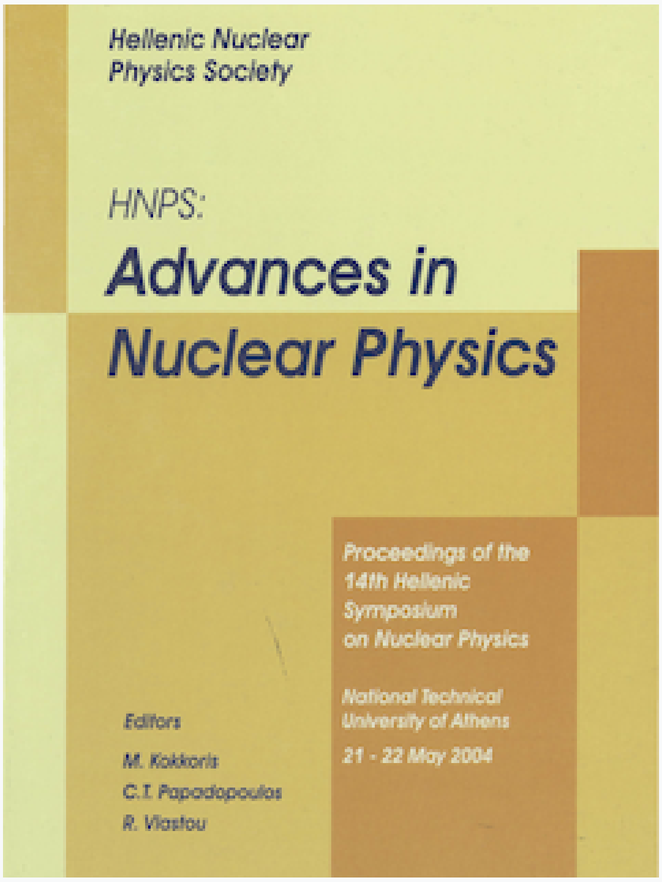
HNPS Advances in Nuclear Physics

Vol 13 (2004)

HNPS2004



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doi: 10.12681/hnps.2949

Rainfall Induced ^{137}Cs Radioactivity in the Aegean Sea

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Abstract

The development of autonomous measuring systems for radioactivity in the marine environment is today of important scientific priority for the marine sciences and especially for the Operational Oceanography. This paper concentrates on the improvement and application of an existing NaI-based gamma ray detection system for radioactivity measurements in seawater. The set up of the system will be described together with the appropriate calibrations in the laboratory. The operation of the system in the Aegean Sea provides the monitoring efficiency at 1461 keV by inter-calibration with an appropriate salinity sensor mounted close to the NaI-detector. The gross counting rate was found constant within the statistical uncertainty of the measurements, during dry periods. The artificial radioactivity (^{137}Cs) in the North Aegean Sea was increased, however, up to seven times after strong rainfall, compared to the mean radiation level as given in literature. This increment remains only for a short period of time (¡ 2 days), due to strong advection processes of the water masses during strong rainfall.

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1 Introduction

The use of in situ gamma spectrometry to monitor radioactivity in the marine environment has become increasingly attractive since many improvements in hardware and software have been achieved to increase the sensitivity of the measuring devices [1-4]. The use of this type of spectrometers offers the advantage of collecting and transmitting the bulk gamma radiation as well as the whole spectrum on a continuous basis. The bulk radiation level varies with time and region due to meteorological conditions, suspended matter, salinity etc. The continuous monitoring of gamma radiation in the marine environment provides very important information on various environmental processes like rainfall wash out [5] and freshwater discharge [6].

For in situ monitoring of seawater radioactivity, gamma-ray spectra are acquired by NaI underwater detectors, installed on floating measuring systems with special equipment for transmitting the data to the operational centre. Continuous measurements in the marine environment are very scare since the radioactivity detection has many difficulties due to the low efficiency of the detection systems (in the order of 10^{-5}), to the high background produced by the Compton scattering of 1461 keV gamma energy of ^{40}K and to the floating measuring system tolerance depending on the various applications in the marine environment.

This study demonstrates the use of an underwater NaI detection system for the investigation of radioactivity rise in the Aegean Sea with the POSEIDON network, in case of rainfall. Preliminary results on the volumetric activity enhancement of ^{137}Cs due the wash out, will be also presented.

