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Response function calculation of an underwater gamma ray NaI(Tl) spectrometer

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Abstract

Autonomous measuring systems for radioactivity in the water environment require caution concerning the specifications of power consumption, stability, communication equipment, tolerance and efficiency. In addition, the calculation of the response function of the whole system is essential for suppressing the background of the measurement and for estimating the concentration of the involved radionuclides, especially given the greater probability of primary gamma photons undergoing multiple scattering events before they interact with the sensing device. In this work, a method is presented that can be used to calculate the response function of a NaI(Tl)-based spectrometer when it is used in the marine environment. The method takes into account both the scattering of photons in the water (analytical calculations) and the detection mechanism of the sensor (Monte-Carlo simulation). In order to validate the method, the calculated response function has been used in a real measuring system in order to estimate the concentration of ^{40}K and thus the salinity of the water.

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Keywords: Marine radioactivity; NaI(Tl) detector; Response function

1. Introduction

The development of a measurement system for radioactivity in the water environment is today of important scientific priority for the marine sciences and especially for the operational oceanography [1]. The average concentration of ^{137}Cs in surface waters of the world's oceans and seas varies from 0.3 Bq/m^3 (Antarctic) to 100 Bq/m^3 (Baltic) [2]. The Baltic Sea, the Black Sea as well as the Mediterranean Sea were the main reservoirs for the released radionuclides due to the Chernobyl accident.

A great deal of effort has been given in the last decade to develop a stationary monitoring network in order to observe the sea for radioactive contamination and especially to detect ^{137}Cs [3,4].

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In the field of underwater gamma ray spectroscopy, the detection systems NaI(Tl) and HPGe are commonly used for in situ monitoring of the radioactivity [5–8]. The HPGe detectors have been used for a number of applications in the marine environment in the last few years, but they could only monitor for a limited period of time and for fields close to power supplies. With respect to NaI detectors, these systems involve a high level of background radiation, rendering them incapable of detecting low-level radioactivity ($<100\,\mathrm{Bq/m^3}$), especially in cases where the radionuclides emit low-energy gamma rays ($<600\,\mathrm{keV}$). Moreover, these sensors show a drift in the produced spectra, changing the exact position of the measured photopeaks and altering certain characteristics of the measured spectrum, like the full-width at half-maximum. Periodic energy calibration can solve the above problems but only for a short period of time (once per month based on our experience), thus increasing dramatically the operational cost of the system due to the large maintenance cruise expenses. On the other hand, an auto gain-stabilized system (e.g. using the position of ^{40}K peak) does not assure the exact reposition of the measured photopeaks, because the energy drift is not a linear function of measured energy [9,10]. The ideal solution to the above problems is the auto-energy calibration, which up to the linear approximation needs a knowledge of the exact position of two photopeaks. Finally, the energy spread of the photons in seawater, the energy drift and the poor energy resolution of the NaI detector reduce the capability of the whole system for underwater operational use.

In order to improve the use of the NaI spectrometer in seawater, a suitable technique has been developed for the calculation of its response function. The idea is that, before simulating the detecting mechanisms of NaI(Tl) detectors, one has to calculate the energy distribution of photons in seawater since primary gamma photons, which undergo multiple scattering events before they interact with the sensing device, contribute significantly to the measured spectrum. 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 concentration ($\mathrm{Bq/m^3}$) of man-made radionuclides in seawater.

2. Response function calculation

The photon energy distribution in seawater can be described in equilibrium by the following equation (the photon energy is represented in multiples of m_0c^2, where m_0 is the rest mass of electron and c the light speed):

$$0 = G\delta(\alpha_t - \alpha_0) + \int_{\alpha_t}^{\alpha_0} n(\alpha)C(\alpha,\alpha_t)\,\mathrm{d}\alpha + 2\delta(\alpha_t - 1)\int_{2}^{\alpha_0} n(\alpha)R(\alpha)\,\mathrm{d}\alpha - n(\alpha_t)S(\alpha_t) \quad (1)$$

