Determination of marine gamma activity and study of the minimum detectable activity (MDA) in 4pi geometry based on Monte Carlo simulation

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Abstract Monte Carlo simulations were performed using the GEANT4 code for the investigation of γ-ray absorption in water in different spherical geometries and of the efficiency of a NaI(Tl) detector for different radionuclides in the aquatic environment. In order to test the reliability of these simulations, experimental values of the NaI(Tl) detector efficiency were deduced and seem to be in good agreement with the simulated ones. In addition, using the simulated efficiency, an algorithm was developed to determine the minimum detectable activity in becquerels per cubic meter in situ as a function of energy for typical freshwater and seawater spectra.

Keywords Marine gamma activity · Minimum detectable activity · 4pi geometry

Introduction

The existence of artificial and natural sources of radioactivity in the marine environment led to the growth of detection systems for the measurement of gamma radiation in water. The quantitative measurements of radioactivity in the aquatic environment demand innovative detection systems (concerning stability, electronics, autonomy), optimum housing constituents, precise calibrations using water tanks that imitate the aquatic environment, and validated codes for simulating the resulting γ-ray spectra. In situ detection systems for long-term aquatic measurements are very scarce due to the power consumption limitation of the systems, the choice of the optimum housing of the crystal, and the high background originating from Compton scattering of 1,461 keV (40K) and 2,615 keV (208Tl) γ-rays of natural constituents of water. NaI(Tl) are the most common crystals for long-term measurements due to their low consumption, good efficiency, and low cost (Povinec et al. 1996), but they have the disadvantage of low energy resolution. Such crystals are contemporarily used in many oceanographic applications, using continuous measurements for radiation monitoring with buoy operation and in situ measurements in seawater (Aakenes 1995; Soukissian et al. 1999; Wedekind et al. 1999; Baranov et al. 2003; Tsabaris and Ballas 2005; Osvath et al. 2005). A lot of effort has been made during the
past few years for the experimental calibration of detection systems in water tanks by diluting calibrated standard sources (Vojtyla 2001; van Put et al. 2004; Tsabaris et al. 2008).

The main problems of the marine radioactivity measurements concern the determination of the detection efficiency and of the minimum detectable activity (MDA) for estimating the smallest amount of a radionuclide’s activity that can be reliably determined. Various methods have been developed for the determination of the MDA or the detection limit (Currie 1968; Aničin and Yap 1987; Currie 1995; Shi et al. 2005), and existing methods have been modified for in situ measurements (Nir-El and Haquin 2001). However, it is difficult to determine the detection efficiencies in the marine environment for all γ-ray energies experimentally, since there are a rather limited number of single-energy, gamma-emitting radionuclides that are soluble in water.

Monte Carlo simulations have been implemented during the last decade in order to study the γ-ray detector response features and efficiency calibration for different geometries and to validate improved performances of new detectors mainly at low energies (Ghanem 2000; Karamanis 2003; Crespin et al. 2004; Fayez and Al-Ghorable 2006; Hurtado et al. 2004). The Monte Carlo technique was also applied for the calculation of theoretically simulated spectra at a required detection position at a given source distribution on the ground (Srinivasan et al. 2001) and for airborne in situ gamma ray measurements (Klušon 2001). Such simulations have also been developed for underwater measurements (Vojtyla 2001; Vlachos and Tsabaris 2005; Vlastou et al. 2006; Mertens et al. 2007), as well as for substrate characterization (Ocone et al. 2004), but those at the open sea marine efficiency calibration in 4pi geometry and studies of the MDA in the whole energy interval are still pending.

In order to determine the efficiency for each γ-ray energy, in the present work, a Monte Carlo simulation was performed using the GEANT4 code for studying the photon interaction in water in different spherical geometries (to ensure 4pi emission) and for calculating the detection efficiency for different radionuclides in the marine environment. In addition, using the simulated efficiency, an algorithm was developed to determine MDA in becquerels per cubic meter as a function of energy, utilizing the Currie method (Currie 1968) modified to account for the particular requirements of the present work. Finally, MDA values were determined for typical freshwater and seawater spectra.

Description of the experimental and simulated setup

System description

For the in situ monitoring of radioactivity in the marine environment, a new 7.5 × 7.5-cm NaI(Tl) scintillator detector, coupled with a photomultiplier and integrated electronics, was developed at the Hellenic Centre for Marine Research. It is an autonomous system with relatively low power consumption, operational on oceanographic buoys and seafloor platforms. The design and special characteristics of the developed system are described elsewhere (Tsabaris et al. 2008). The detection system had to be tested and calibrated in the laboratory, in a water tank, before its deployment in the marine environment. The sensor was mounted in the middle of the water tank, of 5.5 m³ volume, and appropriate radionuclides (99mTc, 137Cs, and 40K) were diluted for the calibration of the detection system. The layout of the water tank, along with the schematic geometry of the detector, is shown in Fig. 1.

Simulation code description

GEANT4 is a Monte Carlo code, which simulates the trajectory of elementary particles through matter. It simulates the tracking of particles, like γ-rays, through an experimental setup for the study of the detector response. It also provides the graphical representation of the setup and of the particle trajectories (CERN 1993). Detailed descriptions of the geometry of the experimental setup with respect to their dimensions, materials, and shapes, as well as of the particle generator, are required by the program to simulate and store