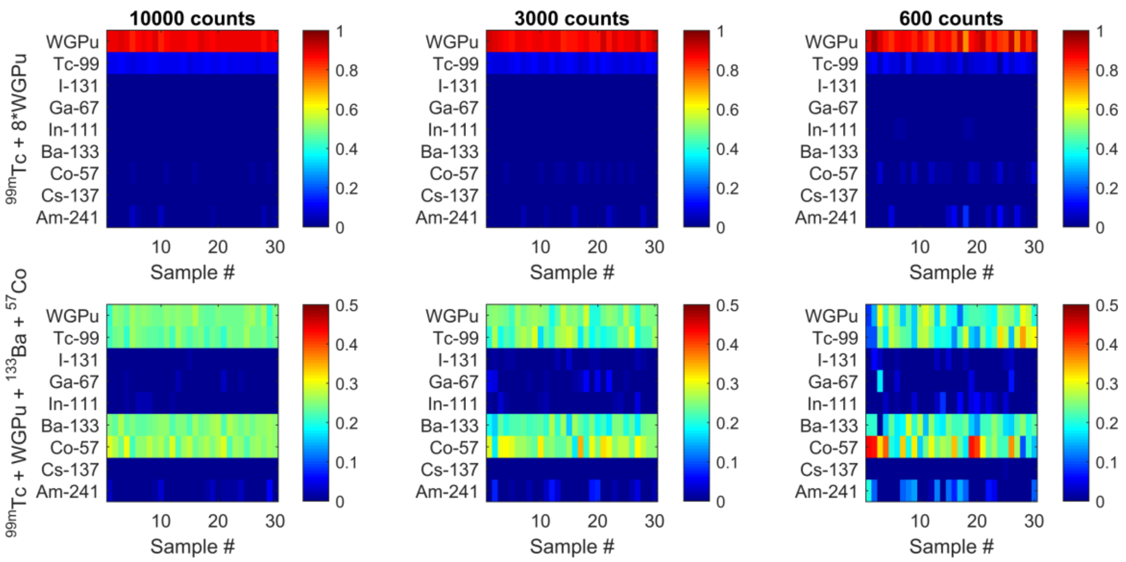

Figure 3. "Simple" (top) and "Complex" (bottom) quantification scenarios. The stilbene detector were irradiated using two sources (i.e., WGPu and ^{99m}Tc (top)) and 4 sources (^{133}Ba, WGPu, ^{99m}Tc and ^{137}Cs, (bottom)) for 30 short time acquisitions.

when compared to the MAP estimation. Correlation matrices like those shown in Fig. 4, obtained using MCMC, are an additional tool when SNM might be mistakenly classified as naturally occurring radioactive materials (NORM).

Figure 4 depicts the a-posteriori correlation matrix for mixing coefficients of one sample of the second mixture shown in Figure 3 (^{137}Cs + HEU + ^{99m}Tc + ^{133}Ba). This correlation matrix shows that the Bayesian estimators (i.e., the MAP and MMSE estimators) struggle to distinguish ^{99m}Tc from ^{123}I, as well as ^{133}Ba from ^{131}I. In other words, the algorithm estimated with high confidence that, for example, either ^{99m}Tc or ^{123}I is present in the mixture, but it cannot necessarily determine which of the two is present. In general, ^{99m}Tc and ^{123}I exhibit very similar PHDs, thus explaining why the Bayesian estimator struggles to discern these sources.

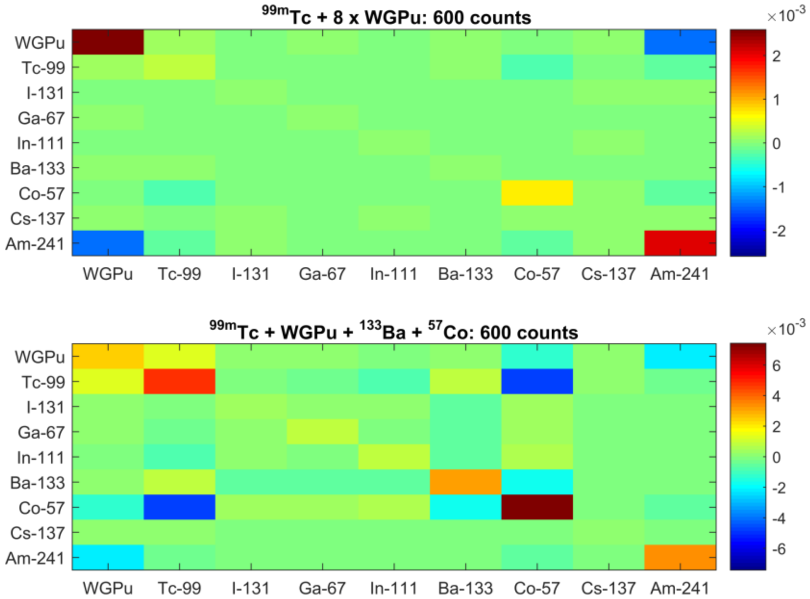

Figure 4. A-posteriori covariance matrices of mixture proportions computed via MCMC for two mixtures from data composed of 600 counts each and measured using a stilbene detector.

IV. CONCLUSIONS

Computational methods were developed to process gamma-ray measurements from mixed sources with up to four different radionuclides out of a library of 12 known radionuclides. The Bayesian MAP and MMSE techniques enabled the identification of sources present in the measured mixtures using a known spectral library. For each mixture, 30 independent samples were tested with each algorithm. The identification obtained with the MMSE method is more robust compared to the MAP. The MCMC approach also yields a-posteriori covariance matrices that can be used to quantify the confidence associated with the classification results and therefore aid the user to determine the need of a secondary inspection in an RPM application. The ability to identify and decompose mixed sources of radiations is crucial for spectroscopic RPMs. Spectroscopic RPMs must be able to detect weak SNM sources masked by a stronger NORM or nuisance radiation source and Bayesian algorithms proved to be useful tools to improve the classification accuracy of high efficiency, yet low energy resolution of organic scintillators.

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