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The development and application of an underwater γ-spectrometer in the marine environment

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Abstract. The development and application of a new detection system is described for autonomously working radioactivity measurements, usable in seawater and river environment. The system is based on a NaI scintillator with specifications for use in the marine environment and for real time acquisition. It is simple, stable for long-term monitoring, and of low consumption. The sensor was energy and efficiency calibrated in the laboratory. Many tests were made for the linearity and the stability of the electronics. The energy resolution calibration of the sensor was performed prior to its location into the tank, using various reference point sources. The system was deployed in open sea in order to measure background and low volumetric activity of ^{137}Cs (19 Bq/m^3). The field measurements in the Aegean Sea offer very promising results concerning the use of the whole system in the marine environment to be used as continuous monitoring and alarm system.

1. INTRODUCTION

The automatic water monitoring networks were developed in order to measure artificial and natural constituents of the seawater. For that purpose, they have to be fully autonomous, with low power consumption, using real time data transfer, and capable of measuring low level of volumetric activity (a few Bq/m^3). In general, the measurement of radioactivity in seawater introduces many difficulties due to following constraints:

a. the sensitivity of the detection system has to be very high due to the dilution factor of the sea. The detection of low-level artificial radioactivity (like ^{137}Cs) becomes difficult due to the Compton scattering of the natural radiation of ^{40}K (about 12500 Bq/m^3 in the North Aegean Sea).
b. the system is placed in the harsh environment of an open sea (sea worthy design and construction) operating in real time data transfer mode.
c. additional measuring sensors should be connected to the system in order to cover a wide range of scientific interests (pH, temperature, salinity and rainfall-meter).
d. maintenance intervals should be at least six months.
e. the detectors/sensors has to be attached on an anchored buoy.

Additional improvements will be needed for allowing:

a. corrections due to voltage drifts of the detector's signal
b. qualitative identification of the participating radionuclides into the measurement (Software development).
c. very long counting time.

Hellenic Centre for Marine Research owns and maintains the POSEIDON buoy network. Radioactivity sensors [1] are installed on respective buoys in three sites at the Aegean Sea [2]. The operation of these systems encountered various problems concerning the stability of the system, the sensitivity, the energy resolution, the dead time and the long-term counting measurements.

In the present work a new developed system is presented, named KA-TE-RINA. The system was tested in the Aegean Sea for a short period. The specifications of the system as well as the improvements for optimal operation are described below.

2. MATERIALS AND METHODS

2.1 System description

A new system for underwater measurements and continuous monitoring has been developed. It monitors gamma ray radioactivity in the energy range from threshold to 2800 keV. The radioactivity sensor of the system consists of a 3"x3" NaI detection crystal, with built-in photomultiplier tube and a preamplifier (developed by Canberra). A separate power supply, together with an electronic card for data acquisition and storage is used. A watertight enclosure, which resides the sensor and the electronics, is also developed. The power consumption of the whole system is relatively low (~1.5 W). The influence of the operating temperature (between 0 and +50^{o}C) to the gain shift of the detector is compensated automatically. The electronics have the following specification for the shaping and amplification of the preamplifier signal (see Table 1). The data acquisition and storage device will be described elsewhere [3].

Table 1. The specifications of the shaping amplifier.

operating voltage	12-18V
shaping method	CR-(RC)^2
shaping time constant	500ns
Input	unipolar (negative or positive)
input impedance	100Ω
gain min	10db
gain max	100db
output dynamic range	80% of Vs into 1k load
Noise	<900μV rms at maximum gain
non linearity	<0.08%

The developed watertight cylindrical enclosure houses the above mentioned sensor unit and the electronic card (see Fig. 1). Following specifications were defined for the enclosure, prior to design:

- Capable to offer free of error continuous sensor functionality up to one hundred meter of water depth.
- (Testing depth of 200 meter).
- Corresponding plugable watertight cabling systems for real time data transmission. Redundant power transmission to/form the sea- surface.
- A lifespan for the enclosure and the cabling system in the deep sea, for at least 5 years.
- Maximum gamma ray permeability through the enclosure.