where $n(\alpha)$ is the concentration of photons with energy α. $G\delta(\alpha_t - \alpha_0)$ is the generation of photons at energy α_t, when G photons per second are generated with energy α_0. $C(\alpha,\alpha_t)$ is the probability per second for a photon with energy α to move to energy α_t due to Compton scattering. Thus, the second term of the second part of Eq. (1) is the number of photons that move to energy α_t due to Compton scattering per second. $R(\alpha)$ is the probability per second for a photon with energy α to be absorbed, leaving in its place an electron positron pair. Finally, the positron will interact with an electron, producing two photons with energy m_0c^2 ($\alpha = 1$). Thus, the third term of the second part of Eq. (1) is the total number of photons with energy $\alpha = 1$ that are produced per second. $S(\alpha_t)$ is the probability per second for a photon with energy α_t to scatter in a different energy due to all scattering mechanisms, i.e. photoelectric effect, Compton scattering and pair production.

The functions $R(\alpha)$ and $S(\alpha)$ are calculated from XCOM software [11]. The function $C(\alpha,\alpha_t)$ is calculated from the Klein–Nishina formula [12], which gives for the angular distribution of scattered photons

$$p(\theta) = \pi r_0^2\left(\frac{\alpha}{\alpha_0}\right)^2\left(\frac{\alpha}{\alpha_0} + \frac{\alpha_0}{\alpha} - \sin^2\theta\right)$$
$$\alpha = \frac{\alpha_0}{1 + \alpha_0(1 - \cos\theta)}. \quad (2)$$

Changing now the variable from θ to α, we get

$$C(\alpha_0,\alpha) = p(\alpha) = \pi r_0^2 \frac{1}{\alpha_0^2}\left[\frac{\alpha}{\alpha_0}+\frac{\alpha_0}{\alpha}-1+\left(1-\frac{1}{\alpha}+\frac{1}{\alpha_0}\right)^2\right]. \quad (3)$$

It is clear now that Eq. (1) is a continuity relation. The meaning of this equation is that in equilibrium, the total number of photons that move from an energy α to another energy is equal to the total number of photons that move to energy α. Due to the existence of the delta functions in the above equation, the solution can be written as

$$n(\alpha) = \lambda(\alpha) + k\delta(\alpha-\alpha_0) + m\delta(\alpha-1) \quad (4)$$

where $\lambda(\alpha)$ is a smooth function in energy α with $\lambda(\alpha)=0$ when $\alpha=\alpha_0$. Substituting the expression for $n(\alpha)$ in Eq. (1), we can calculate the coefficients k and m as follows:

$$k = \frac{G}{S(\alpha_0)} \quad (5)$$

$$m = 2\frac{kR(\alpha_0)+\int_2^{\alpha_0}\lambda(\alpha)R(\alpha)\,\mathrm{d}\alpha}{S(1)}. \quad (6)$$

Substituting k and m in Eq. (4), $\lambda(\alpha)$ can be calculated from the following equation:

$$\frac{GC(\alpha_0,\alpha_t)}{S(\alpha_0)} + 2\frac{(G/S(\alpha_0))R(\alpha_0)+\int_2^{\alpha_0}\lambda(\alpha)R(\alpha)\,\mathrm{d}\alpha}{S(1)} \times C(1,\alpha_t) + \int_{\alpha_t}^{\alpha_0}\lambda(\alpha)C(\alpha,\alpha_t)\,\mathrm{d}\alpha - \lambda(\alpha_t)S(\alpha_t) = 0. \quad (7)$$

Eq. (7) can be solved arithmetically, starting from the fact that $\lambda(a_0)=0$. This distribution of photon energies inside seawater is considered to be the same everywhere and the directions of photon propagation are considered to be uniform. Fig. 1 shows the calculated photon distribution for different values of a_0 with $G=1$. One very interesting point that comes out from Fig. 1 is that the distribution function has a peak at 50 keV, independently of the initial photon energy. This is due to the fact that below 50 keV, the photoelectric effect is dominant in seawater, making all photons

