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R. VLASTOU, C.A. KALFAS

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Set up and application of an underwater γ -ray spectrometer for radioactivity measurements

**C. TSABARIS¹, D. S. VLACHOS¹, C. T. PAPADOPOULOS², R. VLASTOU²
and C.A. KALFAS³**

¹Hellenic Centre for Marine Research
P.O. Box 712, 19013 Anavissos, Greece

²National Technical University of Athens
GR-15780 Athens, Greece

³National Centre for Scientific Research 'Demokritos' (NCSR)
Institute of Nuclear Physics, GR-15310, Greece

e-mail: tsabaris@ath.ncmr.gr

Abstract

The set up and control of an underwater measuring instrument for radioactivity pollution in the marine environment is described. The detection system is based on a NaI scintillator (RADAM III) with modifications for use in the marine environment with on-line measurements. The system is simple, has low power consumption and is stable for long-term monitoring (10 months). Before its deployment, the sensor was calibrated in the laboratory in a tank full of water to reproduce the marine environment. The calibrations were performed, by detecting the 661keV and 1461 keV gamma rays of known activity liquid sources ¹³⁷Cs and ⁴⁰K, respectively. The measured spectra in the laboratory were compared with spectra from a similar detector as acquired in the field. The analysis of the parallel measurement gave satisfactory agreement for the concentration of the potassium (⁴⁰K), as calculated from the salinity in the seawater, thus enabling the system for quantitative measurement of the seawater radioactivity.

Keywords: Underwater sensors; Continuous monitoring; Radioactivity.

Introduction

Marine radiometric techniques, which have been widely used during the last years, are based on the measurement of α β or γ -ray emission of the various radionuclides involved, which may occur from natural and

anthropogenic sources. The emission of natural radionuclides comprises the gamma ray emission of ⁴⁰K, which is a natural constituent of the seawater, and the decay series of ²³⁸U, ²³⁵U and ²³²Th. The main measurable gamma ray spectrum emitted by the anthropogenic radionuclides is due to the following isotopes:

$^{137,134}\text{Cs}$, $^{95}\text{Zr}/^{95}\text{Nb}$, ^{106}Ru and ^{60}Co . Similar gamma ray measurements of radionuclides in the low energy region (< 100 keV) present many difficulties, due to the increase of the photoelectric probability for total absorption of the gamma rays in the seawater and the detector housing. In situ measurements exhibit many advantages (POVINEC *et al.*, 1996 & JONES, 2001) over laboratory analysis, which demands the use of sensitive radioanalytical methods like preconcentration, chemical separation methods as well as long-term measurements in low background counting systems. Some of such measurements have been carried out in the Aegean Sea (FLOROU *et al.*, 1994) and the Eastern Mediterranean Sea (MALANOTTE-RIZZOLI, *et al.*, 1989 & PAPUCCI *et al.*, 1999). This technique, however, does not allow monitoring on a continuous basis or evaluation of the variation of radioactivity level in time and space.

The development of an autonomous measurement system for radioactivity in the water environment is of important scientific priority for the marine sciences (FLEMMING, N.C., 1995). Such a system involves important demands in constructing sensors, which produce reliable measurements for long time intervals, as well as in designing a communication system for on-line transmission of the data. Special attention has also to be given to the tolerance of the system, to cope with the impact of extreme waves in the marine environment, and to the power supply, which is needed for the operation of the central control system and the sensors.

This paper focuses on the improvement and application of an existing gamma ray detection system (RADAM III) for radioactivity measurements in seawater. The set up of the system is described together with the appropriate calibrations and the resulting measurements are presented and discussed.

