Monte Carlo simulation of a NaI detector in the aquatic environment

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Abstract

   NaI(Tl) crystals are used in many marine applications for continuous measurements with buoy operation and autonomous in situ measurements in seawater. Monte Carlo simulations were performed using the GEANT4 code for the investigation of the γ-ray absorption in water in different spherical geometries and for the efficiency of a NaI(Tl) detector of 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 using a special tank filled with water and reference single gamma ray sources (^{99m}Tc, ^{137}Cs and ^{40}K). The cascade reference source ^{111}In was also diluted in tank for comparison with the reproduction spectra of its cascade lines as provided with the GEANT4 code. The results are in good agreement with the simulated ones within uncertainties.

Keywords: Monte Carlo simulation; GEANT4 code; marine efficiency

1. Introduction

   The quantitative measurements of radioactivity in the aquatic environment is a complicated task since it demands robust underwater system, stable electronics, optimum housing material, precise calibrations. *In situ* detection systems for long-term aquatic measurements are very scarce due to the power consumption limitation of the systems and to the high background originating from Compton scattering of 1461 keV (^{40}K) and 2615 keV (^{208}Tl) γ-rays. NaI(Tl) are the most common crystals for long term measurements due to their low consumption, good efficiency and low cost [1], 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 [2-6]. A lot of effort has been made during the last years for the experimental calibration of detection systems in water tanks, by diluting calibrated standard sources [7-9].

   The detection efficiency for aquatic environments can not be determined for all γ-ray energies experimentally, but only using specific gamma ray reference sources and simulation codes. 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 and for calculating the detection efficiency for different radionuclides in the marine environment.

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In an attempt to increase the confidence of the spectra analysis and produce more reliable results in the concentration of low-level environmental radioactivity, simulated spectra of ^{99m}Tc, ^{137}Cs and ^{111}In were compared with real data recorded by the detector in a water tank. In addition the comparison between the measured spectra and the simulated ones could control any possible performance deterioration of the submersible detection system at an early stage.

2. Description of the experimental and simulated setup

2.1. 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 seabed platforms. The design and special characteristics of the developed system are described elsewhere [9]. The detection system had to be tested and calibrated in the laboratory, in a water tank, before its deployment in the marine environment. The layouts of the water tank, along with the schematic geometry of the detector are shown in Figure 1.
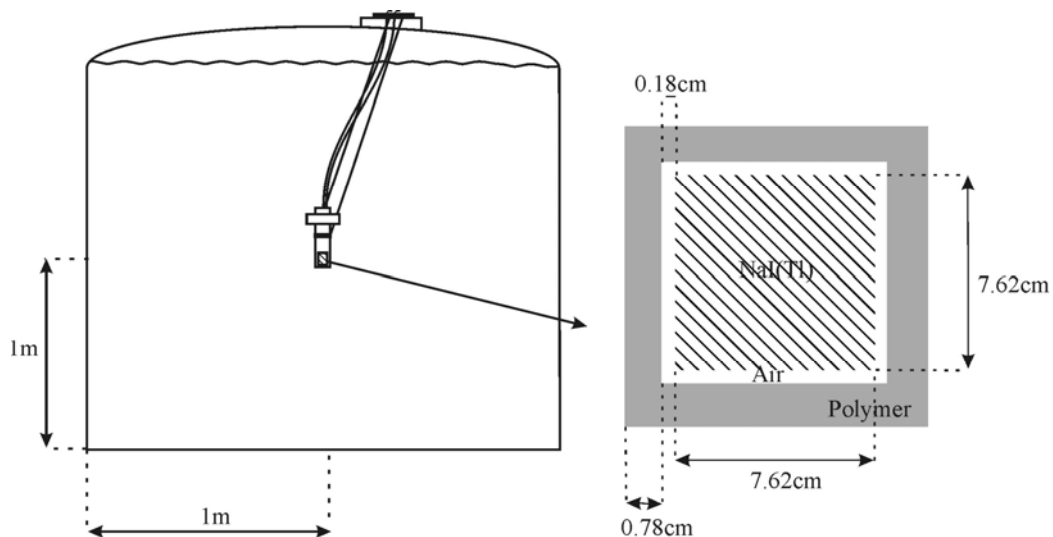

Fig. 1. Layout of the water tank along with the NaI(Tl) detector geometry, used both for the simulations and experimental calibration [11].