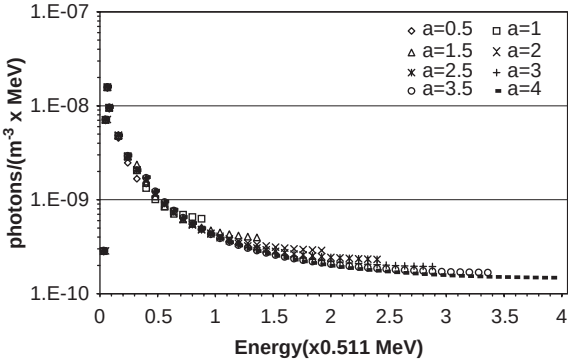

Fig. 1. Energy distribution of photons in seawater for different values of initial photon energy. The initial photon energy E is represented in dimensionless form ($a = E/m_0c^2$, where m_0 is the electron rest mass and c the speed of light).

with energies beyond 50 keV to disappear very fast. This peak will be recorded in the final spectrum and can be used, in accordance with the ^{40}K photopeak, for auto-energy calibration of the measuring system up to the linear approximation, as will be explained later.

Moving to the next step, one can calculate the number of photons that hit the sensor surface per unit time. Taking an infinitesimal surface ds lying on the x–y plane at point (0,0,0), the photons that hit the surface in time dt from one side are those that lie in the semi-sphere with center (0,0,0), radius $c\cdot\mathrm{d}t$ and $z<0$ (c = speed of light) and they move forward the surface. Then the number of photons f that hit the surface ds from one side in time dt can be found to be

$$\frac{\mathrm{d}^2 f}{\mathrm{d}s\,\mathrm{d}t} = \tfrac{1}{4}\rho c \quad (8)$$

where ρ is the total density of photons and c is the speed of light. As expected, this number depends only on the speed of photons and not on their energy. The distribution of angles that the photons hit the surface can be calculated too:

$$p(\theta) = \tfrac{1}{2}\sin\theta\cos\theta. \quad (9)$$

The next step in the folding procedure is to calculate the energy deposited (sensed) by a photon that enters the sensor. This is done by Monte-Carlo simulation in the following way. The sensor contains a cylindrical NaI crystal

surrounded by a waterproof cover as shown in Fig. 2. Thus, we have three areas for the simulation, i.e. the seawater, the cover and the NaI crystal. The cross sections for all scattering mechanisms are calculated using the XCOM software for all the three areas [11]. The photons are represented as objects, which contain their energy E, position p and direction of movement v. The movement of photons between the three areas is calculated with the following algorithm. First a random point in the sensor surface is selected. A photon is placed in that point with a direction that follows the distribution of angles given by Eq. (9). The algorithm starts with the generation of a random number in order to decide if the photon can reach the closest area interface. If yes, the photon is moved according to its direction to the interface and the algorithm starts again. If not, a second random number is generated in order to find the point at which a scattering will occur. After moving the photon to that point, a new random number is generated in order to decide what kind of scattering will occur. Then, the energy and direction of photon is changed and the algorithm starts again. Every time that an amount of energy is transferred to an electron inside the NaI(Tl) crystal, it is considered that this energy will be sensed. The algorithm stops when the photon disappears due to photoelectric effect. Then the total energy sensed by the NaI(Tl) crystal is recorded. A new point in the sensor surface is selected and the algorithm starts again. After an adequate number of iterations (between 20,000 and 30,000), a spectrum with sufficient statistics is produced. Note here that all photons are recorded, even those that do not leave any amount of energy in the NaI(Tl) crystal. This is necessary in order to use the response function for quantitative estimations.

