The Non Negative Least Square Applied to the Full Spectrum Analysis

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INTRODUCTION

An increasing demand of environmental radioactivity monitoring comes both from the scientific community and from the society. This requires accurate, reliable and fast response preferably from portable radiation detectors. Thanks to recent improvements in the technology, γ-spectroscopy with sodium iodide scintillators has been proved to be an excellent tool for in-situ measurements for the identification and quantitative determination of γ-ray emitting radioisotopes, reducing time and costs. Both for geological and civil purposes not only ^{40}K, ^{238}U, and ^{232}Th have to be measured, but there is also a growing interest to determine the abundances of anthropic elements, like ^{137}Cs and ^{131}I, which are used to monitor the effect of nuclear accidents or other human activities. Among the RAD-MONITOR experiment several detection systems have been developed following the request of fast and portable measurements, and a careful selection of the γ-spectra analysis procedure has been consequently performed. The three windows method, suggested by the IAEA [1] has shown limitations, since it becomes imprecise for short time acquisitions and it suffers the poor intrinsic energetic resolution of NaI(Tl) detector. In particular, the Compton continuum around ^{137}Cs photopeak is generally very intense compared to the intensity of 661 keV γ-line. This translates into long acquisition times. Moreover, the intrinsic energetic resolution of sodium iodide detectors prevents to resolve the doublet at 609 keV (^{214}Bi) and 661 keV (^{137}Cs). This issue can be solved only by post processing the data. In any case the windows analysis method requires a prior knowledge of the presence on site of such radioisotope. As a matter of fact, this method is blind to unexpected radionuclides.

Significant improvements in γ-ray spectrum analysis have been obtained by implementing the full spectrum analysis (FSA) method [2]. Since the FSA uses the full extent of the spectrum, as opposed to the three windows method, it needs a much lesser statistic to reach the necessary accuracy. This, in turn, means a drastic reduction in acquisition times.

THE FSA ALGORITHM

The spectra acquired in situ are fitted by a linear combination of the fundamental spectra derived for each isotope from the calibration analysis. The events registered in each channel in the measured spectrum, N, can be expressed as:

$$N(i) = \sum_{k=1}^{4} C_k S_k(i) + B(i) \quad (1)$$

where:

- $N(i)$ are the counts in the channel i,
- C_k are the concentration of the element k,
- $S_k(i)$ are the associated counts to the fundamental spectrum of the element k in the channel i,
- $B(i)$ are the counts in the channel i due to the intrinsic background.

and the index k stays for ^{40}K, ^{232}Th, ^{238}U, and ^{137}Cs. It has become a conventional representation for in-situ measurements to express the concentrations of natural radioisotopes in their respective abundances, where ^{40}K is given in percentage weight while eU and eTh are given in ppm. ^{137}Cs is given in Gy/y.

During the calibration of the system the fundamental spectra (the S matrix) are obtained by solving equation 1 with the radionuclide concentrations (the C_k coefficients) measured in the sites where the calibration measurements were performed. It has to be noted that FSA calibration method produces also the intrinsic background, B, which can be compared with a spectrum acquired with the detector inside a thick lead shielding.

Only the energy range from 300 keV up to 2900 keV is considered in the analysis. Below 300 keV there is a strong presence of the backscattering events which depends on the atomic number and density of the surrounding materials. Above 2900 keV only the cosmic ray contribution is present. Once the first solution has been obtained, in order to improve the χ^2 minimization, a trimming procedure is executed by changing the site concentrations (C_k) in small steps around the measured intervals and repeating the matrix solution.

The simple χ^2 minimization has shown artifacts in the reconstructed [S] matrix due mostly to the correlation of radioisotopes present in different calibration sites. Moreover a possible variation of detector resolution during in situ measurements could lead to crosstalk effects in the analysis. The NNLS constrain has been introduced in the FSA

algorithm in order to prevent any negative concentration results avoiding artifact and leading to more reliable spectra, as shown in figure 1.

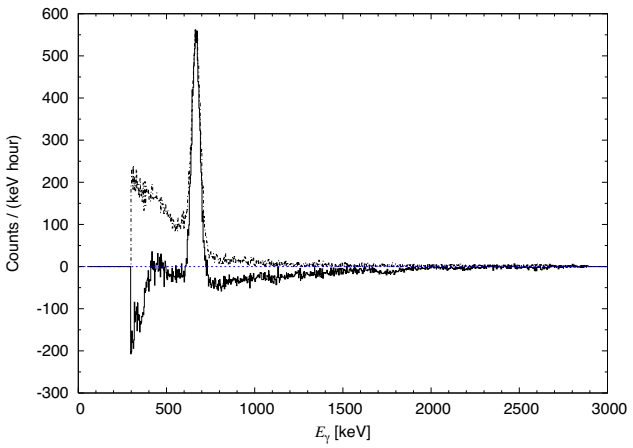

Fig. 1. The reconstructed sensitive spectrum for ^{137}Cs by using simple FSA (full line) and FSA+NNLS (dot dashed line). By implementing the NNLS constraint the non physical effects disappear.

The FSA+NNLS approach was tested on 400 sites where we both collected samples to be analyzed in laboratory, by using the MCA-RAD [4] system, and acquired γ spectra with a NaI(Tl) detector. By comparing the analysis performed in laboratory on the samples and the FSA+NNLS analysis we found results in excellent agreement.

In table 1 there are the correlation factors between the results of the laboratory analysis and three different analysis performed on the γ spectra acquired with the NaI(Tl) detector. We have used both the three windows method suggested by the IAEA and the FSA with or without the NNLS implementation and it is clearly visible how the correlation factors are more reliable by implementing the NNLS constraint. In addiction, we noticed that using the FSA+NNLS method we reduce the time of a measurement by at least a factor 2 with respect in the case of the three windows method.

Table 1. Correlation coefficients averaged for all the data samples. For the IAEA results the χ^2 is not shown due to the absence of a fit procedure.

	K	U	Th	χ^2
IAEA	1.12±0.07	1.11±0.10	1.00±0.09	—
FSA	0.99±0.06	0.78±0.14	0.86±0.07	1.22±0.08
NNLS opt.	1.06±0.06	0.87±0.12	0.94±0.07	1.06±0.05

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