2.2. 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 [10]. 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 the history of each particle from its generation to full deposition of its energy in the detector. The definition of the parameters, which control the Monte Carlo simulation, is of particular importance for the quality of the results. The specific details and characteristics of the code as implemented in a NaI(Tl) system have been extensively described in the past [11].

3. The GEANT4 code simulations

The detection system is designated to measure γ-ray radioactivity in the marine environment. The aim of this work is to accurately convert the photopeak counts corresponding to each radionuclide in the recorded spectrum, to specific activity in Bq/m^3. The specific activity, r, is given by the Equation (1):

$$r(Bq/m^3) = \frac{CPS}{\varepsilon \cdot V \cdot I_\gamma} \tag{1}$$

Where CPS denotes counts per second recorded for the specific radionuclide, ε is the photopeak efficiency at the specific energy, V the volume of water for each γ-ray and I_γ is the emission probability of the specific γ-ray. In order to use Eq. (1) in a global, consistent and reliable way in the case of marine measurements, the product (ε·V), which subsequently is denoted as "marine efficiency" ε_m (in m^3), has to be defined for all γ-ray energies.

The corresponding spherical volume, V_{eff}, surrounding the detector, beyond which γ-rays have practically zero probability to reach and interact with the detector crystal will be defined as the "effective volume" for the specific γ-ray energy.

3.1. Marine efficiency

The calculation of the NaI(Tl) detector efficiency for each γ-ray energy depends strongly on the volume of water mass. Gamma rays of different energies present different absorption in water so that the detector efficiency varies for different effective water volumes. In an attempt to quantitatively study this effect, simulations have been carried out via the GEANT4 code for different radionuclides diluted in different water volumes surrounding the NaI(Tl) detector. As input, three γ-rays, namely 140.5 (^{99m}Tc), 661.6 (^{137}Cs) and 1460.6 keV (^{40}K) were used to produce simulated spectra via the GEANT4 code for various spherical water volumes. For each water volume, the density of counts (counts/m^3) was kept constant, so that the variation of photopeak counts could be studied as a function of water volume. It was observed that the photopeak counts are increasing with volume and exhibit a saturated value for water volumes greater than the effective one. In the case of ^{137}Cs (661.6 keV) the results are shown in Figure 2 along with the fitting curve. The marine efficiency, ε_m, was deduced by dividing the saturated counts with the total number of counts in each spectrum and by multiplying with the respective volume. It was thus also observed that the marine efficiency remains constant for volumes larger than the effective volume.
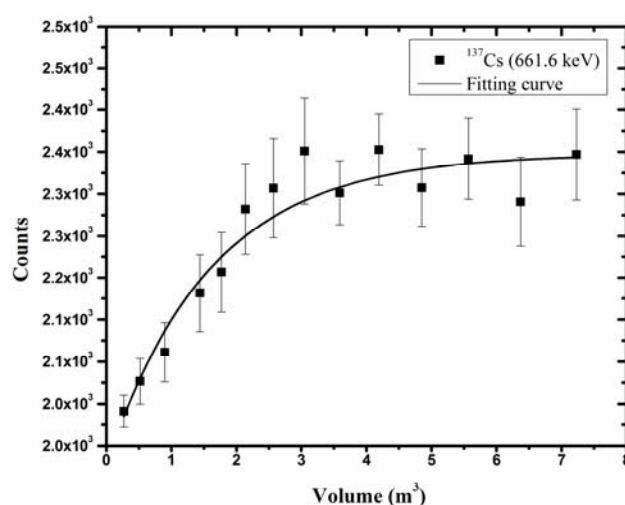

Fig. 2. Graphic representation of detected counts in the 661 keV photopeak of the simulated spectrum, associated to the volume of water with diluted ^{137}Cs. The solid line represents an exponential fitting curve of the simulated values.