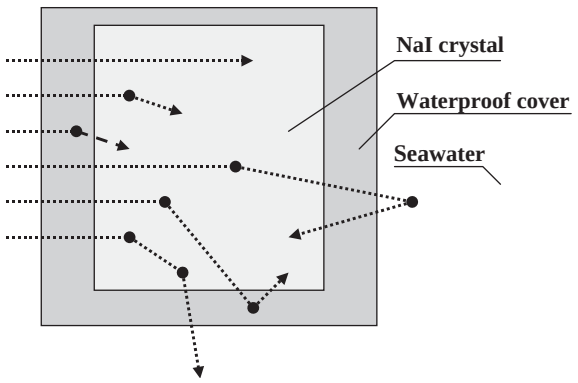

Fig. 2. Monte-Carlo simulation area and different scenarios for the interaction of photons that are handled by the simulation code. Solid circles represent points where the photons change their energy and direction due to Compton scattering and arrows represent points where the photoelectric effect occurs.

At this point we have constructed two sets of arrays, namely D and F. The meaning of their values is: D_{ij} is the density of photons in seawater with energy i given that one photon with energy j is generated per cubic meter and per second. F_{ij} is the number of counts recorded at channel i if one photon with energy j hit the sensor surface per second. It is easily found that the matrix product $S = FD$ has the meaning: S_{ij} is the number of counts at channel i that is recorded by the sensor if one photon with energy j is generated per cubic meter and per second in seawater.

The last step in the folding procedure is the calculation of the resolution matrix R. The meaning of this matrix is: R_{ij} is the number of counts at channel i that is recorded if an amount of energy j is deposed in the sensor due to statistical fluctuations. The rows of this matrix have a Gaussian shape. The elements of R are given by the equation

$$R_{ij} = \frac{1}{\sqrt{2\pi}\sigma} e^{-(i-j)^2/2\sigma^2} \tag{10}$$

where

$$\sigma = \frac{\Gamma}{2\sqrt{\ln 2}} \tag{11}$$

and Γ is the full-width at half-maximum of the Gaussian distribution of counts around a photopeak. Γ depends on energy and is assumed to follow the following relation:

$$\Gamma = \sqrt{c_1 + c_2 E} \tag{12}$$

where c_1 and c_2 are constants that are calculated by experimental calibration of the sensor by using various reference gamma ray sources. Then, the jth column of the product $M = RS = RFD$ is the measured spectrum when a photon with energy j is generated in seawater per cubic meter and per second. Fig. 3 shows the theoretical calculated

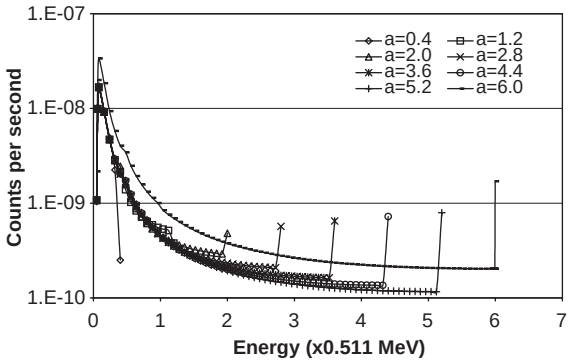

Fig. 3. Ideal NaI spectrum in seawater for different values of initial photon energy. The initial photon energy E is represented in dimensionless form ($a = E/m_0c^2$, where m_0 is the electron rest mass and c the speed of light).

spectra for different initial photon energies and a generation rate of one photon per cubic meter and per second. In this figure, the energy resolution of the sensor was assumed ideal.

3. Experimental setup

The Greek National Center for Marine Research (NCMR) owns and maintains RADAM III sensors constructed by the Norwegian Company OCEANOR. The detector system consists of a detector unit and a power unit shielded by an aluminum and polyester pressure tube. The detector unit is a $3'' \times 3''$ NaI(Tl) detection crystal with a built-in photomultiplier tube, preamplifier, an analog-digital converter, a high-voltage controller, a temperature sensor together with the electronics for data acquisition, storage and transmission. The power unit operated at DC 12 V. The electronics modules are highly miniaturized to fit inside the sensor housing ($80 \times 60\,\text{mm}^2$) and the power consumption is very small ($\sim$1 W). The operating temperature ranges between -10 and $+50^\circ$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(Tl) scintillators. This type of sensor can be used for operational purposes by attaching it on oceanographic buoys. It can operate under severe weather conditions and especially with extreme waves driven by wind forces and high temperature gradients [13].