Materials and Methods

The Hellenic Centre for Marine Research (HCMR) owns and maintains RADAM III

sensors constructed by the Norwegian Company OCEANOR. This system consists of a 3"x3" NaI detection crystal with a built in photomultiplier tube, preamplifier and power supply, together with the electronics for data acquisition, storage and transmission. The electronics modules are miniaturized to fit inside the sensor housing (80x60mm) and the power consumption is very small ($\sim 1\text{W}$). The operating temperature ranges between -10 and $+50^\circ\text{C}$ and its influence on the gain shift of the detector is compensated automatically with thermistor-based hardware. The detection unit maintains the same sensitivity and detection limit as in laboratory analysis using NaI scintillators. Thus, the efficiency and energy resolution are sufficient for alarm or monitoring purposes, provided that the energy of the gamma rays from the various involved radionuclides does not overlap. This type of sensor can be used for operational purposes by attaching it on to oceanographic buoys and can operate under severe weather conditions and especially with extreme waves driven by wind forces and high temperature gradients.

In order to use this system for continuous monitoring the sensor has been energy calibrated and tested for its stability in temperature variations and its energy resolution. Measurements of the detector efficiency and absolute calibration in Bq/m^3 have also been performed. For this purpose, a calibration tank of 5.5m^3 volume, filled with water has been used. The sensor was mounted in the middle of the tank in order to be surrounded by one meter of seawater, which is enough to simulate the high attenuation of the γ -rays in seawater. At the bottom of the tank, an electric pump was used to circulate the seawater, to mix the appropriate radionuclides (^{137}Cs and ^{40}K) with the water and to get homogenous conditions.

At the same time, a parallel measurement was performed with a similar system installed on an Oceanographic buoy in the North Aegean Sea. The detector was placed in a depth of 3m and the measured data were transmitted every 6h to the operational center

of HCMR via Satellite Communication and mobile telephony network (NITTIS, *et al.*, 2000).

Results and Discussion

The sensor was energy calibrated by using point sources of ^{137}Cs and ^{22}Na and then placed in the tank filled with fresh water. Background spectra were collected in order to systematically investigate the natural radioactivity and to test for temperature stability. A typical spectrum is presented in Fig.1, where the photopeaks from the decay series of ^{238}U are clearly seen.

The γ -rays used to perform absolute calibration of the system were the 661 keV of ^{137}Cs and the 1461 keV of ^{40}K with an elemental abundance 0.012% in natural potassium. More specifically, 1000gr natural

KCl were diluted in the tank resulting in $(3025 \pm 55) \text{ Bq/m}^3$ specific activity. Several spectra have been recorded and were found to be identical, indicating the homogeneity of the solution. One of them is shown in Fig.2, where the 1461 keV γ -ray from the disintegration of ^{40}K is illustrated. Furthermore, liquid ^{137}Cs of $490 \pm 25 \text{ Bq/m}^3$ activity was mixed in the water together with 65% HNO_3 0.005N. One of the acquired spectra is presented in Fig.2. The 661keV transition of ^{137}Cs is observed above the high background produced both by the natural background and the Compton scattering of the 1461 keV γ -ray of ^{40}K . To extract the pure 661 keV photopeak, shown as dashed line in Fig.2, the two spectra have been subtracted. The analysis of the spectra has been performed with the 'SPECTRG' software package (KALFAS, 2000). By integrating the net 661 and 1461 keV photopeaks, the