In order to calculate the marine efficiency, ε_m, of the NaI(Tl) detector in the aquatic environment as a function of energy, simulated spectra were generated for the energy range between 100 and 2000 keV. The results are shown in Figure 3 and the error bars in the figure correspond to the statistics of the simulated spectra. The solid line represents the marine efficiency curve, which fits the simulated values in water and is given by the empirical expression [12]:

$$\varepsilon_m = \frac{a \cdot E^b}{c + E^d} \qquad\qquad (2)$$

Where α, b, c and d are fitted parameters and E is the energy of the specific γ-ray.
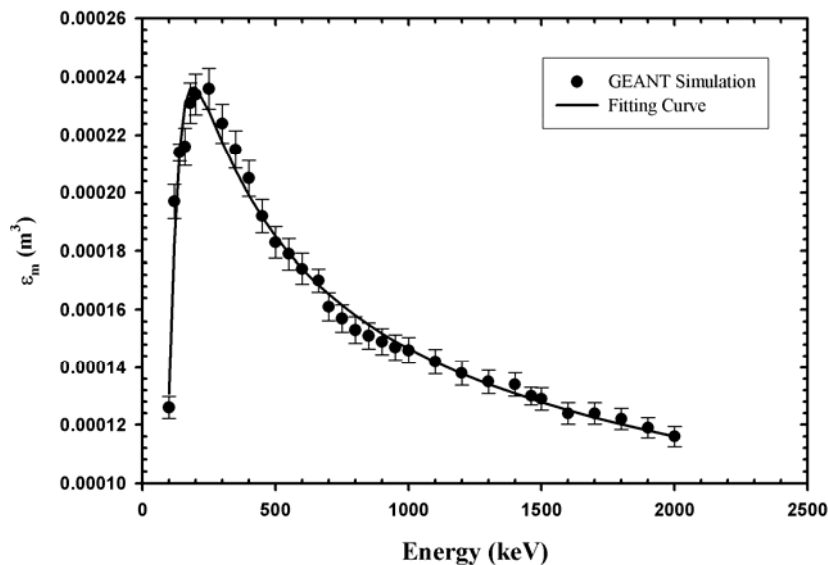

Fig. 3. Simulated marine efficiency values of the NaI(Tl) detector in the aquatic environment along with the fitting curve (solid line) from Eq. (2).

Experimental values [9] of the NaI(Tl) detector marine efficiency, ε_m, (extracted for the 140.5, 661.6 and 1460.6 keV transitions of ^{99m}Tc, ^{137}Cs and ^{40}K, respectively) were deduced and seem to be in good agreement with the simulated ones. The experimental and simulated marine efficiency values seem to agree within the 10% uncertainty values. These results indicate that the marine efficiency of the NaI(Tl) spectrometer as simulated by the GEANT4 code, could be used over the full energy range for the determination of the volumetric activity of radionuclides emitting non cascade γ-rays in water.

