



A two-point in situ method for simultaneous analysis of radioactivity in seawater and sediment

Jun-Ho Lee¹ · Jong-In Byun² · Dong-Myung Lee^{1,2}

Received: 19 June 2019 / Published online: 19 September 2019
© Akadémiai Kiadó, Budapest, Hungary 2019

Abstract

A new in situ radioactivity analysis method was developed to determine the fractional contributions of gamma-ray emitters in seawater and sediment from total measured counts. A semi-empirical formula to determine the fractional count contributions was derived using the variation characteristics of the gamma-ray attenuation rate and geometrical efficiency with the measurement points. The proposed method was employed to make in situ gamma-ray measurements using a CeBr₃ detector for radioactivity analysis of seawater and sediment at a coastal area with a relatively high tidal range. The full energy peak efficiency of the detector at measurement site was obtained using the GEANT4 simulation code. The radioactivity concentration of ⁴⁰K in seawater and sediment was determined using the proposed method and laboratory analysis with sampling. The MDA of the in situ measurement for ¹³⁷Cs and ¹³¹I was also estimated assuming they were deposited on the sediment surface-layer. The validation of this method was demonstrated by comparing with the sampling analysis results.

Keywords In situ gamma-ray measurement · Radioactivity · Seawater · Sediment · GEANT4 code

Introduction

Natural and artificial radionuclides can exist in the marine environment through various pathways, such as weathering and recycling of terrestrial rocks, nuclear bomb tests, nuclear power plant accidents, or illegal radioactive waste disposal [1]. These radionuclides can affect the marine biota and human health, and they can also be used as a radioactive tracer in earth and environment science. Evaluating radioactivity in the marine environment is consequently important. Various evaluation methods have been developed over the past several decades. Typically, sampling and laboratory analysis have been used to evaluate the radioactivity evaluation in seawater and sediment. Although this approach can provide accurate radioactivity assessments, it has several disadvantages such as costly and time-consuming for sample collection, transfer to the laboratory, and pretreatment. For

these reasons, an in situ gamma-ray measurement method without sampling has been widely used in some cases.

The first effort to measure radioactivity in the marine environment was made by the Soviet Union in the late 1950s. Since then many studies on in situ gamma-ray spectrometry in the marine environment have been reported, as presented in the review paper by Jones [2]. Using a static detector system fixed in the sediment, a gamma-ray spectrometry methodology for in situ measurement on the seabed has also been proposed [3–5]. In this method, the seawater contribution was considered by placing the detector in conditions where the influence of the radionuclide in the sediment could be neglected. The system has been effectively utilized for in situ measurement on the seabed for various purposes [5]. However, there are circumstances where the effects of the radionuclide in the sediment cannot be neglected. Therefore, a new method is needed that can evaluate the individual contributions of the radionuclides present in both seawater and sediment.

The aim of the present study is to develop a method of the in situ gamma-ray measurement for the analysis of radioactivity concentration, evaluating the fractional contributions of gamma-ray counts from radionuclides present in both seawater and sediment. For this purpose, a semi-empirical formula with Monte Carlo (MC) simulation was derived to

✉ Jong-In Byun
k975bj@kins.re.kr

¹ University of Science and Technology, 217 Gajeong-ro, Yuseong-gu, Daejeon 34113, Republic of Korea

² Korea Institute of Nuclear Safety, 62 Gwahak-ro, Yuseong-gu, Daejeon 34142, Republic of Korea

consider geometrical efficiency and attenuation differences for seawater and sediment. The method was validated by comparing in situ measurement with sampling analysis for the site, which had a relatively high tidal range. In this paper, the theoretical approach for the proposed method is introduced, and the validation process and the feasibility are discussed.

Theoretical approach

Since the characteristics of the source term in seawater and sediment are different from each other, for in situ gamma-ray spectrometry in the marine environment it is important to evaluate the contribution of radionuclides present in both the seawater and sediment entering the detector. The contributed fraction of gamma-rays count from seawater and sediment can be different, because the thickness of the seawater layer between the detector and seabed can vary. Therefore, the contribution can be determined semi-empirically from gamma-ray measurements taken at different heights from the seabed. The response function of the detector for the photon flux in the measurement setup can be determined by experiment or simulation.

The radioactivity concentrations A_{w,h_i} of the radionuclide in seawater at the detector height h_i above flat sediment can be expressed as

$$A_{w,h_i} = N_{w,h_i} / \varepsilon_{w,h_i} \cdot P_r \quad (1)$$

where N_{w,h_i} is the net count rate (cps) measured from radionuclide in seawater at the detector height h_i . ε_{w,h_i} is the volume efficiency (efficiency \times volume (L)) of the radionuclide in seawater at the detector height h_i . P_r is the gamma-ray emission intensity (Bq^{-1}). Depending on the detector height from the seabed, the net count rate N of the radionuclide in the seawater coming into the detector and detection efficiency ε can be different. However, the radioactivity concentration A_w (Bq L^{-1}) of the radionuclide in the seawater is constant regardless of the detector height. Therefore, Eqs. 1 and 2 can be expressed as follows.

$$A_w = A_{w,h_1} = A_{w,h_2} \quad (2)$$

$$A_w = N_{w,h_1} / \varepsilon_{w,h_1} \cdot P_r = N_{w,h_2} / \varepsilon_{w,h_2} \cdot P_r \quad (3)$$

For the radioactivity concentration A_s (Bq kg^{-1}) of the radionuclide in the sediment, the same principle can be applied, as follows.

