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In situ gamma-ray measurements of marine sediment using Monte Carlo simulation

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Abstract

This work outlines the progress in developing a new method for in situ radioactivity measurements of marine sediments. The method combines the underwater gamma-ray spectrometer (a system named KATERINA based on a NaI(Tl) detector) with Monte-Carlo calculations using the MCNP5 code. This method aims at allowing for an accurate quantitative determination of activity concentrations in marine sediments (using the in situ system), which can be applied in different areas and for variable sediment structures.

As a first step, the MCNP5 code has been successfully applied for the standard 4π geometry in the aquatic environment, reproducing results of the marine efficiency as previously deduced by the GEANT4 code. The experimental set up geometry was introduced in MCNP5 using detailed information for the geometry and the materials. Moreover, a first simulated estimation of the in situ efficiency for sediment measurements is presented for ^{40}K (1460.8 keV). For this purpose a new model was constructed taking into account a typical experimental geometry set-up (with the detector being situated in close contact with the seabed). In order to validate the Monte-Carlo results, activity measurements were also performed in sediment samples collected from Basilica, Cyprus, where the in situ system was deployed. The samples were analysed using a HPGe detector for inter-calibration purposes and the obtained results are discussed.

Keywords: MCNP5; underwater gamma-ray spectrometer;

1. Introduction

Activity concentrations in sediments are correlated with various geological, geochemical and oceanographic phenomena, such as sedimentation rates, radionuclide's bioaccumulation in marine organisms, groundwater submarine diffusions, hydrocarbon manifestations on the seabed and radionuclide re-suspension in the water column. Most of the radionuclides are determined in sediment samples using the laboratory gamma-ray spectrometry method and appropriate calibration extended sources [1, 2].

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The determination of activity concentrations in marine sediments with the in situ method is of great interest, since it provides immediate measured data on a temporal and spatial basis. Till now there are few underwater sensors for radioactivity measurements of the seabed. During the last decades the *in-situ* gamma-ray spectroscopy was introduced for seabed mapping either, by detection systems capable to acquire at a specific energy window or by more sophisticated instruments capable of full gamma-ray spectroscopy [3, 4, 5].

The aim of the present work is to present a preliminary study of the absolute photo-peak efficiency calculation of the *in-situ* spectrometer for the 2π geometry measurement in marine sediment. The efficiency is validated at 1460.8 keV by comparing the experimental value for [40]K (for the 2π geometry) with the theoretical computations as deduced using the Monte Carlo (MC) code MCNP5.

2. Materials and method

Activity concentration measurements have been performed at the coastal site of Basilica (Cyprus) close to a former fertilizer industry. The underwater gamma-ray spectrometer KATERINA has been deployed *in-situ* both, on the seabed using 2π geometry (the detector was placed in close contact with the seabed) and, in the seawater using the standard 4π geometry. The spectrometer was deployed in six different sites (sites 1, 2, 3, 4, 7, 8) in 2π geometry as shown in figure 1. Moreover, a seawater spectrum was acquired in site 1 using the standard 4π geometry. The acquisition live time was 72000 seconds in sites 1, 3, 8 and 10800 seconds in sites 2, 4, 7. The acquisition live time of the seawater spectrum was 62000 sec. The seawater spectrum was considered as a background spectrum and it is properly subtracted from the sediment spectrum.

Furthermore, marine sediments from the same position of the *in-situ* deployment have been collected dried out and measured in the laboratory. Wet mass, dry mass, and volume were measured for all samples before measurement.
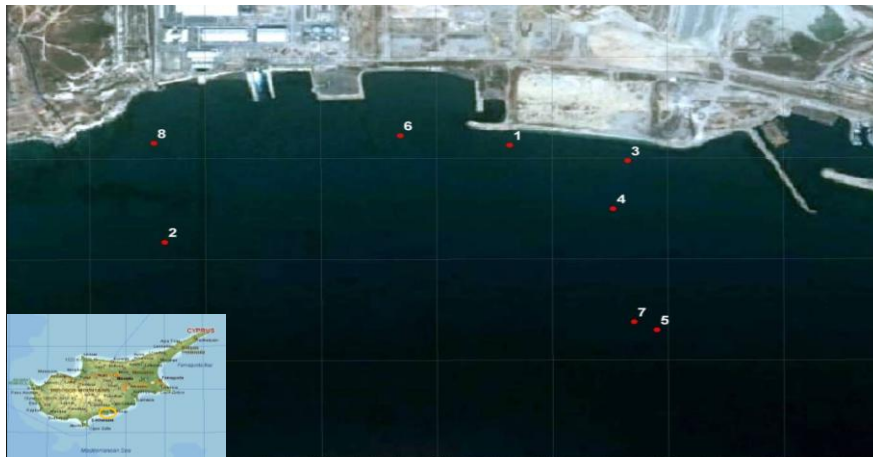

