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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).

Spectral unmixing based on a multiplicative update algorithm for maximum likelihood estimation with Poisson statistics

Mathematical background in short

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

$$\mathcal{P}(s_1,\cdots,s_m|a_1,\cdots,a_n) = \prod_{i=1}^{m} \frac{\left(\sum_{j=1}^{n} \phi_i^j\, a_j\right)^{s_i}}{s_i!} e^{-\sum_{j=1}^{n} \phi_i^j a_j}$$

where n is the number of spectral signatures ϕ^k. The maximization of this expression can be obtained by fulfilling the following first-order optimality condition:

$$\forall k = 1,\cdots,n; \qquad \frac{1}{\sum_{i=1}^{m} \phi_i^k} \sum_{i=1}^{m} \frac{s_i}{\sum_{j=1}^{n} \phi_i^j \hat{a}_j} \phi_i^k = 1.$$

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

$$\forall k = 1,\cdots,n; \qquad a_k^{(\ell+1)} = a_k^{(\ell)} \sum_{i=1}^{m} \frac{s_i}{\sum_{j=1}^{n} \phi_i^j a_j^{(\ell)}} \phi_i^k.$$

Multiplicative update algorithm

Input

Spectral signatures of each γ-emitter to be identified: $\boldsymbol{\Phi} = \left[\phi_i^k\right]$ $(k = 1..n)$; n: number of radionuclides to be identified and natural background

Experimental spectrum: $\boldsymbol{s} = [s_1,\cdots,s_m]$; m: number of channels

Initialization

Maximum number of iterations: p_{max}

Mean counting: $a_k^{(0)} = n^{-1}$

While $p < p_{max}$ do

Multiplicative updating rule: $\boldsymbol{a}^{(p+1)} = \boldsymbol{a}^{(p)} \odot \left(\boldsymbol{\Phi}^T \boldsymbol{s} \oslash \left(\boldsymbol{\Phi a}^{(p)}\right)\right)$

end

Output

Estimated mean counting a_k for each γ-emitter to be identified

The mathematical operators ⊙ (resp. ⊘) are the element-wise product (resp. division). The spectral signatures $\boldsymbol{\Phi} = \left[\phi_i^k\right]$ can be obtained experimentally or by Monte Carlo radiation transport simulations.

Calculations of decision threshold (DT) and detection limit (DL) for five radionuclides used for RPM's testing (^{57}Co, ^{60}Co, ^{133}Ba, ^{137}Cs, ^{241}Am)

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

Each spectral signature corresponds to a 1024-channel γ-spectrum (from 40 keV to 2 MeV) obtained using point sources placed at a distance of about 1 m (^{57}Co, ^{60}Co, ^{88}Y, ^{133}Ba, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{207}Bi, ^{241}Am). The mean counting rate of natural background is about 250 s^{-1}.

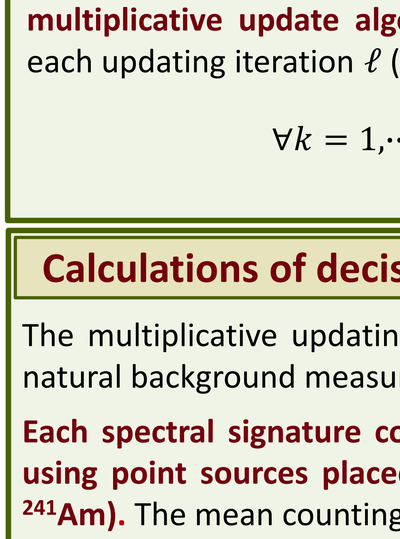
The decision thresholds (DT-5s) for each γ-emitting radionuclide in the dictionary were determined with an alpha risk of 0.1% from Monte Carlo calculations of 10^5 simulated background spectra according to Poisson process (mean counting equal to 1250 corresponding to a measurement duration of 5 s).

In the same manner, **the detection limits (DL-5s) for each γ–emitting radionuclide used for RPM's testing in Table 1 were determined with a beta risk of 0.1%**. In Table 1, the results given by the multiplicative update algorithm (full-spectrum analysis) are compared to the conventional ROI technique. The DL values are significantly lower in the case of ^{133}Ba and ^{60}Co.

	DT-5s	DL-5s	DL / kBq	DL_{ROI} / kBq	$A_{IEC62484}$ / kBq
^{241}Am	28	60	97	148	1740
^{133}Ba	82	160	63	177	333
^{57}Co	57	110	54	89	555
^{60}Co	48	115	36	71	260
^{137}Cs	37	85	59	97	590

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.

Application to the TRI-LATAC system developed at IRSN/LMN for food analysis after a radiological or nuclear accident



TRI-LATAC system

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

The spectral unmixing algorithm was tested in the case of the identification of ^{134}Cs, ^{137}Cs and ^{131}I 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 (^{40}K, ^{131}I, ^{134}Cs, ^{137}Cs, ^{210}Pb, ^{241}Am, natural background) for aqueous solutions, fresh plants and soils in SG500 beakers (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**.

	DL / Bq.kg^{-1} (2 min measurement)			MLP / Bq.kg^{-1}
	Soil	Fresh Plant	Aqueous solution	
^{134}Cs	37	169	48	400
^{137}Cs	49	216	62	
^{131}I	41	179	52	150

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 multiplicative update 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 ZYNQ 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.

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