Progress in radionuclide characterisation in marine sediments using an underwater gamma-ray spectrometer in 2π geometry

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Abstract

The in-situ gamma-ray spectrometry is a well suited method for seabed mapping applications, since it provides rapid results in a cost effective manner. Moreover, the in-situ method is preferable to the commonly applied laboratory measurements, due to its beneficial characteristics. Therefore, the development of in-situ systems for seabed measurements continuously grows. However, an efficiency calibration of the detection system is necessary for obtaining quantitative results in the full spectral range. In the present work, an approach for calculating the full-energy peak efficiency of an underwater in-situ spectrometer for measurements on the seabed is presented. The experimental work was performed at the coastal site of Vasilikos (Cyprus). The experimental full-energy peak efficiency of the in-situ was determined in the energy range 1400–2600 keV, by combining the in-situ and laboratory reference measurements. The experimental efficiency results were theoretically reproduced by means of Monte Carlo (MC) simulations, using the MCNP5 code.

Keywords: Targets Characterization, Ion Beams

1. Introduction

Radionuclide concentration measurements on marine sediments are traditionally performed using sampling techniques and spectrometric analysis in the laboratory. The widely applied method is the laboratory based gamma-ray spectrometry, since the majority of the natural and anthropogenic radionuclides can be detected from their gamma-ray lines. The laboratory method is not cost effective, since it requires manpower and instrumentation for sampling as well as laboratory facilities for the time-consuming procedures involved in the samples preparation. In addition, the laboratory method is applicable only at regions where the conventional grabbing techniques can successfully be employed (for instance non-rocky sea beds).

An alternative to the aforementioned difficulties, is the application of the in-situ gamma-ray spectrometry method. The benefits of the implementation of in-situ techniques for direct measurements on the seabed are well known, and a lot of effort has been devoted to the development of such detection systems [Jones, 2001]. During the last decades, several in-situ systems consisting of different types of detectors have been employed [Van Put 2004; Povinec et al., 1996; de Meijer et al., 2002] for aquatic and marine environment applications. Calibrations are carried out using appropriate laboratory facilities (e.g., special tank), reference calibration sources [Tsabaris et al. 2005] and Monte Carlo simulations [Maučec M., 2004; Vlastou et al., 2006; Bagatelas et al., 2010]. A review of the progress in developing techniques for in-situ measurements is summarized in the work of Jones [Jones 2001] and a recent updated status is also available [Thornton et al., 2013].

In this work an efficiency calibration of the in-situ system KATE [Tsabaris et al., 2008] was performed for measurements on the seabed in $2\pi$ geometry. The system was deployed to acquire spectra in close contact with a specific seabed (the $2\pi$ geometry) as well as in the seawater (the standard $4\pi$ geometry). Additionally, a sediment sample was collected from a site adjacent to the deployment and analyzed in the laboratory. The activity concentrations (Bq/kg dry weight) on the sample were measured at the laboratory by means of high resolution gamma-ray spectrometry. The experimental full-energy peak efficiency of the in-situ system was
calculated in energies covering the interval 1460–2600 keV, by combining the in-situ measurements with the laboratory activity reference measurements. The efficiency results for the specific seabed were subsequently reproduced by implementing the MC MCNP5 code.

2. Experimental Setup and Field Work

The experimental work was performed at the coastal site of Vasilikos (Cyprus). The in-situ detection system utilized in the present application for the gamma-ray measurements on the seabed was developed at the Hellenic Centre for Marine Research [Tsabaris C., et al., 2004, 2005, Tsabaris C., et al., 2008]. The efficiency calibration for measurements on the seabed was held directly on the field by deploying the system in the sea to acquire spectra. For the sediment measurement the detector was situated in close contact with the seafloor (2π geometry measurement) to assure that high counting rates from the radionuclides present in the sediment are recorded. The system was subsequently deployed for an additional measurement in the seawater (4π geometry measurement). For this background measurement the detector was positioned well above the seabed, thus recording events present only in the seawater in 4π geometry. This measurement was performed in order to estimate the contribution of radionuclides present in the seawater during the seabed measurement and properly subtract it. The contribution from each radionuclide is calculated by peak integration in the spectrum acquired in the 4π geometry measurement (seawater measurement). The calculated total count rate in each photopeak is multiplied by a factor of 1/2, and this quantity is subsequently subtracted from the count rate determined in the corresponding peaks of the sediment spectrum (2π geometry measurement).

Moreover, a sediment sample was collected from a site adjacent to the deployment. The grabbing technique was utilized for the sediment sample collection from the seafloor, thus a collection from a maximum depth of 15 cm was achieved. The collected sample was dried out and homogenized prior to the preparation of the final sample in the standard geometry for environmental measurements. The reference gamma-ray spectroscopy measurement was held at the facilities of the National Technical University of Athens (NTUA). Additionally, theoretical estimations were performed using the MCNP5 code to reproduce the experimental efficiency values.

In this work, the previously [Androulakaki et al., 2012] described methodology for the efficiency calculation at the energy 1460 keV (40K gamma-ray line), is expanded to the energies 1460–2600 keV. In this way, activity concentration calculations using the in-situ method are expanded, making possible the quantification of the radionuclides 208Tl (2614 keV) and 214Bi (1765 keV) present on the seabed.

3. Analysis and Results

The volumetric efficiency of the in-situ system for measurements on the seabed, $\epsilon_V$ (in units of lt), was deduced combining the in-situ acquired peak analysis data with the reference concentration activities (as deduced from the laboratory measurement in units of Bq/lt). Special attention was paid to employ proper analysis techniques in cases where in the analyzed peaks contributed more than one photopeaks attributed to radionuclides with neighboring gamma-ray lines (e.g. a total emission probability that equals the sum of the intensities of each gamma-ray line of the radionuclide 214Bi contributing to the peak at 1765 keV was considered). The efficiency results (simulated and experimental) are depicted in Fig. 1 as a function of the gamma-ray energies.

The experimental uncertainty was found 10% for the calculated efficiency at the energy of 2614 keV (208Tl) and 1461 keV (40K) and 20% at 1765 keV (214Bi). The observed uncertainties are attributed to the low activity concentrations of the studied area. The simulation results are in satisfactory agreement (less than 7% relative deviation) with the experimental ones within statistical uncertainties. The accuracy of the obtained results, render the simulations as a powerful tool to be utilized, especially for seabed applications where the variations in the seafloor properties influence the detectors efficiency. A theoretical study of the detection efficiency sensitivity to geophysical sediment parameters is thus considered, as the following step to the present work. Moreover, it is crucial to expand the efficiency calibration in the lower energy region between 300–1460 keV. In order to achieve this goal it is necessary to obtain more experimental data by
deploying the in-situ system in marine ecosystems containing Naturally Occurring Radioactive Materials (NORM). Future deployments are organized in such environments, in order to assure that high quality spectra are acquired.

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