Journal of Environmental Radioactivity 108 (2012) 50–59

Contents lists available at SciVerse ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

In-situ radionuclide characterization of a submarine groundwater discharge site at Kalogria Bay, Stoupa, Greece

Christos Tsabaris [a,*], Dionisis L. Patiris [a], Aristomenis P. Karageorgis [a], George Eleftheriou [b], Vassilis P. Papadopoulos [a], Dimitris Georgopoulos [a], Evangelos Papathanassiou [a], Pavel P. Povinec [c]

[a] Hellenic Centre for Marine Research, Institute of Oceanography, 19013 Anavyssos Attica, P.O. 712, Greece
[b] National Technical University of Athens, Department of Applied Mathematic and Physical Science, 15780 Athens, Greece
[c] Comenius University, Faculty of Mathematics, Physics and Informatics, Mlynska dolina F-1, SK-84248 Bratislava, Slovakia

ARTICLE INFO

Article history:
Received 19 January 2011
Received in revised form
27 July 2011
Accepted 5 August 2011
Available online 8 September 2011

Keywords:
Marine environment
Submarine groundwater discharge
Radon
Thoron
In-situ gamma-ray spectrometry
Kalogria Bay

ABSTRACT

In-situ underwater gamma-ray spectrometer KATERINA was used for continuous measurements of radon progenies (^{214}Pb, ^{214}Bi), thoron progeny (^{208}Tl) and ^{40}K in submarine groundwater discharge (SGD) sites at Kalogria Bay, SW Peloponnesus (Greece). The spectrometer was deployed attached on measuring platform along with two conductivity - temperature data loggers while underwater battery packs supplied the system for acquisition periods up to 25 days. The radionuclide time series together with salinity data were obtained for spring (wet) and summer (dry) seasons. The ^{40}K activity concentrations correlated well with salinity of the emanating groundwater. Although the ^{214}Bi and ^{208}Tl activities showed usually similar trends anticorrelating with salinity, in some cases ^{208}Tl did not follow the ^{214}Bi record due to changes in the dynamics of the groundwater aquifer. As the half-life of ^{220}Rn is very short (55.6 s), its concentration in SGD may depend on the distance from its origin to the monitoring point. The observed temporal variations of ^{214}Bi and ^{208}Tl confirmed advantages of continuous in-situ monitoring of SGD in coastal areas.

1. Introduction

The marine environment and especially near shore environments may be enriched in radionuclides as a result of various human activities and natural phenomena. The primary sources of anthropogenic radionuclides are global fallout from past atmospheric nuclear weapons tests (Livingston and Povinec, 2002), the Chernobyl accident, and discharges of liquid radioactive wastes from nuclear reprocessing facilities (Livingston and Povinec, 2000; Matishov and Matishov, 2004).

On the other hand, transport of radionuclides enriching the coastal zone may also be due to natural phenomena. The most important are discharges of large water masses by rivers, as well as by submarine groundwater springs. The input of radionuclides to coastal regions by rivers can take place by wash out of natural and global fallout radionuclides from soil, as well as technologically enhanced natural radioactive materials within watershed areas, or by disposal of such materials across rivers. On the other hand, groundwater is rich in natural radionuclides which enter into the water body from the subsoil mainly by weathering.

The submarine groundwater discharge (SGD) is an alternative pathway for such materials to be transported into a coastal marine environment (Burnett et al., 2006). The concentrations of radionuclides are usually low, so there is no threat to the environment. However, many studies on SGD showed spatial and temporal variations of several radionuclides which could be useful for better understanding of coastal processes (Burnett et al., 2006). Among them radon and radium isotopes, and the primordial thorium and uranium are the commonest radio-tracers. Radon is considered an excellent indicator for the presence and the intensity of groundwater springs in coastal areas, since it diffuses from the subsoil into the body of groundwater reaching high activity levels, and can be easily detected by various techniques. The most frequently used methods are based on sampling water at specified time intervals. Radon is degassed from the sample, and it is transported by a suitable gas to measuring chambers where particles (alpha and beta) which are emitted due to radon and/or its progeny decays are counted. Although all the measuring process may take place on-site by mobile detection systems (e.g RAD-7), the SGD is not really surveyed continuously as there are gaps between two consequent

* Corresponding author. Tel.: +30 22910 76410.
E-mail address: tsabaris@hcmr.gr (C. Tsabaris).

0265-931X/$ – see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2011.08.005

sample analyses. Moreover, grabbing sediment samples required for calibration is a high cost and time consuming procedure.

It has been recognized during the last years that the utilization of in-situ gamma-ray detection systems would be a useful technique in SGD studies, since such systems are capable for continuous monitoring of radionuclide levels in water (Povinec et al., 1996, 1997; Osvath and Povinec, 2001a; Tsabaris and Ballas, 2005; Tsabaris, 2008), or for mapping the distribution of radionuclides in sediments (Osvath et al., 1999, 2001b). In-situ systems could be fully-integrated, capable to stand many demanding environmental conditions (depth stresses, chemical corrosion, etc.), and operating submerge in the aquatic environment. Also, they offer a full automation of the measuring process, fast and cost efficient determination of radionuclide activities since there is no need for sampling, transport and chemical treatment (Povinec et al., 2001). Additionally, another option could be the remote transmission of data when the monitoring systems are installed on permanent stations far from the laboratories (Wedekind et al., 1999; Debauche, 2004; Osvath et al., 2005; Tsabaris et al., 2005; Tsabaris, 2008).