The thermoplastic material "Acetal" (commercially known as Celcon, Delrin etc.) has been chosen in order to have the lowest photoelectric absorption. The material has excellent resistance in the sea environment, is also known for its very good mechanical strength properties, and it is good machineable.

The wall thickness of the cylindrical enclosure has been calculated using Roark's thick wall formulas [4]. In order to take into account the creeping behavior of the thermoplastic material, additional calculations have been performed [5]. A conservative working strength of 0.9 Ksi has been assumed for Acetal. Similar calculations were performed for enclosure's cup and the bottom geometry. Two radial and one axial Buna N (nitrile) O-rings have been used in order to achieve a very good sealing mechanism despite the frequent reopening of the enclosure (at least during the testing phase).

The calculation of O-rings' parameters (diameter, thickness, material, hardness, groove dimensions, maximum gap etc.) was performed, following the suggestions of the Seal Design Guide from Apple Rubber Products Inc. The enclosure's 3D artistic picture can be seen in Fig. 1. For easy mounting of the sensorial, the unit (sensor + photomultiplier + electronic card) is mounted on a support (sled). The sled is furthermore, permanently fixed with bolts, on the enclosure's cup. A watertight connector- and cabling system was designed according to the specifications mentioned above.

2.2 Calibrations and set up

In order to use this system for continuous monitoring the sensor has been energy calibrated and tested for its stability to temperature variations and its energy resolution. Particularly, the energy and energy resolution calibration has been performed in the lab by using five reference γ-ray sources. In addition, underwater measurements of the detector efficiency and absolute calibration have also been made. A calibration tank of $5.5m^3$ volume, filled with water has been used for this purpose. The sensor was mounted in the middle of the tank, surrounded by at least one meter of water, which is enough to imitate the real marine environment for energies less than 700 keV. At the tank bottom an electric pump was placed in order to circulate the water. The water circulation assures: a) sedimentation avoidance b) good water mixing -with the appropriate reference radionuclide (^{137}Cs)- and c) homogenous conditions.

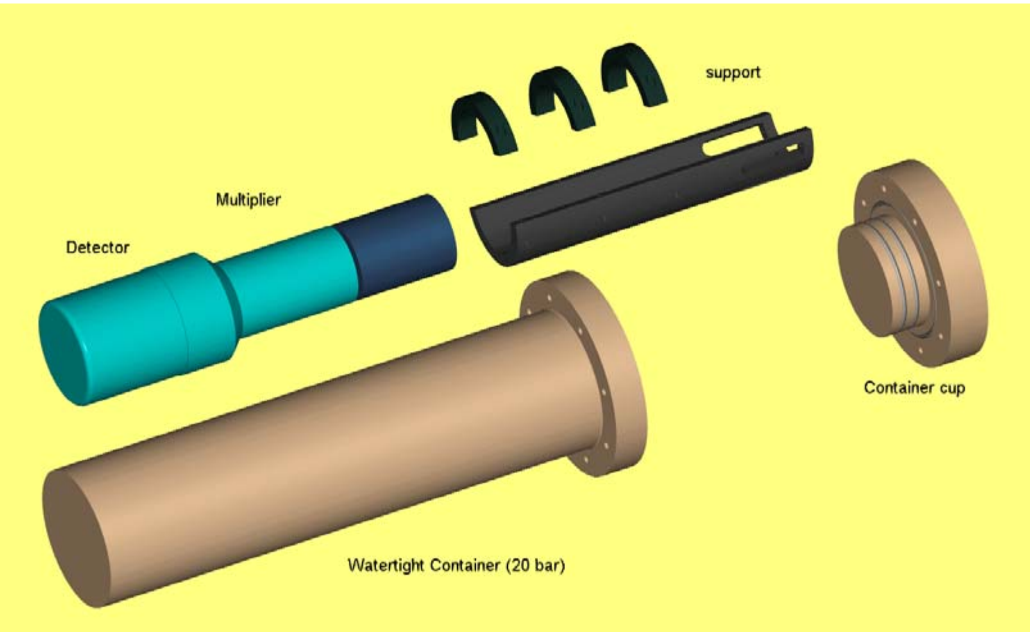

Figure 1. The watertight cylindrical enclosure, which houses the sensor, the photomultiplier and the appropriate sled.