2 Experimental Set-up

The Hellenic Centre for Marine Research (HCMR) owns and maintains RADAM III detectors constructed by the Norwegian Company OCEANOR [7]. The detector system consists of a scintillator system and a power unit shielded by an aluminum and polyester (POM) pressure housing. The detector unit is a 3"x3" NaI crystal with built in photomultiplier tube, preamplifier, an analog-digital converter, a high voltage device, a temperature sensor together with the electronics for data acquisition, storage and transmission. The power unit operates at DC 12V. The electronic modules are highly miniatured to fit inside the sensor housing (80x60mm) and the power consumption is very small (¡2W). The operating temperature ranges between -10 and +50°C and its influence to 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 an ordinary laboratory analysis using NaI scintillators. This type of sensor can be used for operational purposes by attaching it on oceanographic buoys and can operate under severe weather conditions and especially with extreme waves driven by wind forces and high temperature gradients.

The radioactivity detector is housed in the SEAWATCH floating measuring system constructed by the OCEANOR Company. The system has been used for the last few years in various oceanographic applications [8]. A schematic view of the POSEIDON network operation together with the appropriate operation for set up and transmission procedures is shown in Fig. 1. The measured data are saved in a database for further analysis. The network is a real time monitoring and forecasting system for the marine environmental conditions in the Aegean Sea. The observational basis of the network consists of 11 oceanographic buoys that acquire and transmit in real-time oceanographic data at the air-sea interface [9-10].

The communication system allows two-way data transmission between the buoys and the Operational Centre (HCMR) : satellite INMARSAT-C and GSM cell phone communication. The buoys are equipped with both systems. The GSM system is suitable only for buoys close to the coast with low transmission cost. The INMARSAT-C is an autonomous system that allows transmission from open sea and is not affected by weather conditions. The data are automatically transmitted every three hours to the Operational Centre (HCMR). This interval allows an adequate presentation of the daily cycle and at the same time eliminates power and stability problems that could have occurred. An automatic quality control procedure removes spikes and erroneous data that are outside from a predefined range. A second quality control that identifies abnormal trends, usually related to bio-fouling, is performed every month.

The NaI detector system, has been tested and calibrated in the laboratory, in a $5.5m^3$ water tank, before its deployment in seawater. The sensor was mounted in the middle of the tank, surrounded by one meter of water, which is adequate to imitate the real marine environment, due to the high attenuation of the γ-rays in the water. At the bottom of the tank an electric pump was placed in order to avoid sedimentation, to mix the water with the appropriate radionuclides (^{137}Cs and ^{40}K) used for the calibrations and to ensure homogeneous conditions. The water, containing a specific amount of ^{137}Cs and ^{40}K, was circulated and the photopeak contribution from the 662 and 1461 keV gamma rays was acquired. Thus, the relative efficiency of the system could be deduced at these energies. ^{99m}Tc was also mixed in the tank, in order to study the operation of the detection system at low energies (141 keV). The spectra corresponding to these radionuclides were used for the energy and volumetric activity calibration as well as determination of energy resolution and efficiency of the detector when it operates in an underwater mode.

3 Data Reduction

In the present work monitoring data from different regions of the Aegean Sea were investigated. The NaI-detection system was mounted to floating measuring systems and transferred to the regions of Glyfada, Athos and Lesvos

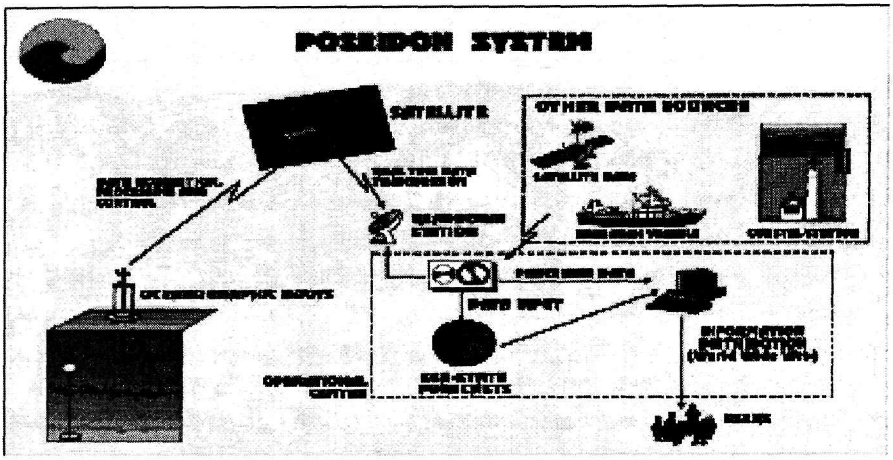

Fig. 1. The POSEIDON network system

with the research Vessel "AEGEAON". The measured data were automatically saved in a database as raw measurements every three hours. Stability checks were carried out and in case of drifts the appropriate corrections were applied to the spectra to maintain the energy calibration. The short period stabilized spectra were then added in order to produce one day spectra for further analysis by using the software code SPECTRG [11].