3.2. Experimental and simulated spectra

In the present work simulated spectra of ^{99m}Tc (140.5 keV), ^{137}Cs (661.6 keV) and ^{111}In (172 keV and 246 keV) were reproduced and compared with the equivalents experimental that were recorded by the detector in a water tank. More specifically, the sensor was mounted in the middle of the water tank, of 5.5 m^3 volume and appropriate radionuclides (^{99m}Tc, ^{137}Cs and ^{111}In) were diluted. The recorded spectra (after the subtraction of the corresponding background spectrum) were used for the comparison with the simulated ones. A rebinning algorithm has been applied to the experimental spectra before the fitting procedure. In following figures (Fig. 4a-c) the experimental spectra are shown, along with the simulated ones.
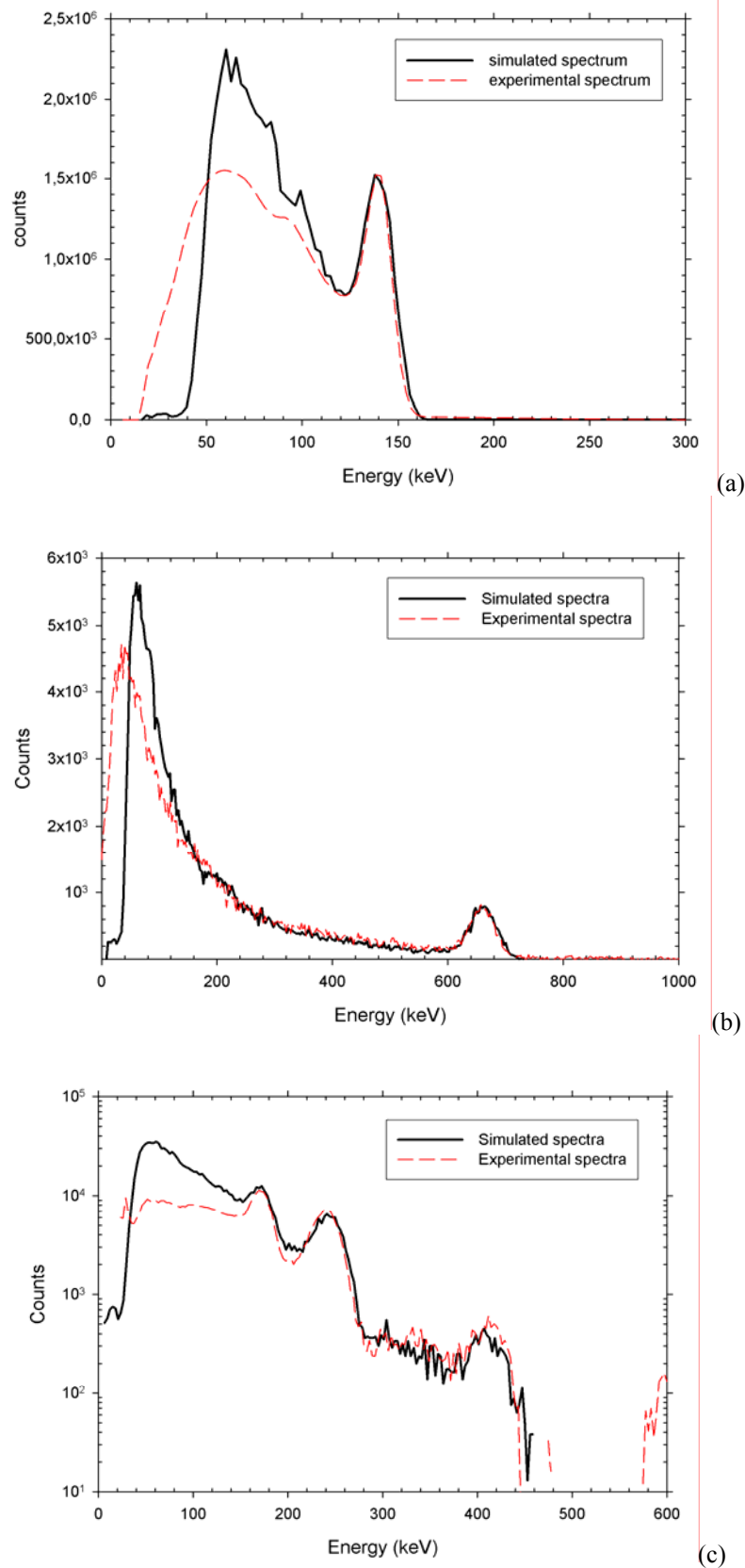
(a)

(b)

(c)

Fig. 4. Simulated and real spectra, acquired by the NaI(Tl) detector in a water tank with diluted (a) ^{99m}Tc, (b) ^{137}Cs and (c) ^{111}In (the background spectrum has been subtracted from the experimental spectra).

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4. Discussion of the results

The GEANT4 code successfully describes the detection process for qualitative, as well as, quantitative marine radioactivity calculation using the NaI(Tl) detection system and produces reliable γ-ray spectra of natural radionuclides collected by a NaI(Tl) detector immersed in an aquatic environment. The simulated spectra can be used to calculate the marine efficiency, ε_m, in a wide energy range and thus to determine the volumetric activity in Bq/m^3 for γ-emitter radionuclides.

It must be emphasized that the calibration factors for the marine efficiency are valid only in the case of single energy γ-emitting radionuclides. In the case of cascade energy γ-emitting radionuclides the results are not appropriate since summing effects should also be taken into account.

The simulated spectra for ^{99m}Tc, ^{137}Cs and ^{111}In agree well with the experimental ones except in the lowest energy region. More specifically, above 180 keV simulated and experimental spectra coincide, but in the low-energy region (<180 keV) the simulation overestimates the experimental spectra. In the case of cascade reference source ^{111}In the simulated and experimental spectrum are in good agreement. So the simulation code reproduces with grate accuracy summing effects from cascade energy γ-emitting radionuclides.

5. Conclusions

A method for improving the accuracy of the determination of radioactivity in the marine environment has been developed, aiming at its integration on geophysical applications, as well as, at real-time data-forwarding buoy applications. The marine efficiency of a NaI(Tl) detector was investigated experimentally and with computer simulations using the GEANT4 code. It was proven that the code provides quantitatively accurate results and is suitable for marine applications. Thus, the simulations constitute a useful tool, especially in the case where the experimental setup for the detector calibration cannot effectively imitate the marine environment, for the investigation of problems of γ-ray absorption in water and solid angle calculations that are present for *in situ* marine measurements.

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