Fig. 1. Basilica coastal zone, Cyprus. Study area - Map of sampling points.

Activity concentrations (dry weight) were determined in laboratory using a 50% relative efficiency HPGe detector. The calculated activities A_{ref} (Bq/L) were used as a reference value

for the experimental estimation of the full-energy peak efficiency for the *in-situ* sediment measurement using the following formula (Eq 1):

$$\varepsilon_V = \frac{\left(cps_s - \frac{1}{2} * cps_w\right)}{I_\gamma * (A_{ref})} \bigg|_{HPGe}^{KATERINA} \qquad (1),$$

where cps_s is the counts per second of the *in-situ* measurement for the sediment, cps_w is the counts per second from the seawater spectrum, I_γ is the gamma line emission probability and ε_V is the photo-peak efficiency (in units of volume) of ^{40}K (at 1460.8 keV) for the 2π geometry.

Monte Carlo simulations are performed in order to validate the experimental results. The MCNP5 model for the *in-situ* measurements was developed under the following assumptions for all sites :

- A sediment composition based on mean fraction values of chemical compounds found with the XRF method for site 1 was kept constant in all sites.
- The experimentally determined water content (%) for site 1, was inserted in the Monte Carlo code for all other sites.
- The experimentally determined wet bulk density for site 1 was inserted in all computation runs.
- A volume source of homogenized material is considered.
- A different effective volume for each radionuclide is considered according to the energy of the emitted gamma-ray. The effective volume is calculated from the total mass attenuation coefficient μ_t which is considered for water saturated sediments as the sum of the mass weighted coefficients of the solid and fluid constituents [6]. The total mass attenuation coefficients were determined for each gamma-ray energy from the web database NIST X-COM.

3. Results - Discussion

The laboratory analysis of the dried sediment samples exhibited low radioactivity levels. The activity concentrations (dry weight) varied between 143 – 160 Bq/L, 5 – 6.5 Bq/L, for ^{40}K and radon progenies (^{214}Pb, ^{214}Bi), respectively. ^{226}Ra concentrations were calculated below 8 Bq/L in all sites.
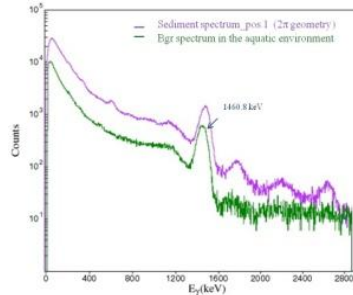

Fig. 2. KATERINA spectra for site

The obtained spectra from the KATERINA detection system for sediment as well as for seawater are presented in the figure 2 (site 1). The two spectra (seawater-4π and marine sediment-2π geometry) exhibit different radionuclide concentrations ([214]Pb, [214]Bi, [228]Ac, [208]Tl) due to the natural constituents of the sediment material. The most evident contribution into the seawater spectrum is the photo-peak of the radionuclide [40]K (at 1460.8 keV) due to the salinity of the sea.

