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Automated quantitative analysis of in-situ NaI measured spectra in the marine environment using a wavelet-based smoothing technique

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ABSTRACT

An algorithm for automated analysis of in-situ NaI γ-ray spectra in the marine environment is presented. A standard wavelet denoising technique is implemented for obtaining a smoothed spectrum, while the stability of the energy spectrum is achieved by taking advantage of the permanent presence of two energy lines in the marine environment. The automated analysis provides peak detection, net area calculation, energy autocalibration, radionuclide identification and activity calculation. The results of the algorithm performance, presented for two different cases, show that analysis of short-term spectra with poor statistical information is considerably improved and that incorporation of further advancements could allow the use of the algorithm in early-warning marine radioactivity systems.

1. Introduction

Continuous monitoring of radioactivity in the marine environment is a major concern for the scientific community, since long-term measurements is a request in many applications of various research areas, such as development of natural resources (radon monitoring for quantification of groundwater discharges), meteorology (radon-progeny monitoring after rainfall for cloud identification), geophysics (radon monitoring as a precursor for seismic activity on submarine faults), radioprotection (nuclear security), radioecology (monitoring of natural radioactivity for industrial waste detection), etc.

In-situ radioactivity measurements in the marine environment using the γ-ray spectroscopy method are mainly performed using two detection systems: HPGe and NaI(Tl), see e.g. (Povinec et al., 1996; Osvath et al., 1999; Osvath and Povinec, 2001; Jones, 2001). The HPGe detectors have been used for quite a few applications in the marine environment during the last years, but they could only operate for a limited period of time (due to the limited holding time of cryostat-dewar) or in regions where power is supplied. The underwater NaI detectors are commonly used during the last decade in stationary marine networks (Wedekind et al., 1999; Tsabaris and Ballas, 2005; Osvath et al., 2005) for operational purposes due to their low cost, low consumption and fair applicability without cooling. However, the use of NaI detectors in marine monitoring and operational applications has specific limitations due to the following reasons: (a) ^{40}K introduces high radioactivity values in seawater ($\sim$12–13 kBq/m^3) and distributes the photons according to the Compton scattering mechanism at energies below 1461 keV, which means that a high natural background radiation (^{40}K) is present in sea; (b) NaI detectors exhibit a voltage drift of the system electronics due to temperature variations, so that a periodic energy calibration is needed for spectrum stabilization and reliable analysis; (c) NaI spectrometers are characterized by poor energy resolution. Moreover, the acquired spectra from such networks are transmitted every three hours and a time-consuming and laborious work is needed for further analysis and data reduction: the raw data concern the number of the detected gamma rays, so that full spectrum analysis has to be done in the laboratory with the aid of available user-dependent spectrum analysis packages. This is far from the desirable target of direct information for the level of radioactivity in the operational center.

Various efforts towards the direction of automated analysis of γ-ray spectra have been published in the last forty years, the main point being the automatic photopeak determination and isotope identification. Extensive literature as concerns the methodologies on automatic photopeak searching can be found in Silagadze (1996). An interactive program performing qualitative and quantitative isotope analyses using median estimates of peak areas in γ-ray spectra, obtained from semiconductor detectors, was presented by Kondrashov et al. (1993). A semi-automated

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isotope identification software, based on an interactive graphical method and working for various kinds of detectors, was also described in Brutscher et al. (2001). Recently, the performance of a nuclide algorithm for qualitative identification, based on a signal shape correlation technique and using a rather intuitive parameter input, has been evaluated through analysis of a considerable number of NaI(Tl) and HPGe modeled spectra (Russ, 2007). In any case, the development of a software for fully automated analysis of measured γ-ray spectra using NaI detection systems in the marine environment is an open issue.

Wavelets have been widely used for the last two decades as a powerful tool for signal analysis and modeling in engineering, biomedical and environmental fields. With regard to applications of γ-ray detectors, wavelet analysis techniques have been applied either to nuclear pulses, as e.g. for spectroscopy enhancement (Garcia-Belmonte et al., 1996), denoising purposes (Zhang et al., 2006) and, quite recently, pulse shape discrimination between beta and gamma rays (Yousefi and Lucchese, 2009) as well as between neutrons and gamma rays (Yousefi et al., 2009), or to the spectrum itself, as e.g. for exploring the quality of peak detection and localization for NaI spectra (Sullivan et al., 2006, 2007). This work, which belongs to the latter category, presents in detail an algorithm for automated analysis of in-situ γ-ray measured spectra as obtained from NaI detectors in the marine environment, using a wavelet denoising technique for smoothing the spectra. The algorithm incorporates an automated energy calibration in order to specify the energies for the detected photopeaks. The nuclide identification is performed through a database of nuclides often appearing in the marine environment. Implementation of the developed method provides both qualitative and quantitative user-independent information on the detected nuclides. The efficiency of the method is shown through two test cases of measured spectra obtained in a laboratory tank and at the open sea, using a NaI underwater measuring system.