By comparing the one day acquired data over long periods, several changes have been observed, exceeding the statistical variations, between dry and wet periods. Such changes originate predominantly from rain showers [12, 13] and contain both natural and artificial radionuclides. The data analysis for the estimation of possible ^{137}Cs contamination was processed by subtracting the measured spectrum before rainfall from the measured spectrum just after the rainfall. Several tests have been carried out by comparing measured spectra acquired during dry periods, in order to specify the background spectrum at each location. Before the subtraction, the two spectra were calibrated precisely for energy and corrected for the dead time of the system. The correction of the spectra for possible voltage drifts was performed with gain-stabilization procedure by using the two photopeaks at 50 keV (lowest gamma ray energy in seawater) and 1461 keV (^{40}K gamma ray emission).

4 Results – Discussion

The main characteristic of the measured data during dry periods, is that the gross counting rate of the system remains stable within the limits of the statistical uncertainties, while counting rate variations can be encountered between different regions. As an example, in Fig. 2, the variation of gross

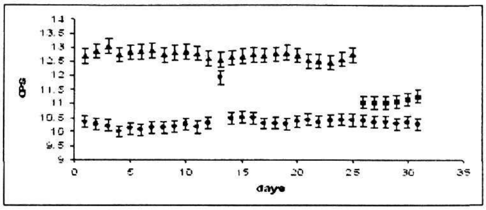

Fig. 2. The counting rate (cps) at Lesvos in May 2000 (triangles), at Athos in May 2000 (rectangles) and at Athos in August 2001 (points).

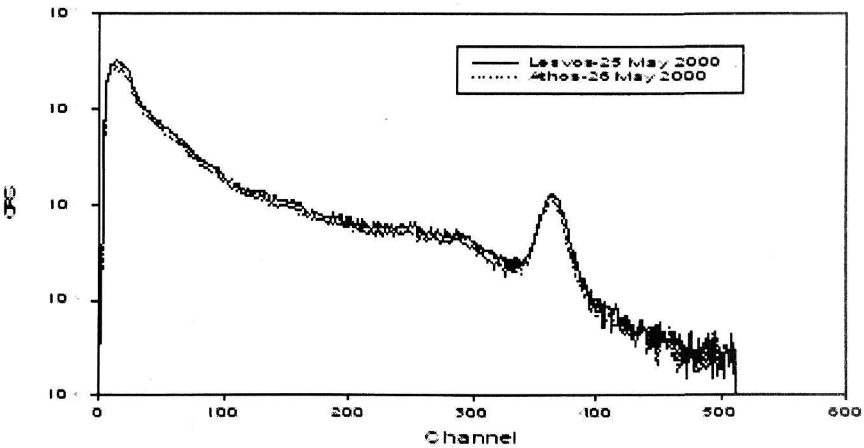

Fig. 3. Measured counting rate (cps) as a function of energy (channels) at Lesvos (25 May 2000 - line) and at Athos (26 May 2000 - dot line).

counting rate (total number of counts per second) is depicted in May 2000 for a period of 25 days at Lesvos (triangles) and then for 6 days at Athos (rectangles). The mean value of gross counting rate at Lesvos is 13.7cps, while at Athos 11.2cps. This difference could be attributed to the higher salinity of the seawater at Lesvos island. To corroborate this argument, the detailed gamma-ray spectra taken on the 25^{th} May at Lesvos and 26^{th} May at Athos, are presented in Fig.3. It can be clearly seen that the 1461keV gamma-ray of ^{40}K, as well as its Compton tail at lower energies, is considerably stronger at Lesvos than at Athos, indicating the enhanced salinity in the water region of Lesvos. Concerning artificial radioactivity (mainly due to ^{137}Cs), the radiation level was stable within the statistical uncertainty in both aforenamed locations.

