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Spectral identification of a ^{90}Sr source in the presence of masking nuclides using Maximum-Likelihood deconvolution

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ABSTRACT

A technique for the spectral identification of strontium-90 is shown, utilising a Maximum-Likelihood deconvolution. Different deconvolution approaches are discussed and summarised. Based on the intensity distribution of the beta emission and Geant4 simulations, a combined response matrix is derived, tailored to the β^- detection process in sodium iodide detectors. It includes scattering effects and attenuation by applying a base material decomposition extracted from Geant4 simulations with a CAD model for a realistic detector system. Inversion results of measurements show the agreement between deconvolution and reconstruction. A detailed investigation with additional masking sources like ^{40}K, ^{226}Ra and ^{131}I shows that a contamination of strontium can be found in the presence of these nuisance sources. Identification algorithms for strontium are presented based on the derived technique. For the implementation of blind identification, an exemplary masking ratio is calculated.

1. Introduction

The last decade has seen several novel technologies for spectrum analysis, driven mostly by the evolving requirements of both nuclear security and safety. Challenged by a rising threat of nuclear terrorism, the security branch seeks for a variety of instruments that can be handled by more or less untrained personnel to perform different tasks of nuclear inspections. Additionally, with the recent events of the reactor incident at the Fukushima plant, Japan, a series of important safety considerations was brought back into the focus of the nuclear physics community.

Still, there is a high demand for reliable technologies that provide an automatised and correct threat announcement once radioactive emissions are detected. The decision whether a source is considered as threat or not is based on an investigation of the radiations isotopic composition, usually performed by analysing its spectroscopic fingerprints. As a rather complex procedure, the latter requires detailed knowledge of the associated spectra and is mostly mastered by experts specialised in this field. Of course, it is necessary to provide automated detection and identification tools to border securing officers, firefighters and police squads, as to mention just a few examples of user groups that cannot be trained to become experts in spectral analysis in addition to their own important set of skills. Here, it is crucial to map the expert knowledge into algorithmic schemes to assist the personnel with its tasks at hand.

For all these radiation analysis products the key idea is to deter the user from the issues and complexity of the detection and analysis process itself and to keep most of the physical core tasks, including the identification, hidden as a black box. Algorithms were developed to learn and identify sources based on their spectral shape and commercial detection products exist that are designed specifically for the identification of nuclear sources.

Furthermore, in the field of food and contamination analysis, those technologies play an increasingly relevant role. With the fallout from nuclear accidents affecting rural and farming areas dedicated to the cultivation of nutrition, it becomes necessary to carefully observe the world-wide cargo routes with specialised equipment. As of today, such evaluations are done by laboratories with dedicated instruments, food monitors, that are commonly fed with piecewise tests of food samples. Caused by the Fukushima incident, vast amounts of ^{137}Cs, ^{131}I, ^{132}Te and ^{90}Sr contaminated large rural areas around the plant. The short living isotopes as ^{131}I, having a half life of about 8 days or ^{132}Te with 3.2 days quickly vanished, leaving the long-living isotopes ^{137}Cs with 30 years and ^{90}Sr with 28 years as the main contributions.

But not all nuclides are equally important and a group of around forty sources can be regarded as highly relevant, nearly all of which feature a distinct γ-pattern. Modern equipment is more or less able to identify these sources and there are standards for nuclear security, Ref. [1] is just one example, specifying an exact list of nuclides that is desired to be found.

In nearly all scenarios stated above, the strontium isotope ^{90}Sr is an exception. After the first nuclear tests and their world wide

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fallout, a number of works [2,3] investigated its impact on biological material and way to determine its concentration in essential nutrition products like milk [4]. From the scintillation perspective, strontium yields a very decisive problem: it is a β^- emitter and when measured with e.g. a sodium iodide scintillation detector, it produces a continuous spectrum that has no characteristic peaks. For most algorithms this fact leads to the difficulty to identify ^{90}Sr correctly, especially if it is masked by nuisance isotopes. As a matter of fact, the identification of strontium in a simple, algorithmic way using low-cost equipment for its detection is very desirable and, as we already briefly discussed, many types of nuclear equipment could be significantly improved by it. Any subsequent threat categorisation [5] would benefit from such a solution.

In the course of this work, we will consequently focus on ^{90}Sr and present such an algorithm that identifies its continuous radiation even in the presence of masking sources. Threat isotopes emit γ-radiation with distinct peak pattern. More or less all algorithms somehow deduce their result from the peak pattern, either by locating the peaks and intensities or simply by matching the whole shape with the reference data. Without neglecting that germanium semi-conductor detectors provide probably the most accurate spectroscopic quality in terms of resolution and offer a high precision analysis of the radiation, our method is specifically designed to sodium iodide scintillator detectors, because they are a workhorse in the nuclear detection industry, cheaper and far more widely distributed than the high-resolving germanium.