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. Measurements of the detector efficiency and absolute calibration in Bq/m^3 have also been performed. For this purpose, a calibration tank of 5.5 m^3 volume filled with water has been used [14]. The sensor was mounted in the middle of the tank in order to be surrounded by 1 m of water, which is enough to imitate the real marine environment, due to the high attenuation of the gamma rays in the water. At the bottom of the tank, an electric pump was placed in order to circulate the water to avoid sedimentation, mix the water with the appropriate radionuclides (^{137}Cs and ^{40}K) and achieve homogeneous conditions. After the various settings and calibrations, the sensor was moored on an oceanographic buoy owned by NCMR and placed at North Aegean Sea (location 39.96N, 27.72E). Every 3 h, the measured spectrum was transmitted using GSM telephony and Inmarsat C satellite communication.

4. Results and discussion

In order to test the validity of the calculated response function, we tried to estimate the salinity of seawater from the measured spectra. More precisely, since potassium constitutes 1.1% of the salts that are diluted in seawater, the salinity is given by

$$S = 90.9\,\frac{C[\text{K}]}{\rho} \tag{13}$$

where C[K] is the concentration of potassium (g/l) and ρ is the density of seawater (1027 g/l). On the other hand, the abundance of ^{40}K is 0.0117%. Then, the salinity is given as

$$S = 756.5\,C[^{40}\text{K}] \tag{14}$$

where $C[^{40}\text{K}]$ is the concentration of ^{40}K in seawater (g/l) which can be found by the measured

spectrum and the calculated response function. On the other hand, the salinity of the seawater can be very easily calculated by conductivity measurements. Fig. 4 shows the measured spectrum in comparison with the spectrum produced by the simulation code, taking into account only the contribution of ^{40}K. Note here that although the simulated spectrum contains only the contribution of ^{40}K, the measured spectrum also contains a large amount of natural radionuclides (like U and Th series) which are present in the seawater. Thus, we are able to compare the two spectra only in the area near the ^{40}K photopeak, where the contribution of the background radiation is about one order of magnitude smaller compared to the ^{40}K contribution. The simulated spectrum was scaled in order to fit the measured spectrum at the photopeak located at 1461 keV. The scaling coefficient corresponds to a salinity of the seawater of 36.4 psu while the measured salinity was 36.6 psu. This agreement validates the simulation result concerning the quantitative estimation of the emission of ^{40}K in seawater. Fig. 5 shows the estimated and measured salinity

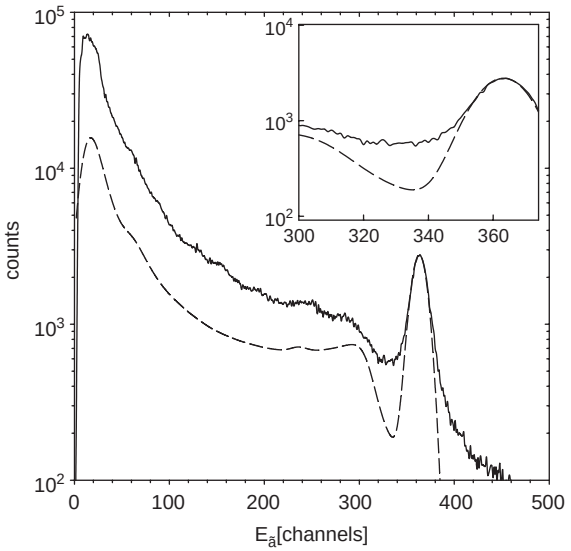

Fig. 4. Gamma ray spectrum acquired in the field using the RADAM III (Oceanor) NaI(Tl) (3″ × 3″) sensor (solid line) and simulated spectrum (dashed line) for the 1461 keV (^{40}K) gamma ray contribution considering that the salinity of seawater is 36.4 psu. The measuring time is 3 days for both spectra.