2. Spectrum smoothing

Short-time in-situ NaI measurements in the marine environment provide spectra of highly irregular shape due to poor statistical information. The algorithm for automated analysis of the γ-ray spectra presented herein is based on a simple method, the success of which depends on the optimal fitting of a smooth curve to the original data of the irregular input spectra. A considerable number of traditional or more advanced methods exists for fitting data, such as classical or generalized (Savitzky–Golay) moving average filtering, various local regression smoothing methods, smoothing splines, etc. The reason of proposing a Wavelet-based Smoothing Technique (WST) is that its multiparametric nature allows smoothing along the whole energy range of the measured spectrum, providing a very efficient fit to the original data and, at the same time, a highly smoothed spectrum appropriate for straightforward automated analysis. Practically, through WST – widely used in applications for signal denoising – the original measurements for each channel are replaced with new, ‘‘denoised’’ values; that’s why WST is alternatively mentioned in the text as wavelet denoising technique. Before proceeding to the description of the algorithm, basic information on the tools of the wavelet approach used in this work will be given briefly: the discrete wavelet transform and the corresponding denoising process.

2.1. Discrete wavelet transform

A wavelet $\psi(t)$ is a waveform of effectively limited duration that has an average value of zero and tends to be irregular and asymmetric. The wavelet transform decomposes a function into a weighted sum of wavelets derived from scaling (dilation) and translation (shifting or positioning) of a single mother wavelet $\psi(t)$. Scaling allows the characterization of the frequency contents of the function, while translation enables the localization of different frequency contents. The wavelet transform of a function s based on discrete analysis, called Discrete Wavelet Transform (DWT), is the family of coefficients

$$C(p,q)=\int_R s(t)\frac{1}{\sqrt{p}}\psi\left(\frac{t-q}{p}\right)dt \quad (1)$$

where $p=2^j$, $j\in Z^2$ and $q=k2^j$, $(j,k)\in Z^2$, are the scale and translation parameters, respectively. The reconstruction process, called Inverse Discrete Wavelet Transform (IDWT), can be formulated as

$$s(t)=\sum_{j\in Z}\sum_{k\in Z}C(j,k)\psi_{j,k}(t). \quad (2)$$

Using DWT, the signal is decomposed into high-scale (low-frequency) components, called ‘approximations’, and low-scale (high-frequency) components, called ‘details’. The orthogonal, compactly supported wavelets are proper for discrete analysis by applying a fast decomposition and reconstruction algorithm for DWT (Mallat, 1989). Given a signal s of length N, the DWT consists of $\log_2 N$ steps at most. Starting from s, the first step produces two sets of coefficients: approximation coefficients and detail coefficients. These vectors are obtained by convolving s with a low-pass filter for approximation and a high-pass filter for detail, followed by dyadic downsampling. The decomposition procedure is iterated until a predetermined number of steps (levels) L, with successive approximations being decomposed in turn, so that the signal to be broken into many lower resolution components. Conversely, starting from the last pair of approximation and detail coefficients, the IDWT reconstructs the previous approximation coefficients, inverting the decomposition step by dyadic upsampling (inserting zeros) and convolving the results with the reconstruction low-pass and high-pass filters.

2.2. Wavelet denoising process

Smoothing of the original spectrum is performed by an automatic wavelet denoising process, using the MATLAB Wavelet Toolbox 4 (Misiti et al., 1997–2007). The underlying model for the original spectrum is of the form $s(n)=f(n)+\sigma e(n)$, where $f(n)$ is the smoothed spectrum, $e(n)$ is a Gaussian white noise with zero mean and unit variance, σ is the noise level, and variable n represents the equally-spaced channels. The smoothing procedure contains noise filtering and recovery of $f(n)$, and consists mainly of three stages:

(i) Computation of the wavelet decomposition of the spectrum s at a specific level L by selecting the mother wavelet and the level;
(ii) Application of a thresholding approach to the detail coefficients for each level, whereas only the portion of the details that exceed a certain limit is discarded.
(iii) Computation of the wavelet reconstruction, based on the original approximation coefficients of level L and the modified detail coefficients of levels from 1 to L.

3. The algorithm for automated spectrum analysis

The idea of an automated γ-ray spectrum analysis for NaI detectors in the marine environment is based on two main points: first, the capability of the wavelet denoising technique to produce a smooth-shaped version of the measured spectrum