We will describe a procedure that discovers the ^{90}Sr source based on response modelling and likelihood maximisation. Using the original distribution function of β^- emitters, we will construct a generalised response matrix, the so-called endpoint matrix that combines the unique features of the β^-.

The procedure will identify the continuous contributions of a β^- source in a bare and slightly shielded configuration and in the presence of other sources, explicitly considering the masking with strongly scattered sources. While we focus on the isotope ^{90}Sr, the method remains applicable for other β^- sources, too. We do not intend to distinguish between multiple β^- materials. Note that shielding has an enormous influence on the shape of the strontium spectrum, because its most characteristic counts are detected in the first hundred channels. Our model includes an average shielding that shows good results for the low and medium shielded cases.

Our document is structured as follows: after this short introduction we will present a brief summary of spectral deconvolution techniques in Section 2, beginning with their relevance in the context of other publications and concluding with the formal statement of the Maximum-Likelihood expectation maximisation (MLEM). As the latter algorithm requires the knowledge of the so-called response matrix, Section 3 will deal with the derivation of a β^- specific response model and its inclusion in the MLEM algorithm. The final deconvolution has been tested with ^{90}Sr and masking sources in Section 4. Section 5 gives an overview of the application of our deconvolution technique as identification algorithm for ^{90}Sr. Here, the unique characteristics of the deconvolved solution are used to establish a decision threshold. Finally Section 6 yields an outlook on upcoming applications and extensions of our algorithm.

2. Maximum-Likelihood deconvolution of γ-spectra

In this section we will concentrate on the question, how far the impact of certain physical interactions that define spectrum characteristics can in fact be mathematically reversed by a procedure which is called deconvolution or sometimes simply inversion. As the name already suggests, the nature of the interactions is restricted to systems where the complete answer function is produced by convolving responses of the subprocesses.

Original roots of inversion equations can be found in the prominent geological discipline of seismic exploration, where in 1954 Robinsion [6] presented a way for predictions based on the decomposition of time series, heavily inspired by the fundamental works by Wiener during that time.

Deconvolution is a longstanding topic in spectrum analysis and has also been discussed in the literature for quite a while. From the emission of radiation to the spectral acquisition, multiple physical processes like material absorption, the photo effect inside a detector or the Compton-scattering take place and for our following concept we assume that these steps can be reversed for a given source. Spectral deconvolution was indeed thoroughly reviewed by Bouchet [7] in 1995, comparing different deconvolution techniques and summarising their advantages and disadvantages. The conclusion of his work pointed already towards two rather similar algorithms, one based on an iterative maximisation of a likelihood estimator and the other applying an iterative entropy maximisation, both turning out to have significant potential for use in the spectrum analysis domain.

A-priori knowledge is the fundamental input of the deconvolution and in the context of spectrum deconvolution this is given in terms of the detectors response. The latter can be modelled by a matrix that relates the incoming energies (that hit the detector) with the output spectrum. A short illustrative introduction on these matrices is given in the next section.

In a rigorous application related approach, Meng and Ramsden [8] revisited three of these algorithms to investigate possible benefits for a virtual resolution improvement of low-cost scintillators like cesium iodide or sodium iodide. They explored the possibilities of symmetric detector concepts, leveraging the benefits of a homogenous response matrix and their processed spectra featured much sharper peaks, making the spectra easier to interpret by eye or by algorithmic means. As a main result, they provided evidence that the Maximum-Likelihood technique is superior to its alternatives and appears to be the method of choice for spectrum deconvolution tasks.

With the market requesting cheaper radiation detection equipment, development activities went towards plastic scintillators, having a remarkable sensitivity for radiation but unfortunately a very low intensity of the photo-peaks. For most of their energy ranges, they feature a purely continuous spectrum that is dominated by a smooth, edge-like structure produced by Compton-scattering. Response matrices were calculated for these extreme types of spectra and it is possible to virtually reconstruct the photo-peaks with astonishing quality, as e.g. discussed by Butchins et al. [9].

The deconvolution of plastic spectra shows in principle that continuous spectra and continuous responses are suitable to perform a stable inversion. Consequently it is also possible to unfold other continuous spectra, as in our case produced by β^- emitters, as well.

The way the deconvolution methods work can be easily understood as follows: the original spectrum of a radioactive source – as the experimental result – is a projection onto a given discrete channel space. Once a relationship between the incident energy and the out coming spectrum is established – mathematically this relationship is the response matrix – an inversion can be calculated that maps the measurement back on the incident energies. The decisive point is that, while the measurement as such always contains the information core, the deconvolution matrix and the subsequent inversion represent a defined scheme by which information is interpreted. In other words, the deconvolution reduces the solution space to the most-likely solutions which must be known a-priori for a given detector.

2.1. Construction of the detector response matrix

In previous works, response matrices for γ-detection have been successfully applied to various problems, as shown e.g.