The energy calibration and the efficiency calibration of the system for underwater use, has been calculated by using two reference gamma ray sources: ^{40}K and ^{137}Cs. The used γ-rays for the 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. In specific, l000 gr natural KCl were diluted in the tank resulting in (3025±55) Bq/m^3 volumetric activity. Several spectra were recorded and were found to be identical, indicating thus the homogeneity of the solution. Furthermore, liquid ^{137}Cs of (320±21) Bq/m^3 volumetric activity was mixed with water together with 65% HNO_3 0.005N [6].

In February 2004 KA-TE-RINA was placed in a special holder and was deployed in a depth of 3m using the HCMR's Research Vessel "Aegeao". Even the instant site-coordinates of the detector were those of ship's drifting motions, this did not affect the whole measurement. It can be assumed that for unchanged weather conditions in the short measurement period and in specific area, the seawater homogeneity is also ensured. Salinity data were also simultaneously measured at the same depth by means of an on board CTD. The efficiency at 1461 keV gamma ray energy at the open sea has been calculated using the measured salinity values. The developed system has been tested furthermore, in five different sites in the Aegean Sea. The site coordinates are given in Table 2.

Table 2. The site coordinates in the Aegean Sea. The measurement depth was 3m.

Position	Deep Basin 3	Deep Basin 2	LESVOS	Deep Basin 6	Varkiza-Athens
Latitude	39 02.23	40 07.01	39 18.29	37 42.54	37 42.21
Longitute	24 39.11	24 32.62	25 39.69	25 24.85	23 48.68

It is intended to embed the developed system, in the near future, on a specific oceanographic buoy [7]. Using appropriate facilities and previous experience, the measured data will then be transmitted to the operational centre of HCMR. The communication link uses either mobile (GSM) or Iridium satellite telephony.

3. RESULTS-DISCUSSION

3.1 Validation of the system

First, the NaI sensor was set up using the PCI-slot card (ASA 100) developed from Canberra Company for NaI detectors. The optimum operating voltage was 1000 V and the amplification gain was adjusted for detecting gamma rays until 2800 keV. A typical spectrum is shown in Fig. 2, measured in the laboratory using a ^{60}Co reference source.

The system was then tested, after being fully developed (NaI detector + new electronics + watertight enclosure) for continuous monitoring measurements in the marine environment. Similar measurements were performed using same radioactive source for validating the device. The derived spectrum is shown in Fig. 2. The comparison of the two spectra is very satisfactory, in the entire energy range. Both spectra have been energy calibrated first, using two photopeak energies of ^{60}Co reference source and presuming the linearity of electronics.

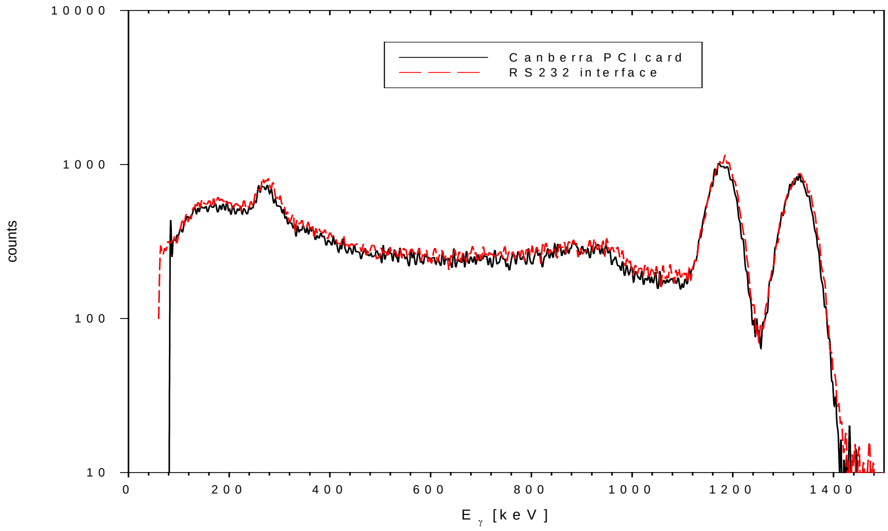

Figure 2. The measured spectra as acquired by the Canberra PCI card as well as by the developed electronics with RS232 communication. The two measurements were performed in the same geometry by using the ^{60}Co reference source and the measuring time was 100 s.