$$A_s = A_{s,h_1} = A_{s,h_2} \quad (4)$$

$$A_s = N_{s,h_1} / \varepsilon_{s,h_1} \cdot P_r = N_{s,h_2} / \varepsilon_{s,h_2} \cdot P_r \quad (5)$$

where A_{s,h_i} is the radioactivity concentrations of the radionuclide in sediment at the detector height h_i above the seabed. N_{s,h_i} is the net count rate (cps) measured from the radionuclide in the sediment at the detector height h_i . ε_{s,h_i} is the mass efficiency (efficiency \times mass (kg)) of the radionuclide in the sediment at the detector height h_i . Equations 3 and 5 can be rewritten as

$$R_w = N_{w,h_1} / N_{w,h_2} = \varepsilon_{w,h_1} / \varepsilon_{w,h_2} \quad (6)$$

$$R_s = N_{s,h_1} / N_{s,h_2} = \varepsilon_{s,h_1} / \varepsilon_{s,h_2} \quad (7)$$

where R_w and R_s mean the ratio of detection efficiencies or the full peak count rates for the radionuclides in seawater or sediment at height h_1 and h_2 above the seabed. For the count rates of radionuclide measured using a detector in the marine environment, this indicates the contribution ratio to total count rate from the radionuclide present in both seawater and sediment.

The ratio R_T of the total count rate obtained simultaneously for the radionuclide present in both seawater and sediment at the detector height h_1 and h_2 above seabed can be expressed as

$$R_T = \frac{N_{T,h_1}}{N_{T,h_2}} = \frac{N_{s,h_1} + N_{w,h_1}}{N_{s,h_2} + N_{w,h_2}} \quad (8)$$

where N_{T,h_1} and N_{T,h_2} are the total count rate obtained simultaneously for the radionuclide present in both seawater and sediment at the detector height h_1 and h_2 . Substituting Eqs. 6 and 7 into Eq. 8, the ratio of the count rate for the radionuclides present in both seawater and sediment at the detector height h_1 or h_2 can be expressed as

$$\frac{N_{s,h_1}}{N_{w,h_1}} = \frac{(1 - R_T/R_w)}{(R_T/R_s - 1)} \quad \text{or} \quad \frac{N_{s,h_2}}{N_{w,h_2}} = \frac{(1 - R_w/R_T)}{(R_s/R_T - 1)} \quad (9)$$

Using Eq. 9, the individual count rates for the radionuclide present in both seawater and sediment can be determined from the total count rate. That is, R_w , R_s , and R_T are required to evaluate the fractional contribution from the total count rate of the radionuclide simultaneously measured in both seawater and sediment. Each of the count rate obtained using Eq. 9 can be applied to Eqs. 3 and 5 to calculate the radioactivity concentration for the radionuclide in seawater and sediment.

Application

System configurations

To validate the theoretical approach for in situ gamma-ray spectrometry in the marine environment, a 2"Ø × 2" CeBr₃ (Scionix-Holland, Inc.) detector was used, with a FWHM (Full Width at Half Maximum) of about 4.2% for 661.6 keV gamma-rays of ¹³⁷Cs. The CeBr₃ crystal was coupled to the 2"Ø × 2" photomultiplier tube (Hamamatsu series). To acquire and analyze the spectra, the ORTEC digiBase-E and MAESTRO software were used.

The watertight enclosure housing was designed using 5 mm thick Acetal with the chemical formula CH₃CH(OC₂H₅)₂ material, to protect the detector and its electronics from the seawater. Acetal is a material with good mechanical strength properties and excellent resistance to marine environments [6]. Polyurethane foam with a thickness of 1 cm was used in the detector head to protect the CeBr₃ crystal in the Acetal housing. The cable used for power supply and data transmission was also made of waterproof materials and sufficient flexibility. To reliably seal the enclosure housing, an O-ring and high vacuum grease were used. A stainless steel tripod was used to hold the detector at a constant distance above the sediment surface. Each leg of the tripod in contact with the sediment surface was made of a circular plate with a radius of 4 cm and a thickness of 0.5 cm to prevent the detector from sinking into the sediment. Also, three 1 m long anchors were attached to each leg of the tripod to prevent the tripod from vibrating or being moved by tidal currents during measurement.

In situ measurement and sampling analysis

We have chosen a reference site whose tidal sea level increased and decreased periodically with time. This phenomenon facilitated the detector set-up, the measurement, and seawater and sediment sampling in the marine environment without a diver and ship. In particular, it allowed the accurate characterization of the sediment according to depth. The measurement was carried out on the Yellow Sea seacoast near Gusipo port in Korea (35°26'56.8"N 126°26'06.5"E), where the low tide and high tide occur every 6 h. In this area, the tide range between low tide and high tide is about 6 m for the largest and about 2 m for the smallest. Near the coast, the sediment surface is exposed at low tide and the sea level is about 2.5 m at high tide. Information about tides in this area can be found in the KHOA Smart Tide Forecast prepared by the Korea hydrographic and oceanographic agency [7]. The in situ