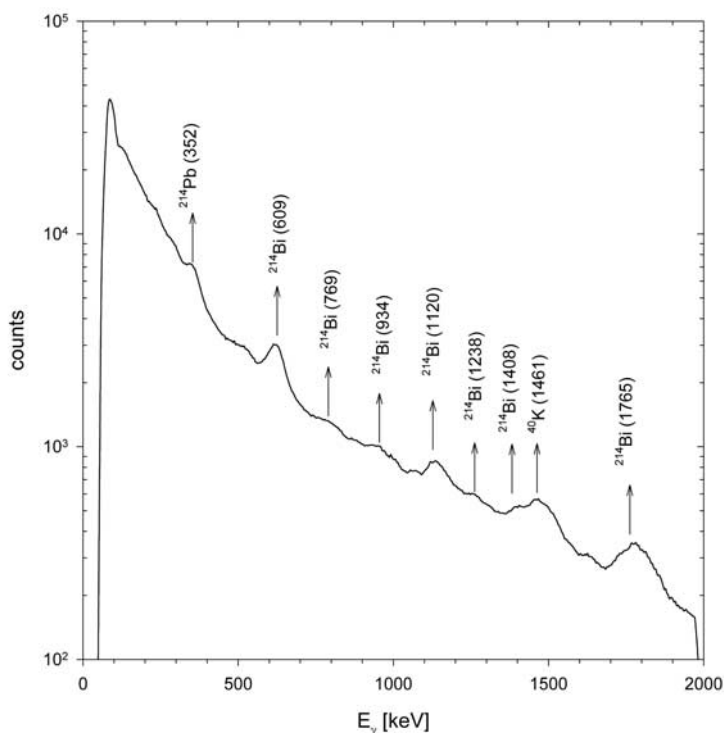


Fig. 1: Background spectrum as acquired with the RADAM III system (3 days). The energy and origin of the individual photopeaks are indicated in the Figure.

efficiency of the detector has been deduced as follows:

$$\begin{aligned} \text{Absolute total efficiency (661keV)} \\ = (3.4 \pm 0.2) \times 10^{-5} \end{aligned}$$

$$\begin{aligned} \text{Absolute total efficiency (1461 keV)} \\ = (2.4 \pm 0.1) \times 10^{-5}. \end{aligned}$$

In addition, the specific activity R in Bq/m^3 corresponding to N (counts/sec) in the photopeak of the γ -ray has been calculated to be:

$$R \text{ for } ^{40}\text{K} (\text{Bq/m}^3) = 7.1 \times 10^4 \times N (\text{counts/sec})$$

$$R \text{ for } ^{137}\text{Cs} (\text{Bq/m}^3) = 6.4 \times 10^3 \times N (\text{counts/sec})$$

The error has been estimated to be $\pm 5\%$ and is mainly due to the uncertainty in the activity of the radioactive source.

A parallel measurement has also been performed using the Real-time technique in

real marine environment. A similar RADAM III system was also calibrated and installed on a buoy at the North Aegean Sea. The spectra were recorded and transmitted to the operational center of HCMR via satellite communication. A typical spectrum is depicted in Figure 3, where the contribution of the characteristic 1461keV γ -ray of ^{40}K in the entire energy interval is clearly shown (Compton edge, Compton tail and photopeak). By analyzing the photopeak and using the absolute calibration in Bq/m^3 , the concentration of natural potassium in the seawater has been extracted as $(427 \pm 20) \text{ gr/m}^3$, corresponding to a salinity $(38 \pm 2) \text{ psu}$. This result verifies salinity measurements in the same region, which vary between 35 and 38.

In order to improve the use of the RADAM III spectrometer in seawater, a suitable technique will be developed for the