Gross counting rate for Athos is also shown in Fig.2 (points) for a month period in August 2001. The observed average value is 10.2cps, very close to the corresponding one measured in May 2000 (rectangles). On the 13^{th} August 2001, however, a significant enhancement of the total gamma count rate is observed, reaching 11.9cps, following a rainfall on the previous day. It is well known [12] that the rain showers induce radioactive wash out of natural origin. Furthermore, from the gamma-ray spectrum taken on the 13^{th} August 2001, shown in Fig.4, it is obvious that artificial radioactivity due to ^{137}Cs, was also detected after the rainfall. Similar phenomena have also been observed in other regions of the Aegean sea, after rainfall. Due to the poor energy resolution of the NaI detector, a nuclide specific analysis of complex spectra is not possible. Nevertheless, in the case shown in Fig.4, an attempt was made to distinguish between the 662keV γ-ray of ^{137}Cs and the 609keV of ^{214}Bi from the radioactive series of natural ^{238}U. An algorithm was adopted for the deconvolution of two energy photopeak contributions. The fitting procedure was applied in the energy interval from 535 to 715 keV. The FWHM at the lowest energy is wider due to the ^{208}Tl contribution at 583 keV, which increases the width of the deconvoluted low energy photopeak. The analysis of the spectrum is presented in Fig. 4 along with the identified energy, the width (FWHM) and the net area results. However, a more complex analysis of the spectrum is currently under investigation.

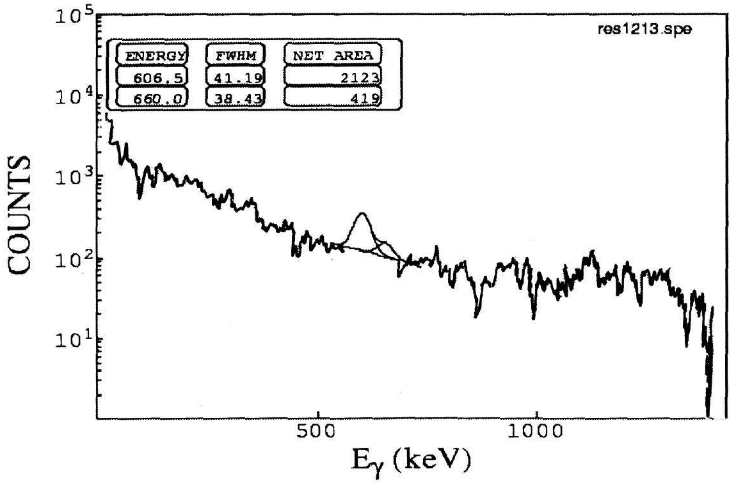

Fig. 4. The deconvolution result for separating the 609 keV (^{214}Bi) contribution from 662 keV^{137}Cs) in the gamma-ray spectrum on the 13^{th}August 2001.

The γ-activity variation was transformed in Bq/m^3, as described in [1], by taking into account the yield at 662 keV (84.62%), the measuring time (1 day), the infinitive volume for the specific γ-ray and the efficiency of the system in seawater [13]. The preliminary results for ^{137}Cs activity variation in the region of Athos are given in Table 1 for periods with strong rainfall. The maximum detected volumetric activity for ^{137}Cs on the 13^{th} August 2001, was estimated to be (31 $\pm$ 3) Bq/m^3. This value is significantly higher compared to the background radiation level of 3.5-5.5 Bq/m^3 of ^{137}Cs in the north Aegean Sea

[14-16]. Such variations are expected since ^{137}Cs is an artificial radionuclide, which is transported from land by wind and deposited on the sea surface by rainfall. It remains only for a short period of time (¡2 days), due to strong advection processes of the water masses at the specific region during the strong event.

Table 1
^{137}Cs activity variation in the region of Athos for periods with strong rainfall.

STATION	**DATE**	**^{137}Cs x10^{-3}(cps)**	**Volumetric Activity of ^{137}Cs (Bq/m^3)**
ATHOS	October 2000 8^{th} 9^{th} 10^{th}	background not detected within 1day background	< LLD
ATHOS	November 2000 26^{th} 27^{th} 28^{th} 29^{th}	background 3.3 4.1 background	18 $\pm$ 2 23 $\pm$ 2
ATHOS	July 2001 10^{th} 11^{th} 12^{th}	bckground not detected within 1day background	< LLD
ATHOS	August 2001 12^{th} 13^{th} 14^{th}	background 6.8 background	31 $\pm$ 3
ATHOS	November 2002 6^{th} 7^{th} 8^{th}	background 4.0 background	21 $\pm$ 2

5 Conclusions

The improvements for the operation and application of RADAM III detection system installed at oceanographic buoys, offer qualitative and quantitative information of the various radionuclides which are detected in seawater. Such radionuclides may arise from natural radioactivity (decay of ^{40}K and the daughter products of ^{232}Th and ^{238}U radioactive series) as well as from possible artificial radioactivity (^{137}Cs). The application of the detection system in the Aegean Sea indicates that rainfall may increase the gross activity of seawater.