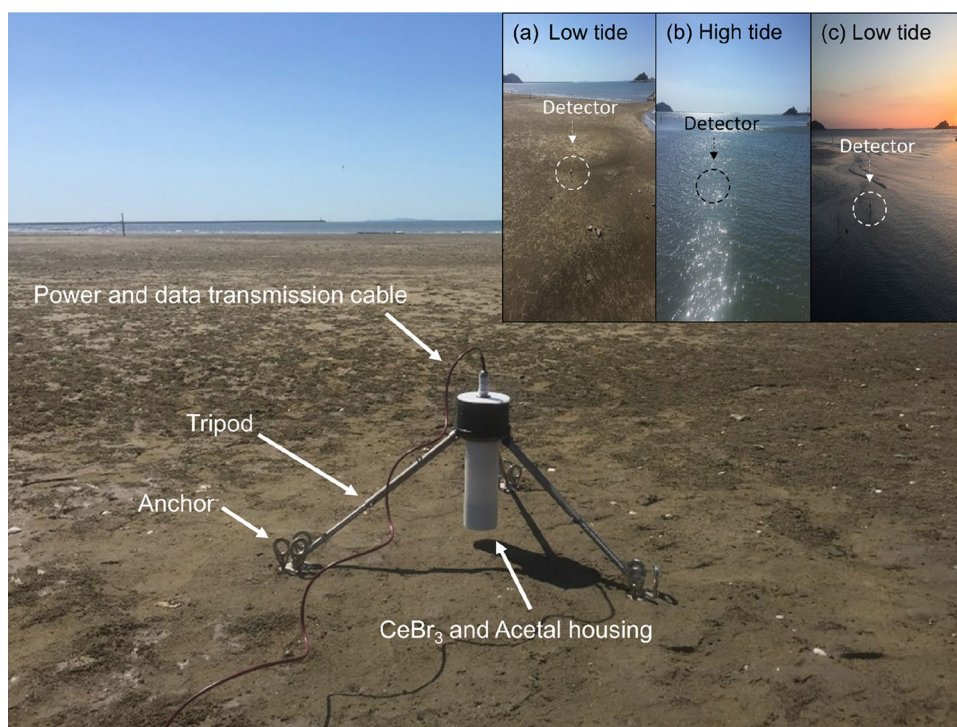
gamma-ray measurement for R_7 in Eq. 9 was performed by measuring ⁴⁰K in the marine environment, which is relatively abundant compared to other radionuclides in both seawater and sediment. The CeBr₃ detector has internal background peaks produced by ²²⁷Ac, which interfere with the 1460.8 keV for ⁴⁰K [8]. Therefore, a self-background measurement of the CeBr₃ detector was carried out with a low-level lead shield (< 5 Bq kg⁻¹ for the 50 mm thick inner lead layer and < 100 Bq kg⁻¹ for the 100 mm thick outer lead layer). The results from the self-background measurement result were used to subtract the overlapping internal background peaks from the ⁴⁰K measured in the marine environment [9].

The distances of 5 and 12 cm between the detector and the sediment surface were chosen for h_1 and h_2 in this study because the difference in the gamma-ray flux of ⁴⁰K from the sediment at a the gap of 7 cm was approximately double assuming that the depth distribution of ⁴⁰K in the sediment is homogeneous. The theoretical background of these gamma-ray fluxes is described by ICRU 53 report [10]

At low tide, the stainless steel tripod with the detector was placed on the sediment surface, as shown in Fig. 1. The distance between the detector and the sediment surface was fixed at 5 cm. The in situ measurement was performed on about an hour before the maximum high tide which is the smallest change in sea level. The measurement time was 7000 s. During the in situ measurement, a seawater sample of 20 L was taken near the measurement point for analysis in the laboratory (sample number 1). Then, at low tide when the sediment surface was again exposed, the distance between the detector and sediment surface was adjusted from 5 to 12 cm. Again, the in situ measurement was performed on about an hour before the maximum high tide. During in situ measurement, 20 L of seawater sample was also taken near the measurement point (sample number 2) to check for variation in the radioactivity concentration of ⁴⁰K for seawater in initial and last measurements.

To maintain the density and water content of the sediment when the in situ measurement was taken, the sediment sample was collected when the sea level was about 3 cm above the sediment surface, by coring to a depth of 30 cm at the center of the measurement site using a cylindrical sampler with a diameter of 14.8 cm. To prevent seawater and sediment from escaping beneath the coring sampler while collecting the sediment, the bottom of the coring sampler was covered with 1 mm thick galvanized steel plate. Immediately after sampling, the collected sample was divided into five depths as follows: 0–5, 5–10, 10–15, 15–20 and 20–30 cm. In the laboratory, samples were ground to a particle size of less than 200 µm, respectively. Then, each sample was used to fill a cylindrical plastic bottle. The samples were measured using a BEGe (Broad Energy Germanium) detector with

Fig. 1 The CeBr_3 detector and its housing at the low tide in the field; **a** before measurement, **b** during measurement, and **c** after measurement



a relative efficiency of 65%. Then, the gamma-ray energy spectra were analyzed to determine the radioactivity concentration for ^{40}K . Photopeak efficiency calibrations of the BEGe detector were carried out using full energy peak counts from a standard reference source containing ^{241}Am , ^{109}Cd , ^{57}Co , ^{139}Ce , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{88}Y and ^{60}Co (Eckert and Ziegler series) which had geometry identical to that of the sediment sample. True coincidence summing and self-attenuation corrections were performed using the GESPECOR software [11]. The calibrations of the BEGe detector were validated with IAEA 447 reference materials.

For seawater samples, HCl (Hydrochloric acid) was added to recovery the ^{40}K adsorbed on the inner wall of the sample bottle. In the laboratory, the suspended solid materials contained in the seawater were removed using filter paper. The filtered seawater was transferred to a 10 L polyethylene Marinelli beaker to get enough counts. Then, the measurement was carried out using a coaxial type HPGe detector with a relative efficiency of 30% to analyze the radioactivity concentration for ^{40}K in seawater. To directly calibrate the radioactivity concentration of ^{40}K (natural abundance 0.0117%) in seawater and reduce the radioactive wastes produced by the standard reference source, the KCl with a purity of 99.5% was used. 250.1 g of KCl were diluted in a polyethylene Marinelli beaker whose geometry was identical to that of the seawater sample, resulting in 4.04 kBq total activity. The used KCl was validated with the standard reference source used to calibrate the BEGe detector.

