Spectral unmixing applied to fast identification of γ-emitting radionuclides using NaI(Tl) detectors

Objectives

Fast and robust spectroscopic identification of γ-emitting radionuclides is key for:
- Radiation portal monitors (RPM) (e.g. located at borders) used to prevent illegal nuclear material trafficking;
- in situ environmental measurements following a radiological or nuclear accident. The development of reliable automatic systems is essential to minimize the number of expert interventions in decision-making due to large flow of measurements. For this purpose, a new algorithm based on spectral unmixing has been developed for fast automatic identification of γ-emitting radionuclides in natural background radiation.

Constraints

The automatic identification code was designed to meet the constraints of measurements at low statistics with RPMs equipped with scintillation detectors (plastic, NaI(Tl)):
- Short measurement duration (few seconds) corresponding to γ-ray spectra of a few hundred to few thousand counts;
- Low false-alarm rate (alpha risk < 0.1%) in automatic decision-making;
- Full-spectrum analysis without using region-of-interest (ROI) techniques;
- Implementation in embedded digital systems (FPGAs, microcontrollers).

Mathematical background in short

A measured γ-spectrum is \( s = [s_1, \ldots, s_m] \) is composed of \( m \) channels. In each channel \( i \), the counting can be modeled with Poisson statistics depending on parameter \( \lambda_i \). For spectral unmixing purpose, the Poisson parameters \( \lambda_i \) are linear combination of individual spectral signatures \( \phi_k \) (detector response) related to the γ-emitting radionuclides (with index \( k \)) to be identified and the respective mean counting \( a_k \) is to be estimated. Considering the measured spectrum \( s \), the likelihood of the mean counting \( a_k \) is given by the following joint probability (Poisson statistics):

\[
Pr(s_1, \ldots, s_m|a_1, \ldots, a_k) = \prod_{i=1}^{m} \frac{e^{-\lambda_i} \lambda_i^{s_i}}{s_i!}
\]

where \( n \) is the number of spectral signatures \( \phi_k \). The maximization of this expression can be obtained by following the first-order optimality condition:

\[
\mathcal{V}_k = 1, \ldots, n; \quad \frac{1}{\lambda_k} a_k e^{-\lambda_k} \mathcal{Z} = 0
\]

Based on the above condition, each mean counting \( a_k \) can be estimated using a multiplicative update algorithm with non-negative constraint designed so that at each updating iteration \( \ell \) (with normalized spectral signatures), as follows:

\[
\mathcal{V}_k = 1, \ldots, n; \quad a_k^{\ell+1} = a_k^{\ell} \left( 1 + \frac{1}{\lambda_k} \sum_{i=1}^{m} \frac{S_i}{s_i} \phi_i \right)^{-1}
\]

The multiplicative update algorithm was tested using a dictionary of 10 spectral signatures including natural background measured with an unshielded 3’x3’ NaI(Tl) detector.

The spectral decision threshold (DT) and detection limit (DL) for five radionuclides used for RPM’s testing are summarized in Table 1.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>DT</th>
<th>DL</th>
<th>A_{\text{ECEM}}/kBq</th>
</tr>
</thead>
<tbody>
<tr>
<td>241Am</td>
<td>28</td>
<td>60</td>
<td>97</td>
</tr>
<tr>
<td>137Ba</td>
<td>82</td>
<td>160</td>
<td>63</td>
</tr>
<tr>
<td>57Co</td>
<td>57</td>
<td>110</td>
<td>54</td>
</tr>
<tr>
<td>60Co</td>
<td>48</td>
<td>115</td>
<td>36</td>
</tr>
<tr>
<td>133Cs</td>
<td>37</td>
<td>85</td>
<td>59</td>
</tr>
</tbody>
</table>

Table 1: DT and DL obtained by spectral unmixing (in red). The DL values in Bq unit are compared to the results based on the ROI technique and activities of RPM’s testing sources specified in IEC 62484:2010.

The aim of the TRI-LATAC detection system (3’x3’ NaI(Tl) detector) is to implement automatic decision-making for non-expert users for first screening of food samples after a radiological or nuclear accident.

The spectral unmixing algorithm was tested in the case of the identification of 134Cs, 137Cs and 241Am according to the Council regulation 2016/52 (Euratom) defining maximum permitted levels (MLP) of radioactive contamination of food.

The DL values (beta risk = 2.5%) were calculated using a dictionary of 7 spectral signatures (K_k, 111In, 131I, 137Cs, 212Bi, 241Am, natural background) for aqueous solutions, fresh plants and soils in SG5000 Becker (500 mL).

As presented in Table 2, a measurement duration lower than 3 min is sufficient to comply with the MLPs specified in the case of infant food.

Table 2: DL values obtained by spectral unmixing compared with maximum permitted levels (MLP) in the case of infant food (Council regulation 2016/52 (Euratom)).

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Soil</th>
<th>Fresh Plant</th>
<th>Aqueous solution</th>
<th>MLP/Bq kg⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>134Cs</td>
<td>37</td>
<td>169</td>
<td>48</td>
<td></td>
</tr>
<tr>
<td>137Cs</td>
<td>49</td>
<td>216</td>
<td>62</td>
<td></td>
</tr>
<tr>
<td>241Am</td>
<td>41</td>
<td>179</td>
<td>52</td>
<td></td>
</tr>
</tbody>
</table>

Table 2: DL values obtained by spectral unmixing compared with maximum permitted levels (MLP) in the case of infant food (Council regulation 2016/52 (Euratom)).

Conclusion

The multiplicative update algorithm was developed for maximum likelihood estimation with Poisson statistics using a joint probability that accounts for the spectral signatures of the γ-emitting radionuclides to be identified and natural background. Combined to full-spectrum analysis, the spectral unmixing algorithm takes into account optimally statistical fluctuations of γ-ray spectra for decision-making at low statistics.

The spectral unmixing algorithm is also well-adapted for implementation in embedded digital systems. Automatic decision-making can be obtained with a maximum execution time lower than 100 ms in a low-cost Xilinx ZYNYC FPGA. This value complies with measurements with RPMs. The next step of the study will be the improvement of decision-making in the case of complex mixtures of γ-ray emitters.