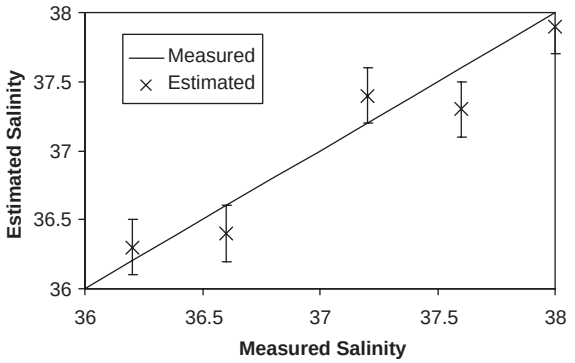

Fig. 5. Measured and estimated salinity of seawater. The error bars height is less than 2% of the estimated value.

of seawater for different seasons over the year. In all cases, the estimation error was less than 2% of the estimated value. These results show that the calculated transfer function can be used for accurate quantitative estimations. Returning to Fig. 4, it is clear that although there is a good agreement between the measured and calculated spectra around the photopeak of ^{40}K, there is an underestimation of the calculated spectrum in the area between the photopeak and the Compton edge as it is shown in the embedded enlarged figure. This is probably caused by the use of a Gaussian-like function to simulate the resolution of the sensor. A more accurate function has to be used because the energy resolution calibration of the sensor shows that the shape of the photopeak is fitted better at energies below $E_0 - \Gamma/2$ with an exponential-like function, where E_0 is the energy at the photopeak and Γ is the full-width at half-maximum. The use of such a function will improve significantly the simulation in the NaI(Tl) crystal at the energy interval between the Compton edge and the associated photopeak energy.

Both spectra, simulated and measured, show a peak at 50 keV, which is caused by the distribution of the energy of the photons in seawater, as mentioned earlier. This peak and the natural decay of ^{40}K can be used for energy auto-calibration of the sensor. More precisely, there will always be two dominant peaks in a measured spectrum in seawater: at 50 and 1461 keV (^{40}K). These two peaks are adequate for the energy calibration, assuming that there is a linear and uniform

dependence of measured energy on recording channel of the sensor. The exact location of the peak around 50 keV can be calculated from Eq. (7) by setting the derivative of $\lambda(a_t)$ equal to zero. The results show a slight dependence on the scattering environment (47.5 keV for salinity 30 psu, which is increased to 49 keV for salinity 40 psu) and are independent of the initial photon energy. The detailed calculations will be presented elsewhere [15].

Finally, since the measured spectrum is a sum of convolutions of the transfer function with delta functions, the knowledge of the transfer function can be used with well-known methods, such as filtering algorithms, for the qualitative and quantitative estimation of different sources of radioactivity. The basic idea of these methods is to minimize the noise of statistical origin, taking into account the basic characteristics of the recorded signal, like the Gaussian shape and the change of the full-width at half-maximum with energy [16]. After extraction of the first photopeak (at the higher energy), the Compton continuum is calculated (from reference spectra or model curves) and subtracted in order to extract the next photopeak. With an exact knowledge of the transfer function of the measuring system, these methods can be applied in underwater gamma spectroscopy too. A comparative study of the applicability of these methods using the calculated transfer function is at present under investigation.

5. Conclusions

The response function of a NaI(Tl)-based system has been estimated theoretically by using both analytical and Monte-Carlo techniques and taking into account all the responsible processes and interactions of gamma rays in water as well as in the material of the detector and its housing. The calculated response function was tested for quantitative estimation of ^{40}K in the North Aegean Sea. The validation of the method was performed by comparing the measured salinity of the seawater with the salinity value produced by the calculated concentration of ^{40}K. Moreover, the analytical calculation of the distribution of photon energy in seawater shows that a photopeak around 50 keV will always be present in the measured spectrum , which with the ^{40}K photopeak can be used for auto-energy calibration of the measuring system.

Finally, the calculated transfer functions can be used as reference spectra to well-known filtering techniques for the qualitative and quantitative estimation of different sources of radioactivity in seawater.

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