Efficiency calibration

To obtain R_w and R_s in Eq. 9, the MC simulation with the GEANT4 (GEometry ANd Tracking) code was implemented by performing the geometrical setup of the detector and source volume. The GEANT4 code is a simulation toolkit written in the C++ programming language, which simulates the physical interactions that occur when particles pass through a material. Although the GEANT4 code has higher initial entry barriers to use than other simulation codes because the users must code directly, it is a versatile and flexible program, and available under an open source license [12–14].

The geometric modeling of the CeBr_3 and its housing was based on the real dimensions and materials provided by the manufacturer, as shown in Fig. 2. A 5 mm thick Acetal housing was 9.3 cm in diameter, 35.5 cm in length with a density of 1.4 g cm^{-3} . The polyurethane foam with a thickness of 1 cm was 7.7 cm in diameter, 6.2 cm in length and a density of 0.025 g cm^{-3} . The photomultiplier tube has dimensions of 5.08 cm in diameter, 9.8 cm in length and a density of 0.8 g cm^{-3} . For the digiBase-E, the dimensions were 6.2 cm in diameter, 11 cm in length and a density of 1.1 g cm^{-3} . The photomultiplier tube and digiBase-E applied mean density and were assumed to be made of Pyrex glass and bakelite, respectively. To validate these system geometries in the GEANT4 code, the experiments were performed using the point like sources of ^{151}Eu (121.8 keV), ^{137}Cs (661.6 keV) and ^{60}Co (1173.2 and 1332.5 keV), respectively. The point

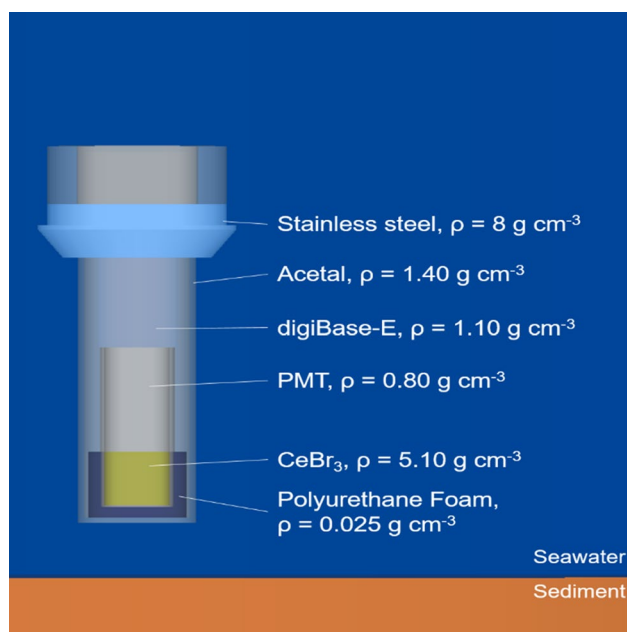


Fig. 2 Geometric modeling of the CeBr_3 detector and its housing using the GEANT4 code

sources were measured at points (on the front and sides of the housing) of about 30 cm from the center of the crystal in the laboratory. The same was also simulated in the GEANT4 code. Then, the detection efficiencies obtained from the experiments were compared with those from the simulation.

The source geometry can be broken down into two types: seawater and sediment. Both source geometries were modeled as cylinders using the sampling information from the in situ measurement site. When modeling the geometry of seawater, the density used was applied as an average of two samples taken in the field because there is little difference in the density of seawater samples. The simulation of the detection efficiency for the radionuclides in the sediment were implemented using three different depth profiles for gamma-ray emitter distributions in the sediment: (1) the real depth profiles of ^{40}K obtained by sample collection and laboratory analysis at the in situ measurement site to validate the proposed method, (2) the homogenous distribution of ^{40}K under the condition of unknown real depth profiles, and (3) freshly deposited source distribution on the seabed surface. The chemical composition of sediment used in the simulation was taken from Al-Masri et al. [15] because for gamma-ray energy higher than around 100 keV, mass attenuation coefficients do not significantly depend on the sample type.

Since the simulation requires a finite volume, the effective volumes of the source terms, which is the largest volume where the photons can reach the detector, were determined. For seawater, the effective radius was determined by varying the radius of the seawater cylinder at a detector height of 12 cm

from the sediment surface. The height of the seawater cylinder was fixed at 100 cm from the top of the Acetal housing at a detector height of 12 cm. Because of the attenuation of seawater, the effective radius of the sediment was conservatively determined to be equal to the effective radius of the seawater. The effective depth of the sediment was determined by varying the depth of the sediment cylinder at a detector height of 5 cm from sediment surface while the radius of the sediment cylinder was fixed at 100 cm. A single energy of 3000 keV, which is the highest energy among the natural gamma-ray, was used for the gamma source and set to be homogeneously distributed through the source volume. It was considered the effective volumes of seawater and sediment that were determined using an energy of 3000 keV would also be satisfactory for the below energies of 3000 keV. The number of the beams was kept constant at 3×10^8 .

