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Radioactivity measurements in the aquatic environment using in-situ and laboratory gamma-ray spectrometry

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HIGHLIGHTS

- In-situ underwater gamma-ray spectrometry method is validated by the lab method.
- MC simulations using MCNP5 reproduced experimental energy spectra and efficiency.
- MDA of the in-situ method was an order of magnitude lower than the lab method.
- The in-situ method was sensitive and cost-effective compared to the lab method.

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ABSTRACT

The in-situ underwater gamma-ray spectrometry method is validated by inter-comparison with laboratory method. Deployments of the spectrometer KATERINA on a submarine spring and laboratory measurements of water samples with HPGe detector were performed. Efficiency calibrations, Monte Carlo simulations and the Minimum Detectable Activity (MDA) estimations were realized. MDAs varied from 0.19 to 10.4 (lab) and 0.05 to 0.35 (in-situ) Bq/L, while activity concentrations differed from 7% (for radon progenies) up to 10% (for ^{40}K), between the two methods.

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1. Introduction

Monitoring of radionuclides in the marine environment is of significant importance, since human activities (e.g. mineral exploration, mining, production of electrical energy) may lead to accidental or intentional dumping of nuclear radioactive materials in the sea, resulting in enhanced levels of radioactivity (Matishov and Matishov, 2004). Furthermore, natural radionuclides (mainly radium and radon isotopes) are used as tracers in hydrogeological applications such as the investigation of submarine groundwater discharges (Burnett et al., 2006; Schubert et al., 2006, 2008) and geophysical studies such as monitoring of radioactive gasses in groundwater (Tsabaris et al., 2011) as well as in hydrocarbon bearing zones (Gadallah et al., 2010).

Marine radioactivity measurements are performed mainly by two approaches (a) sample collection/grabbing in combination with on-site or laboratory measurements and (b) in-situ instrumentation in direct contact with the environment under study. The laboratory method is widely applied using the standard gamma-ray spectrometry for complex geometries between source and detector (Saegusa et al., 2000; Venturini and Vanin, 1993). The in-situ approach, has reached a high level of analytical performance during the last decade, mainly using the underwater gamma-ray spectrometry (Povinec et al., 2006, 2008; Tsabaris et al., 2010, 2012). This approach is also combined with stationary floating platforms for real-time data transmission (Aakenes, 1995; Wedekind et al., 1999; Tsabaris and Ballas, 2005; Tsabaris, 2008a).

Two kinds of detection systems have been mainly utilized for in-situ gamma-ray applications, those based on High Purity Germanium (HPGe) semiconductor crystals and those based on

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NaI(Tl) scintillators (Povinec et al., 1996). Despite their superior resolution, HPGe systems exhibit several limitations for autonomous operation, concerning high power consumption (100–300 W) and short acquisition period (~4 h) of deployment due to the cooling necessity (with liquid nitrogen). On the contrary, NaI (Tl) detection systems exhibit higher efficiency and much lower consumption (~2 W), which render them suitable for prolonged underwater deployments. However, the disadvantages of the NaI(Tl) systems compared to HPGe ones are their poor energy resolution and the voltage drifts of the amplification signal, which can take place during the measurement, producing overlapping peaks.

The most important task using an underwater spectrometer is the calculation of the system's detection efficiency. 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 (Vojtyla, 2001; van Put et al., 2004; Tsabaris et al., 2008b). Recently, experimental gamma-ray spectra in water were compared with Monte Carlo simulations using the GEANT4 code (Vlastou et al., 2006). A methodology for the determination of the detection efficiency of the in-situ system in water (marine efficiency ε_m), based on experimental measurements and simulation data, is described in Bagatelas et al., 2010.

The calculation of the detection efficiency for extended samples using HPGe detectors is still a complex task, since calibration sources should be prepared with composition and density similar to the real samples', and should be measured in the same geometry as well. The experimental determination of absolute photo-peak efficiency can be performed only in specific energies according to the reference calibration source. The detection efficiency is calculated in a wide range of energies (not only at the energies of the reference sources), by fitting the experimental data with standard mathematical functions from literature (Debertin and Helemer, 1988; Dias et al., 2004). Alternatively, appropriate simulation codes (e.g. MCNP5 (X-5 Monte Carlo Team, 2003), GEANT4 (GEANT4 Collaboration, 2003), PENELOPE (Salvat et al., 2006) and FLUKA (Ferrari et al., 2005)), may provide absolute photo-peak efficiency estimations at each required energy and for any geometry. Among the simulation codes, the MCNP and GEANT codes have been applied to gamma-ray spectroscopy yielding a satisfactory agreement between experimental and simulated data (Rodenas et al., 2000; Karamanis et al., 2002; Saegusa et al., 2004; Lépy et al., 2010).