3.2 Calibrations

The sensor was deployed in a tank filled with drinking water. In order to investigate the variation of the natural radioactivity and to test for temperature stability background spectra were systematically analysed. The gamma ray emission from the decay series of ^{238}U can be clearly seen in the measured background spectra. Typical background spectrum is presented in [8] accompanied with a study for the gas loss of ^{222}Rn. During the first month the variation of the background counts was significant, since the half-life of ^{222}Rn is only 3.825 days. In order to have a constant background spectrum, the reference sources were mixed into the water after passing of a month from the experiment setting.

The efficiency calibration of the detector was made using two reference sources: ^{40}K and ^{137}Cs. The acquired spectrum is shown in Fig. 3.

The 661keV transition of ^{137}Cs is sitting on the high background produced both by the natural background and the Compton scattering of the 1461 keV γ-ray of ^{40}K (see Fig. 3). The analysis of the spectra has been performed with the "SPECTRG" software package [9]. By integrating the net 661 and 1461 keV photopeaks, the efficiency of the detector in the water environment has been deduced as follows:

Absolute total efficiency (661keV) = (4.2 ± 0.2)x10^{-5}

Absolute total efficiency (1461 keV)=(3.5 ± 0.1)x10^{-5}.

It is clearly seen that the counting rate of the system is very low when measurement is performed into the water. The efficiency of the new developed system (KA-TE-RINA) is on the other hand about 30% higher compared with a similar underwater system [8]. This result improves the measurement method drastically, since statistics is a very important criterion for low level radioactivity measurements, and in specific using NaI detectors.

The gain stabilisation of the system (voltage drift elimination) was performed using the 1461 keV of ^{40}K the 2615 keV of ^{208}Tl and the 50 keV of the seawater threshold. The linearity of the system was checked measuring the emission of other nuclides, like ^{214}Bi and ^{214}Pb. The non-linearity of used electronics is very low, i.e. the non-linear amplification coefficient is of order 10^{-9}.

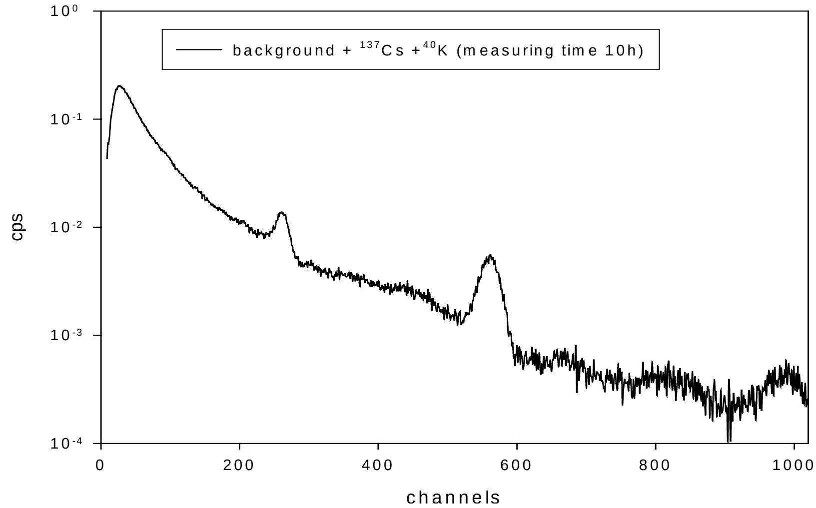

Figure 3. The acquired spectrum with the calibration reference sources: ^{40}K and ^{137}Cs.