The study of the variation of artificial radioactivity, provides a very important result for ^{137}Cs pollution. It was observed that during strong rainfall an enhancement of the volumetric activity of ^{137}Cs occurs. Despite of the poor energy resolution of the NaI spectrometer, a fitting procedure was tried, in an attempt to separate neighboring photopeaks and identify ^{137}Cs in the spectrum. The volumetric activity of ^{137}Cs after strong rainfall was thus extracted for some regions in the Aegean sea and was found to fluctuate between 18 and 31 Bq/m^3, which is 5 to 7 times higher compared to background radiation measurements.

The operation of the RADAM III detector can be applied for monitoring quantitatively the natural and artificial radioactivity in the marine environment. It can also be used as an early warning system very effectively, since the background radiation at 3m depth is well specified experimentally in three regions in the Aegean Sea. The system has been used in conjunction with the monitoring network "POSEIDON" and provided very useful information for the variation of the γ- activity level in the Aegean Sea with rain showers. In future, the correlation of the observed enhancement of radioactivity with other environmental parameters will be investigated.

Acknowledgements

The authors would like to thank the POSEIDON group for the continuous technical support for the operation of the RADAM III detectors installed at the POSEIDON network.

References

[1] Tsabaris, C. and Ballas, D., "On line gamma-ray spectrometry at open sea". App. Radiation & Isotopes 62, 83-89 (2005).

[2] van Put, P., Debauche, A., de Lellis, C. and Adam, V. Performance level of an autonomous system of continuous monitoring of radioactivity in seawater. Journal of Environmental Radioactivity 72 177-186 (2004).

[3] Vlachos, D.S. and Tsabaris C. "Response Function Calculation of an Underwater Gamma Ray NaI(Tl) Spectrometer", Nuclear Instruments & Methods in Physics Research A 539, 414-420 (2005).

[4] Vlastou, R., Ntziou I., Th., Kokkoris, M., Papadopoulos, C. T. and Tsabaris C. "Monte Carlo simulation of γ-ray spectra from natural radionuclides recorded by a NaI detector in the marine environment", Applied Radiation and Isotopes, in press (2005).

[5] Paatero, J. "Wet deposition of radon-222 progeny in northern Finland measured with an automatic precipitation gamma analyzer". Radiation Protection Dosimetry 87, 273-280 (2000).

[6] Burnett, C. W. and Dulaiova, H. "Estimating the dynamics of groundwater input into coastal zone via continuous radon-222 measurements", Journal of Environmental Radioactivity 69, 21-35 (2003).

[7] Aakenes, U. R., "RADAM - Expandable gamma monitoring units for military and civil use", International Conference on Environmental radioactivity in the Artic, Oslo, 315-323 (1995).

[8] Hansen, S.E., and Stel, J.H, "SEAWATCH: Performance and future In Operational Oceanography". The challenges for European Co-operation, Elsevier Oceanography Series, 62, Elsevier Science B.V. (1997).

[9] Nittis K., Soukkisian T., Chronis G. and the POSEIDON project group, "Real time monitoring and forecasting in the Aegean Sea by the POSEIDON system". Protection and Restoration of the Environment V, Thassos, Greece 3-6 July (2000).

[10] Soukissian, T.H., Chronis, G. T. & Nittis, K., "POSEIDON: Operational Marine Monitoring System for Greek Seas. Sea Technology 40, 31-37 (1999).

[11] Kalfas, C.A., "SPECTRG-Software Package for Analysis of Gamma-Ray spectra". Personal Communication (2000).

[12] Wedekind, Ch., Schiling. G., Gruettmueller, M., and Becker, K., "Gamma-Radiation monitoring network at sea". Applied Radiation and Isotopes 50(1999)733.

[13] Tsabaris, **C.**, Vlachos, D. S, Papadopoulos, C.T, R. Vlastou and C. A. Kalfas, "Application of an Underwater γ- ray Spectrometer for Radioactivity Measurements". Proceedings of the International Symposium on North Aegean System Functioning and Interregional Pollution, Kavala, p.31 (2000).

[14] Florou H. and Kritidis P. "The dispersion of Cs-137 in the Aegean Sea". Radiochimica Acta 66/67, 415-417 (1994).

[15] Papucci, C. and Delfanti, R.. "^{137}Caesium distribution in the eastern Mediterranean Sea: recent changes and future trends". The Science of the Total Environment 237, 67-75 (1999).

[16] Delfanti, R., Tsabaris, C., Papucci, C., Kaberi, H., Lorenzelli, R., Zervakis, V., Tangherlini & M., Georgopoulos, D., "Re-distribution of ^{137}Cs Chernobyl signal in the Aegean Sea". Proceedings of the International Conference on Isotopes in Environmental Studies-Aquatic Forum 2004, p.7, Monaco (2004).