In this work, an inter-comparison exercise has been realized, comparing results of the two methods (in-situ and laboratory), aiming at testing the performance of the in-situ underwater gamma-ray spectrometer KATERINA (Tsabaris et al., 2008b) – the abbreviation comes from the Greek words Innovative Sensor for Artificial and Natural Radioactivity (Tsabaris et al., 2008c) – under realistic conditions. For this purpose, a method was developed for the direct measurement of water samples by an HPGe detector under two geometries, as described in detail below.

2. Materials and methods

2.1. Study area and field work

The inter-comparison exercise was performed using water masses (in-situ) and water samples (lab) from the same marine environment, namely a submarine groundwater discharge located in the Bay of Stoupa, (southwestern Greece). The groundwater is emanated from fissures in the bedrock at a depth of 25 m with high flow rates (several decades cm/s) (Tsabaris et al., 2012) forming an aggressive environment. Several deployments of the in-situ system and a number of sample collections were performed by divers during the period from July 2009 till May 2010, covering a full hydrological cycle of the submarine spring. The system KATERINA was positioned and secured immediately above the spring, where the crystal position was approximately 2 m above the seafloor and 23 m below the sea surface. The grabbing of the water samples took place exactly from the same point, whenever this was feasible. To avoid radon gas losses and ground-seawater exchange, the sample containers should be closed immediately after grabbing. The containers should be made from material durable to withstand depth pressures and with low radiation absorption for the efficient detection of gamma-rays. To meet the requirements, cylindrical bottles with frustum ending (1/4 of the total height) made from heavy-duty borosilicate glass were selected. Both bottles were 7 mm thick with round opening (20 mm radius) and had diameters of 86 and 126 mm, total heights of 160 and 240 mm and volumes of 0.6 and 2.2 L, respectively. The samples were transported to the laboratory the day after each sampling (to reduce the ^{214}Bi and ^{214}Pb concentration losses due to ^{222}Rn decay ($T_{1/2}$=3.82 d)) and measured directly with an HPGe detector without any pre-treatment or pre-concentration. Decay corrections were performed for the radon progenies in order to calculate their activity concentrations at the sampling date and time.

2.2. In-situ method

The deployments were performed using the autonomous in-situ underwater gamma-ray spectrometer KATERINA. Initially, the measuring system was energy calibrated and tested for its stability with respect to temperature variations and energy resolution. The calibrations were carried out inside a tank having a volume of 5.5 m^3 filled with freshwater (Tsabaris et al., 2008b).

The specific activity r, in Bq/L, is given by the equation

$$r(\mathrm{Bq}/L) = \frac{CPS}{\varepsilon_m I_\gamma} \quad (1)$$

where CPS denotes counts per second recorded for each photo-peak, I_γ the gamma-ray emission intensity and ε_m the marine photo-peak efficiency, namely the product (εV) of the detector efficiency ε and the effective volume of water V that is measured for each gamma-ray.

The marine photo-peak efficiency ε_m and the Minimum Detectable Activity (MDA) of the system have been calculated and presented in detail elsewhere (Bagatelas et al., 2010). The MDA calculations for five main radionuclides as derived from laboratory measurements in the water tank for a 24 h acquisition time are presented in Table 2.

2.3. Laboratory method

2.3.1. Experimental set up

All laboratory measurements were carried out at the facilities of Hellenic Centre for Marine Research (HCMR). The detection system consists of a p-type coaxial HPGe detector (ORTEC GEM-FX8530P4) with 50% nominal relative efficiency and with a resolution of 2.15 keV at 1.33 MeV along with a computerized MCA system (CANBERRA Model 8715) for the data acquisition. A lead shielding (58.5 mm thick and 21 mm height) surrounded the detector in order to reduce the ambient gamma-ray background.

The calibration of the HPGe system for two extended geometries was performed, following a similar methodology, as described in Tsabaris et al., 2007. Two radioactive reference sources of Europium ^{152}Eu/^{154}Eu were produced with the same

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