Results and discussion

Efficiency calibration result

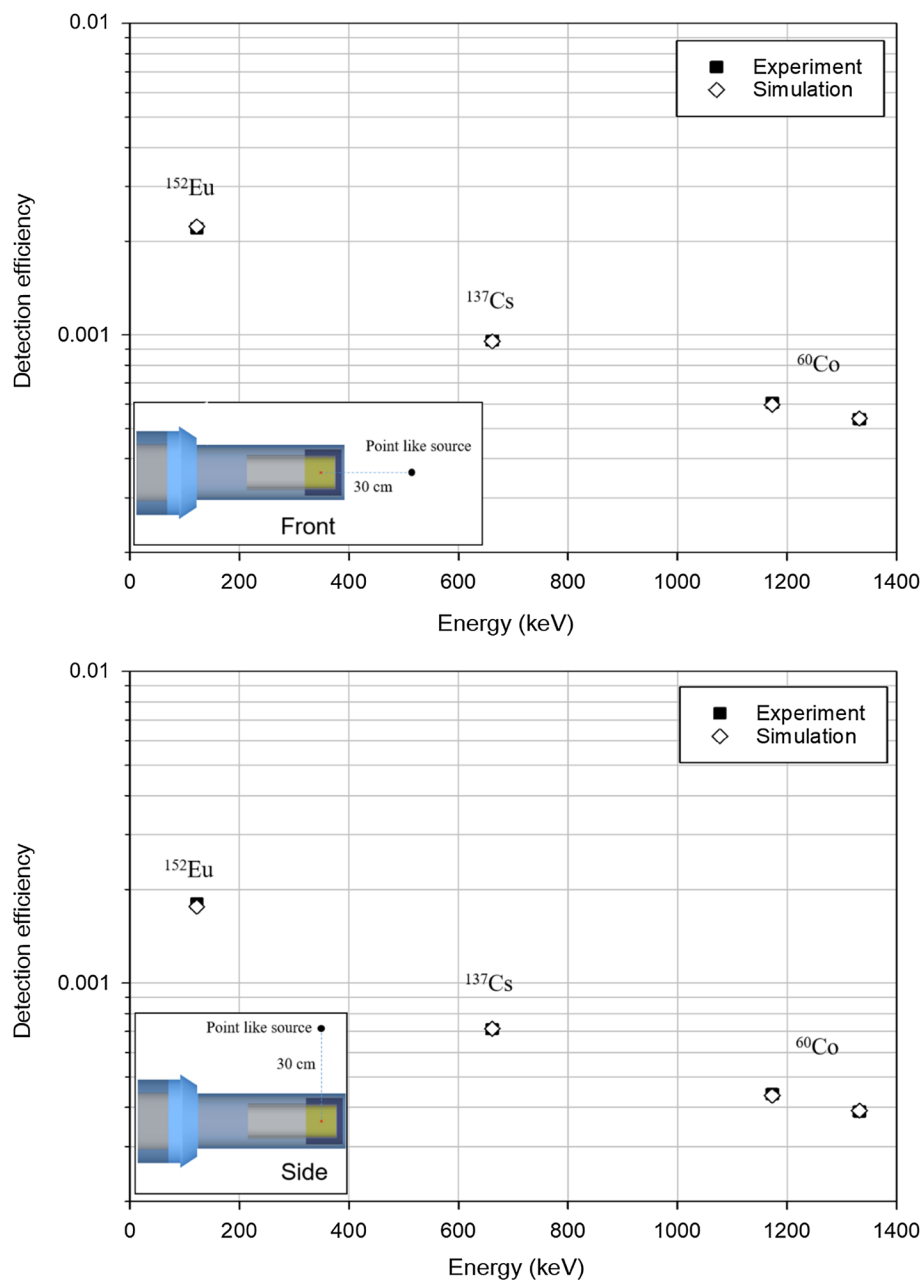
A comparison of the detection efficiency results from the experiments and simulations are shown in Fig. 3, in the energy range of 121.8–1332.5 keV. The differences in the detection efficiency between experiment and simulation were less than 3%, which demonstrates the validity of the models by the GEANT4 code.

Figure 4a shows the results for volumetric detection efficiency when varying the radius of the seawater cylinder. The volumetric detection efficiency forms a plateau for radii from 80 to 140 cm. As a result, the source radius of seawater was conservatively determined to be 100 cm on the plateau. The source radius of the sediment was also determined to be the same as that of seawater. Figure 4b shows the results for volumetric detection efficiency when varying the depth of the sediment cylinder. The volumetric detection efficiency reaches a plateau at a depth of 20 cm. When modeling the source volume of the sediment, the depth of the sediment was determined to be equal to the sampling depth 30 cm. The source geometries of seawater and sediment are shown in Fig. 5. Based on these, the ratio R_W and R_S of the volumetric detection efficiencies for ^{40}K (1460.8 keV) in seawater and sediment were obtained. In Fig. 5a, the efficiency of the sediment to validate the proposed method was determined as the total efficiency ε_T as follows

$$\varepsilon_T = \sum_{i=1}^5 \frac{A_i}{A_T} \cdot \varepsilon_i \quad (10)$$

where i is the number of sediment layers according to depth (1: 0–5 cm, 2: 5–10 cm, 3: 10–15 cm, 4: 15–20 cm and 5: 20–30 cm), A_i is the radioactivity in the corresponding sediment layer obtained from the sampling analysis of the

Fig. 3 Comparison of the detection efficiency from experiments and simulations



sediment, A_T is the sum of radioactivity in all sediment layers and ε_i is the efficiency for the corresponding sediment layer by the GEANT4 code. This total efficiency ε_T was calculated at the detector heights of 5 and 12 cm, respectively.

In a practical application, since the radioactivity analysis by in situ measurement is usually performed without sampling for source distribution profiling, the source term of the sediment was simply modeled using just the mean water content of 26.2% and density of 1.62 g cm^{-3} , as shown in Fig. 5b. This means the depth distribution of the radionuclide in the sediment is homogeneous. The R_S in Eq. 7 was determined using the simulation results $\varepsilon_{s,5\text{cm}}$ and $\varepsilon_{s,12\text{cm}}$ from this sediment model.