3.3 Field data

Field measurements were performed in the open sea. The device was deployed at five different sites and at a depth of 3m. The measured spectra looked identical except of the spectrum derived in Lesvos. The increase of ^{40}K contribution (20%) on the specific spectrum indicates more saline water of the region. The result is also verified by the CTD measurement. Similar behavior was observed in the past [2]. An interesting result is the ^{137}Cs increment, in case of a preceded rainfall. This was encountered in the region of Varkiza (close to Athens). If one compares the measured data in Varkiza with the other data sets, observes a variation of the counting rate around 661 keV (see Fig. 4).

The volumetric activity increment of ^{137}Cs (Bq/m^3) due to rainfall, is calculated by using the Eq.1:

$$r(Bq/m^3) = \frac{cps}{\varepsilon \cdot I \cdot V} \tag{1}$$

where
cps: the counts per second for ^{137}Cs,
ε: the efficiency of the underwater system at the specific energy,
I: the yield and
V: the totally absorbed volume of the specific gamma ray.

The volumetric activity variation due to rainfall is (19 ± 4) Bq/m^3. The relatively large error (± 4Bq/m^3) is due to poor statistics (measuring time only 3h) and due to the various voltage drifts. The analysis of the spectra was made with the "SPECTRG" software. The software uses a house developed de-convolution algorithm [9].

4. SUMMARY – CONCLUSIONS

A new sensing radioactivity apparatus is developed for measuring artificial and natural radioactivity in the marine environment. The developed hardware and software for underwater gamma ray spectrometry has been tested and calibrated in the laboratory for its stability and its sensitivity for ^{137}Cs. The device could observe volumetric activity variation of (19 ± 4) Bq/m^3 of ^{137}Cs at open sea within the period of 3h. The derived sensitivity is about 30% higher when compared with a similar system (RADAM III) used in the POSEIDON network [7]. The spectra with the KA-TE-RINA system are gain stabilized by using three photopeaks: 2615 keV (^{208}Tl), 1461 keV (^{40}K) and 50 keV (threshold energy in seawater).

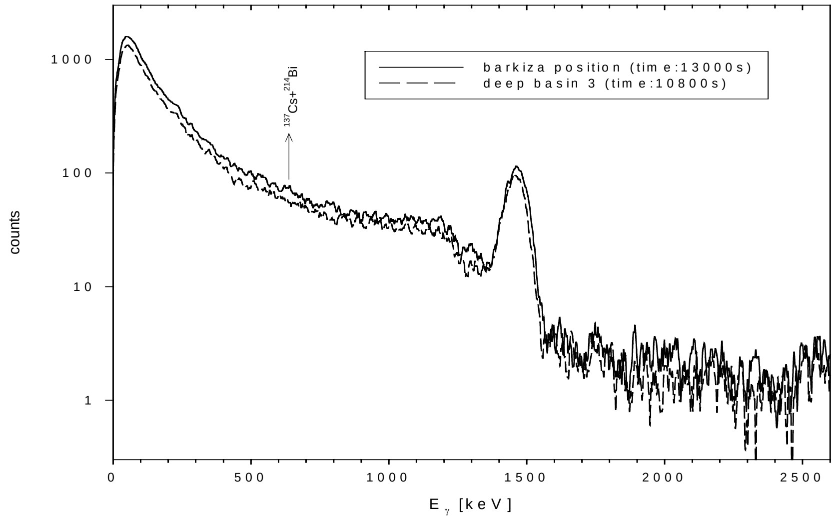

Figure 4. The measured spectrum at the DB3 (3m) and at Varkiza (Athens) position in a depth of 3m. The measuring times are 10800 and 13000 seconds respectively. The measured spectrum in Varkiza is after a rainfall.

In the future, the integration of the developed system on a real-time data-forwarding sea surface buoy will furthermore complete the aim of this work. This is consistent to the further idea of building an operational and low cost buoy network, for monitoring radioactivity levels in the open sea. It is intended, to embed the device in a floating measuring system, for the surveillance and monitoring of gamma radiation in the marine environment. Additionally a similar system could be applied in rivers and lakes as an early alarm system for possible water pollution.

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