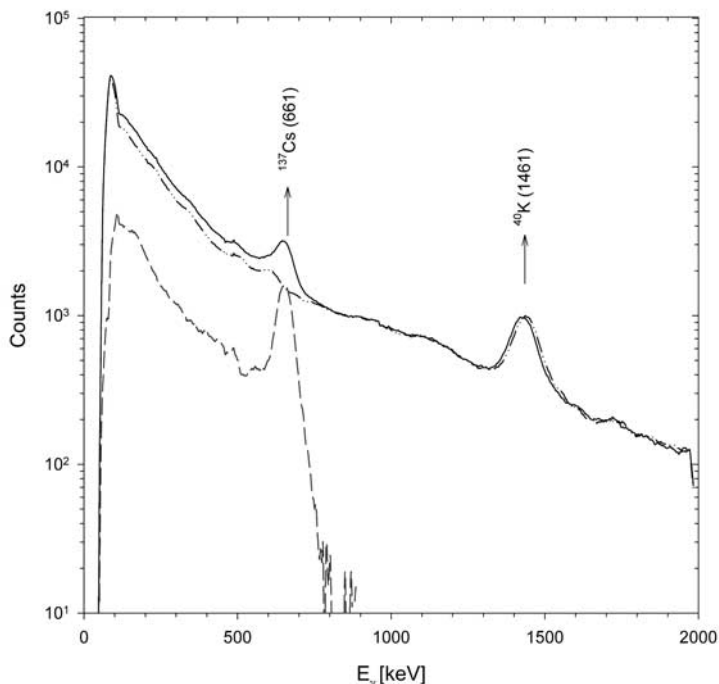


Fig. 2: Gamma ray spectrum recorded in the laboratory (3 days) by mixing in drinking water KCl (dashed double dot line), and adding known activity concentration of ^{137}Cs (solid line). The dashed line represents the pure ^{137}Cs spectrum deduced by subtracting the previous two spectra.

calculation of its response function. The idea is that, before simulating the detecting mechanisms of gamma spectrometer, one has to calculate the energy distribution of photons in seawater (primary gamma photons undergoing multiple scattering events before they interact with the sensing device) since they have a significant contribution in the measured spectrum. In addition, the development and application of this technique provides the possibility to improve the lower limit of detectability of the system and thus to identify the volumetric activity (Bq/m^3) of anthropogenic radionuclides in seawater. The next step in the folding procedure is to calculate the energy deposited (sensed) by a photon that enters the sensor by taking into account the three interaction mechanisms: Photoelectric effect, Compton scattering and Pair production.

Conclusions

The RADAM III NaI scintillator system for underwater gamma ray spectrometry has been tested and calibrated in the laboratory for radioactivity pollution monitoring in marine environment. The absolute efficiency and calibration in Bq/m^3 of the detector have been determined by using ^{137}Cs and ^{40}K radionuclides of known activity.

In order to test the deduced values of absolute calibration, parallel measurements with a similar detection system installed on a buoy in the North Aegean Sea, have also been performed. By analyzing the recorded data, the concentration of natural potassium in seawater has been extracted and found to be in good agreement with values given in the literature.

The poor energy resolution of the system makes it incapable of precise peak

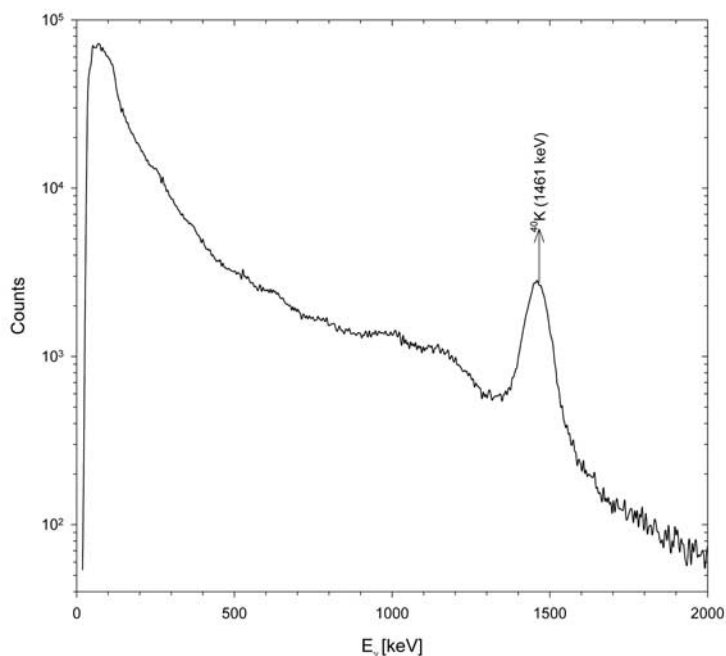


Fig. 3: Gamma ray spectrum acquired in the field by using similar detection system and the real-time technique. The measuring time is 3 days.

identification of the various gamma ray contributions as produced from the natural constituents of the water. This problem will be solved by estimating theoretically the γ -ray spectra using Monte Carlo techniques and taking into account all the responsible processes and interactions of gamma rays in the water as well as in the material of the detector. The simulated results will be validated by the experimental measured efficiency, when the underwater system is acquiring in a depth of at least 1m.

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