In situ measurement and sampling analysis results

For seawater samples, the difference in the radioactivity concentration of ^{40}K in the two samples was within 2%. It can be seen that the radioactivity concentrations for ^{40}K in seawater obtained in the two in situ measurements are in good agreement. The weighted average value of the radioactivity concentration for ^{40}K in seawater was $11.1 \pm 0.3 \text{ Bq L}^{-1}$. For sediment samples, the radioactivity concentration for ^{40}K varied from 585 ± 8 to $685 \pm 10 \text{ Bq kg}^{-1}$ -wet depending on the depth, as shown in Table 1. The weighted average value $636 \pm 5 \text{ Bq kg}^{-1}$ -wet represents that the total ^{40}K radioactivity per total mass in all layers. The ^{40}K radioactivity values

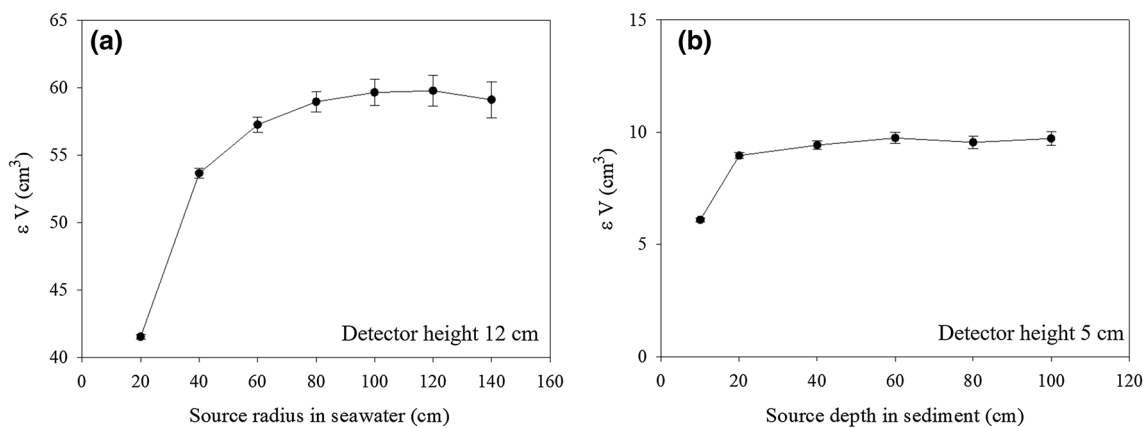


Fig. 4 The volumetric detection efficiency for various source terms in the GEANT4 code

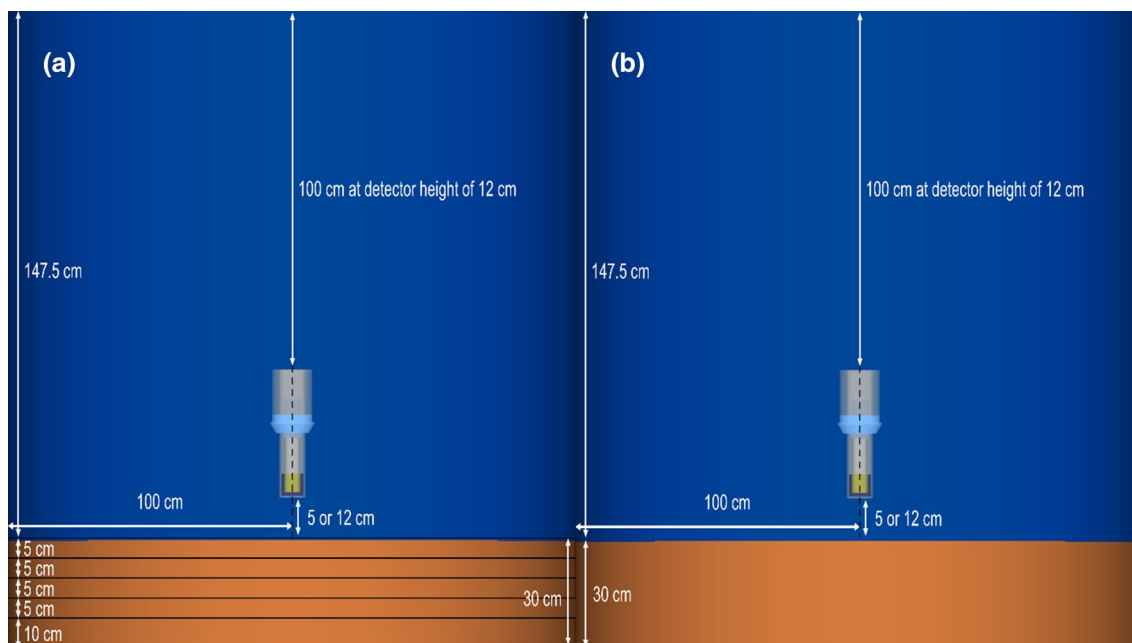


Fig. 5 The geometry of the source term used in the GEANT4 code. **a** The model using the sample information with sediment depth, **b** the model without the sample information assuming that the depth distribution of the radionuclides in the sediment is homogeneous

Table 1 Sampling information and radioactivity analysis results for ⁴⁰K in sediments