A detailed MCNP5 model was developed using the accurate detector dimensions, as given by the manufacturer. This model successfully reproduced the efficiency results at energies above 300 keV according to previous estimations of the marine efficiency [7] using the GEANT4 code. Moreover, the model was applied for simulating the experimental spectra of [137]Cs (661.7 keV) and [40]K (1460.8 keV). The experimental spectra were acquired using the *in-situ* spectrometer, reference sources and a water tank in a controlled laboratory environment [8]. The results are shown in the following figure:
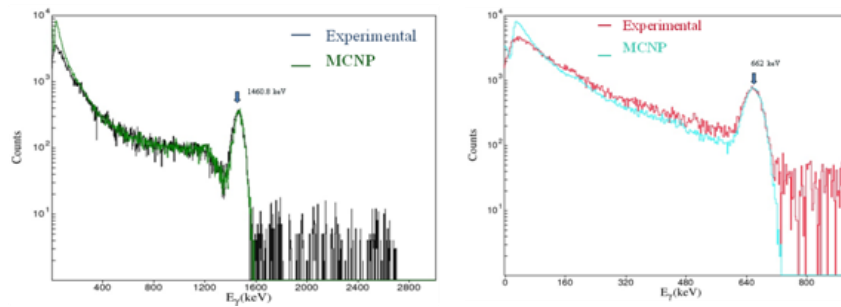

Fig. 3. Comparison of experimental with theoretical (MCNP5) spectra for [40]K (left) and [137]Cs (right).

The next step, following the validation of the MCNP5 model in the aquatic environment (4π geometry), was to perform appropriate adjustments in the code for estimating the system efficiency in 2π geometry for measurements in the marine sediments. Laboratory results for the wet bulk density, the porosity, and the chemical composition of the collected sediment sample in site 1, where included in the description of the source in the MC code. The final MC model for the 2π geometry shown in figure 4 consists of the detector model, sediment source and surrounding environment.
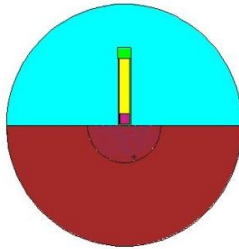

Fig. 4. MC model for 2π geometry

The theoretical value for the absolute photo-peak efficiency in marine sediment was found 0.0286 L, with acceptable statistical precision [9] of 5.2% (% relative error). The experimental efficiency calculation using Eq.1 resulted a value of (0.027 ± 0.002) L for [40]K in site 1. The comparison between the experimental and theoretical results showed satisfactory

agreement within 6%. After the validation of the theoretical model in 2geometry for site 1, the derived MC efficiency value for ^{40}K was further used for the *in-situ* activity concentration calculations of the seabed at the sites 2, 3, 4, 7 and 8 (see Figure 1). The *in-situ* values are compared with those obtained by means of a laboratory based HPGe detector for the aforementioned sites in Table 1.

Table 1

Comparison of in situ and lab results for activity concentrations (^{40}K) in marine sediments

	Site 1 (Bq/L)	Site 2 (Bq/L)	Site 3 (Bq/L)	Site 4 (Bq/L)	Site 7 (Bq/L)	Site 8 (Bq/L)
Lab						
	158.4 ± 9.4	149.1 ± 13.0	151.6 ± 10.5	143.0 ± 9.8	145.4 ± 10.0	154.9 ± 9.2
In - situ						
	148.8 ± 10.5	133.9 ± 12.9	174.1 ± 12.3	134.7 ± 11.1	146.2 ± 12.4	188.8 ± 13.3
deviation (%)	-6.5	-11.4	14.8	-6.1	0.5	21.9

A mean deviation of 9 % was found between laboratory and in-situ results. The maximum discrepancies are observed in sites 3 and 8 (15 and 22 %, respectively). These discrepancies (in sites 3 and 8) could be attributed to the assumption that the ^{40}K concentration of the seawater does not vary in the studied region. The deployment of the *in-situ* system in the 4π geometry should be considered as an additional measurement for every sediment measurement (in 2π geometry). The aforementioned discrepancies can also be attributed to the assumptions made for the theoretical estimations (as explained analytically in a previous section). For instance, sediment characteristics such as water content, and wet/dry density can differ significantly in the same region [10]. These parameters should be specifically determined in advance before being introduced in the MC code in a systematic way. The estimated efficiency is not generalized for other seabeds due to the different sediment characteristics (e.g. different size of grains, mineralogy).

4. Conclusions

A new approach for *in-situ* quantitative marine sediment measurements has been presented. The importance of obtaining more experimental data from regions with different geological formations is evident. Future work involves such measurements for methodology generalizations over a wide range of energies.

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