Recently in-situ gamma-ray continuous monitoring systems have been successfully applied in SGD studies (Povinec et al., 2006a; Tsabaris et al., 2010) since groundwater is rich in radon and its decay products are usually gamma-ray emitters. The in-situ gamma-ray spectrometry can be applied for simultaneous quantitative determination of all radionuclides present in the emanating groundwater (Tsabaris et al., 2008; Bagatelas et al., 2010).

The aim of this study is to present results from the application of the in-situ underwater gamma-ray spectrometer named KATERINA (the abbreviation comes from the Greek words Innovative Sensor for Artificial and Natural Radioactivity) in the study of submarine groundwater springs at the Kalogria Bay, located southwest of the Peloponnesus Island (Greece).

2. Materials and methods

The underwater gamma-ray spectrometer KATERINA was deployed in the Kalogria Bay for several periods from July 2009 till May 2010. The system operated autonomously without computer connection, pre-programmed to acquire gamma-ray spectra every 12 h. The spectra were buffered into an internal memory, which were later recovered from the system. External battery packs were used as a power supply providing operation time up to 25 days. The gamma-ray spectrometer was accompanied with two conductivity-temperature logger (CT) providing data to calculate salinity values. The results are presented as time series in order to investigate radionuclide activity variations with salinity records of the emanating groundwater.

2.1. Study area

The Kalogria Bay is located north of Stoupa town, in the southwest Peloponnesus (Messinia Prefecture). The area is characterized by the presence of numerous minor SGDs, visible as small gyres at the sea surface (small circles in Fig. 1a). The major SGD site is located ~100 m offshore and creates at the sea surface two impressive gyres, with diameter varying between 25 and 60 m (Fig. 1b). The SGD emanates groundwater as a vertical jet from a depth of 25 m. The spring is partly covered by a limestone block with dimensions approximately 5 × 4 × 1 m. Divers observed smaller jets emanating groundwater from the sides of the rock block, from surrounding rock fissures, as well as three minor SGDs in a distance of a few meters from the major SGD. During the winter period, the discharge was so strong that divers could hardly approach the core of the SGD and place the measuring platform safely on the sea bed.

2.2. The in-situ underwater gamma-ray spectrometer

The underwater gamma-ray spectrometer KATERINA is a fully-integrated unit since there is no need of computer connection to acquire and store the data. All the modules are enclosed inside a special housing made by acetal, which allows deployment depths up to 400 m, and at the same time exhibits the lowest gamma-ray attenuation. The detection module is based on a 3" × 3" NaI(Tl) scintillation crystal in contact with a photomultiplier tube with build-in high voltage controller. The initial signal is processed consecutively by a preamplifier, an amplifier, and then by an analog to digital converter. Electronic drifts of the amplification gain, which are usually observed in field installations are auto-compensated by a dedicated circuit on the amplifier module. The signals then pass through a multi-channel analyzer, and the obtained data are stored in a non-volatile memory module with microcontroller (after a preset of the acquisition time). A power unit is included to distribute the input voltage (12–15 V DC) from an external source (underwater battery pack) to the electronic modules.

In order to use the spectrometer for the determination of the radon ^{222}Rn daughters (^{214}Bi and ^{214}Pb), the thoron ^{220}Rn daughter (^{208}Tl) and ^{40}K, the system was energy calibrated and tested for its stability with respect to temperature variations and energy resolution. The calibrations were done inside a laboratory tank of 5.5 m^3 filled with freshwater. At the bottom of the tank, an electric pump circulated the water to insure homogeneity. The water was spiked for calibrations with standard solutions of mono-energetic radionuclides ^{137}Cs, ^{40}K ^{99m}Tc and ^{111}In. The whole procedure of the calibration is out of the scope of this work and it is described in details in a previous work (Tsabaris et al., 2008).

After the analysis of the spectra the measured counts under any photopeak are converted to activity data (in Bq/m^3) by the following equation (Eq. (1)):

$$r\left(Bq/m^3\right) = \frac{\mathrm{cps}}{\varepsilon_m I_\gamma} \quad (1)$$

where cps denotes the counts per second recorded for each photopeak, ε_m is the efficiency for an effective volume of water, and I_γ is the emission probability. The efficiency of the system, e_m, was previously determined experimentally at 141, 661 and 1461 keV (Bagatelas et al., 2010). Monte Carlo simulations were also carried out for calculating the efficiency of the system at any gamma-ray energy (Bagatelas et al., 2010). The minimum detectable activity (MDA) of the system was studied in both freshwater and seawater environments (Bagatelas et al., 2010). As concerns radon progeny ^{214}Bi at 609 keV, the MDA was calculated to be 0.03 and 0.05 Bq L^{-1} in freshwater and seawater, respectively. The MDA of the system depends on the ^{40}K concentration in the water. However, the expected activity of ^{214}Bi in groundwater is at least by one order of magnitude above the MDA of the system.

Moreover, the background of the system due to potential radon daughter's accumulation on the housing material (acetal) as a result of previous deployments into radon rich environments was examined. Gamma-ray spectra were acquired in several deployments in a reference lake where ^{222}Rn activities were below the MDA of the system. The spectrum analysis exhibited that the gamma-ray lines from radon progenies were not present, implying that any background activity from the radon progenies into the material of the enclosure is below the MDA of the system.

The analysis of the spectra was performed using the SPECTRW software (Kalfas, 2010). The quantification for single gamma-lines (concerning mainly the radon/thoron progenies ^{214}Pb, ^{214}Bi and ^{208}Tl) was carried out following the methodology already described (Bagatelas et al., 2010). The ^{208}Tl is a decay product of ^{224}Ra with