Depth (cm)	Density (g cm ⁻³ -wet)	Water content (%)	Radioactivity concentration for ⁴⁰ K, <i>k</i> = 1 (Bq kg ⁻¹ -wet)
0–5	0.80	27.3	685 ± 10
5–10	1.55	20.2	633 ± 9
10–15	1.92	31.9	585 ± 8
15–20	1.93	23.6	658 ± 9
20–30	1.92	24.0	642 ± 9

of each layer were used as weighting factors to calculate the total efficiency in Eq. 10, and to validate the proposed method. Figure 6 shows the CeBr₃ gamma-ray energy spectra measured with distances of 5 and 12 cm distance between the detector and the sediment surface in the marine environment. In these spectra, the *R_T* based on 12 cm was determined using the peak count rate of ⁴⁰K. Applying these ratios *R_W*, *R_S* and *R_T* to the two in situ measurements method in Eq. 9, the radioactivity concentrations for ⁴⁰K in seawater and sediment at a detector height of 12 cm were

Fig. 6 CeBr_3 spectra in the marine environment with detector height

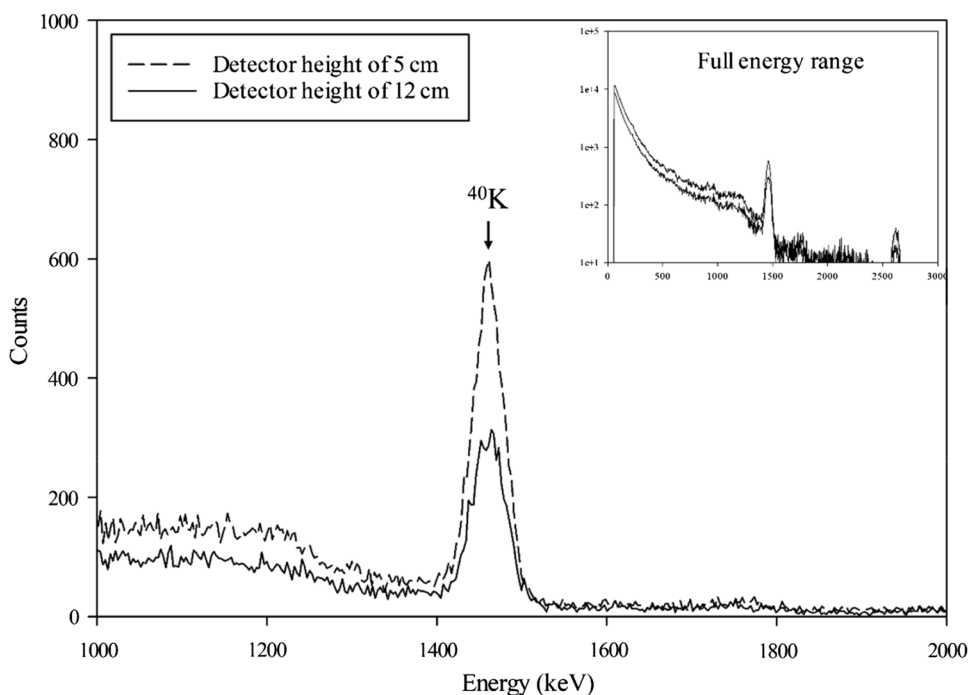


Table 2 Comparison of in situ CeBr_3 and sampling analysis of the radioactivity concentration of ^{40}K in seawater and sediment

Environment	Radioactivity concentration of ^{40}K ($k=1$)		In situ method/Sampling analysis	
			With real depth profiles	With homogeneous distribution
	In situ method (this study)	Sampling analysis		
Seawater (Bq L^{-1})	11.4 ± 2.5	13.7 ± 2.3	1.03 ± 0.23	1.23 ± 0.21
Sediment (Bq kg^{-1} -wet)	575 ± 29	563 ± 28	0.90 ± 0.05	0.88 ± 0.04

calculated from the total count rate of ^{40}K present in both seawater and sediment. The results were compared with those from the sampling analysis, as shown in Table 2. The ratio of the radioactivity concentrations for ^{40}K between in situ with sample information and sampling analysis showed 1.03 ± 0.23 in the seawater and 0.90 ± 0.05 in the sediment, respectively. The radioactivity concentrations for ^{40}K by the in situ method without sample information were $13.7 \pm 2.3 \text{ Bq L}^{-1}$ in seawater and $563 \pm 28 \text{ Bq kg}^{-1}$ in sediment. The ratios between the in situ method without sample information and the sampling analysis were 1.23 ± 0.21 in the seawater and 0.88 ± 0.04 in the sediment, respectively, which show larger than the ratio results by in situ using sample information. This means that if the depth distribution of the radionuclide in the sediment is known, the two in situ measurements method we have presented is well suited to determining the radioactivity of radionuclides in both seawater and sediment. Even if the depth distribution of the radionuclides in the sediment is not available, as is typical in

Table 3 MDA values of in situ CeBr_3 for ^{131}I and ^{137}Cs in radiation emergency situation (with a detector height of 5 cm from the sediment surface)

Nuclide	Energy (keV)	MDA ^a	
		Seawater (mBq L^{-1})	Sediment (Bq m^{-2})
^{131}I	364.0	684	521
^{137}Cs	661.6	634	423

^aCurrie method with a 95% confidence level

practical applications, this method is satisfactory for determining the radioactivity in marine environments. Also, in accident situations, such as when radionuclides leak into a marine environment, this method seems to be more effective because the depth distribution in the sediment is not important for artificial radionuclides deposited on the sediment surface at the beginning of the accident.

Table 3 shows the MDA (Minimum Detectable Activity) of ^{131}I and ^{137}Cs in the spectrum measured at a detector height of 5 cm, assuming a radiation emergency situation. The MDA was calculated as

$$MDA = \frac{2.71 + 4.65 \cdot \sqrt{B}}{t \cdot K_i \cdot P_r \cdot \epsilon \cdot V} \quad (11)$$

where B represents the background counts for the corresponding energy region, t is measurement time, K_i is decay correction factors, P_r is the gamma-ray emission intensity, ϵ is the detection efficiency and V is the volume (L) of the seawater and area (m^2) of the sediment [16]. For detection efficiency, the simulation in seawater was performed assuming that each artificial radionuclide was homogeneous in the seawater. For detection efficiency in the sediment, the simulation assumed that each artificial radionuclide was deposited at 2 cm from the sediment surface.

Conclusion

We have presented a new in situ measurement method to determine the radioactivity concentration in seawater and sediment by determining the fractional contribution from both seawater and sediment in the total counts of radionuclides. The ratio of the total count rate for ^{40}K , which is the natural radionuclide abundant in seawater and sediment, was obtained by performing two in situ gamma-ray measurements with different detector heights in the marine environment. The detector set-up, and sampling of seawater and sediment were carried out at high and low tides. The ratios of detection efficiencies for the seawater and sediment source terms were obtained using the GEANT4 code. The source term of the sediment was modeled using the density and water content according to the sediment depth. For the calculated detection efficiency from the simulation with the density and water content according to the sediment depth, the radioactivity for ^{40}K in samples analyzed in the laboratory was used as the weight factor. The radioactivity concentration for ^{40}K in seawater and sediment obtained with the new in situ method was compared with the results from the sampling analysis.

In conclusion, the two in situ measurement method was proven to be a reliable approach for evaluating the fractional contribution of any radionuclide present in both seawater and sediment by in situ gamma-ray spectrometry. The present study confirmed that this method was satisfactory based on the knowledge of the depth distribution of radionuclides in the sediment, and that it can be used in practical applications. This method can also be used at the detector heights other than those selected in this study, and can also be applied to other detectors, such

as the $\text{LaBr}_3(\text{Ce})$ and portable semiconductor detectors. The method will be helpful in the evaluation of artificial radionuclides released into the marine environment due to nuclear accidents or radiological emergency. As a further study, it would be suggested to measure and evaluate areas with artificial radionuclides by in situ gamma-ray spectrometry applied this method.

References

1. Tsabaris C (2008) Monitoring natural and artificial radioactivity enhancement in the Aegean Sea using floating measuring systems. *Appl Radiat Isot* 66:1599–1603
2. Jones DG (2001) Development and application of marine gamma-ray measurements: a review. *J Environ Radioact* 53:313–333
3. Androulakaki EG, Kokkoris M, Skordis E, Fatsea E, Patiris DL, Tsabaris C, Vlastou R (2016) Implementation of FLUKA for γ -ray applications in the marine environment. *J Environ Radioact* 164:253–257
4. Androulakaki EG, Kokkoris M, Tsabaris C, Eleftheriou G, Patiris DL, Pappa FK, Vlastou R (2016) In situ γ -ray spectrometry in the marine environment using full spectrum analysis for natural radionuclides. *Appl Radiat Isot* 114:76–86
5. Androulakaki EG, Tsabaris C, Eleftheriou G, Kokkoris M, Patiris DL, Pappa FK, Vlastou R (2016) Efficiency calibration for in situ γ -ray measurements on the seabed using Monte Carlo simulations: application in two different marine environments. *J Environ Radioact* 164:47–59
6. Tsabaris C, Bagatelas C, Dakladas T, Papadopoulos CT, Vlastou R, Chronis GT (2008) An autonomous in situ detection system for radioactivity measurements in the marine environment. *Appl Radiat Isot* 66:1419–1426
7. Korea Hydrographic and Oceanographic Agency (2016) KHOA smart tide forecast and Korea real time database for North–East Asian regional global ocean observing system (NEAR-GOOS). <http://www.khoa.go.kr/swtc/main.do>
8. Quarati F, Dorenbos P, van Der Biezen J, Owens A, Selle M, Parthier L, Schotanus P (2013) Scintillation and detection characteristics of high-sensitivity CeBr_3 gamma-ray spectrometers. *Nucl Instrum Methods A* 729:596–604
9. Jun-Ho Lee, Jong-In Byun, Dong-Myung Lee (2019) In-situ CeBr_3 gamma-ray spectrometry for radioactivity analysis of soil. *J Radioanal Nucl Chem* (in press)
10. ICRU 53 (1994) Gamma-ray spectrometry in the environment. International Commission on Radiation Units and Measurements. Bethesda, Maryland. Report 53
11. Arnold D, Sima O (2004) Extension of the efficiency calibration of germanium detectors using the GESPECOR software. *Appl Radiat Isot* 61:117–121
12. Allison J, Amako K, Apostolakis J, Arce P, Asai M, Aso T, Bagli E, Bagulya A, Banerjee S, Barrand G, Beck BR (2016) Recent developments in Geant4. *Nucl Instrum Methods A* 835:186–225
13. Geant4 Collaboration (2016) Toolkit developers guide. Available online at: <http://cern.ch/Geant4>
14. Joel JSC, Maurice NM, Jilbert NME, Ousmanou M, David S (2018) Monte Carlo method for gamma spectrometry based on GEANT4 toolkit: efficiency calibration of BE6530 detector. *J Environ Radioact* 189:109–119

15. Al-Masri MS, Hasan M, Al-Hamwi A, Amin Y, Doubal AW (2013) Mass attenuation coefficients of soil and sediment samples using gamma energies from 46.5 to 1332 keV. *J Environ Radioact* 116:28–33
16. Currie LA (1968) Limits for qualitative detection and quantitative determination. Application to radiochemistry. *Anal Chem* 40:586–593

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.