Research Article

Activity Concentrations of Sr-90 and Cs-137 in Seawater and Sediment in the Gulf of Tonkin, Vietnam

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In this study, we measured the activity concentrations of Cs-137 and Sr-90 in surface seawater and surface sediments at the Tra Co, Bach Long Vi, and Ky Anh locations in the Gulf of Tonkin, Vietnam, from December 2018 to October 2019. The average activity at the selected locations was 1.22, 1.43, and 1.33 Bq/m³ for Cs-137 and 0.88, 1.17, and 1.09 Bq/m³ for Sr-90 in surface seawater samples and 0.74, 1.01, and 0.81 Bq/kg dry for Cs-137 and 0.49, 0.49, and 0.43 Bq/kg dry for Sr-90 in sediment samples. The ratio of the average activity concentration (Cs-137/Sr-90) in the surface seawater was 1.42, 1.22, and 1.22 at the Tra Co, Bach Long Vi, and Ky Anh locations, respectively. These are somewhat low compared to the global ratios (1.6 and 1.8). Meanwhile, Cs-137/Sr-90 ratios in the sediment samples at the selected locations were 1.51, 2.06, and 1.88, respectively, which is equal to or greater than the corresponding value for global sedimentation according to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Correlations between Sr-90, Cs-137, and organic carbon content were detected in this study. The results showed that Cs-137 has a high correlation with the organic carbon content in sediment, while Sr-90 has a low correlation.

1. Introduction

The Gulf of Tonkin is located at the coordinates of 17°00′–21°40′ north latitude and 105°40′–109°40′ east longitude, stretching to nearly 5 geographical latitudes with a distance from north to south of approximately 500 km from the coast of Vietnam. The Gulf of Tonkin is one of the major bays in Southeast Asia and the world, with an area of approximately 126,250 km², widest width of approximately 320 km, and narrowest width of approximately 220 km. It is a place where natural resources are stored, especially seafood and oil [1].

Artificial radioisotopes penetrated the marine environment either as a result of global nuclear tests (the 1950s–1960s), radioactive waste discharge, or the Chernobyl incident (1986), but the most common cause is radioactive dust due to atmospheric nuclear tests. The concentrations of Cs-137 and Sr-90 entering the marine environment from radioactive dust due to atmospheric nuclear tests are reported to be 603 PBq and 377 PBq, respectively [2].

Recently, due to the accident at the Fukushima nuclear power plant, as well as an increase in the commercial activity of nuclear power plants around the Gulf of Tonkin from China (4 NPPs operational and 2 NPPs under construction) [3], there is a risk of introducing radioactive pollutants Cs-137 and Sr-90 into the marine environment in general and the Gulf of Tonkin in particular. Thus, artificial radioisotopes (Cs-137 and Sr-90) contribute to the public dose due to human consumption of marine food. Thus, studies and
surveys must produce a database of radioisotopes in marine environmental objects in order to assess the impact of these radioactive nuclei on human health and the environment [2].

Cs-137 and Sr-90 have a relationship with regard to organic carbon content [4]. The activity of Cs-137 is highly correlated with organic carbon, while that of Sr-90 is lower. It was shown that Cs-137 is more highly absorbed in soils with high organic content.

Our work concerns the determination of radioactivity of Sr-90 and Cs-137 in seawater and marine sediment and the correlation between them in the Gulf of Tonkin of Vietnam. The correlations of organic carbon with Cs-137 and Sr-90 in the sediment samples are also calculated in this study. The collected data will be very useful in assessing the impact of current and future applications of atomic energy on Vietnam’s marine environment and public health.

2. Sampling and Research Method

2.1. Study Areas. Seawater samples and marine sediments were collected from 3 locations (1 offshore location and 2 nearshore locations) in the Gulf of Tonkin. The locations are listed in Table 1, and the sample collection locations are shown in Figure 1.

Depending on the flow of seawater in the Gulf of Tonkin, the samples were collected at times when the flow moved from north to south, and vice versa [1]. These samples were collected in December 2018, April 2019, June 2019, and October 2019.

2.2. Sampling and Sample Pretreatment

2.2.1. Sampling. A boat traveled to the sampling area at 5 km from the shore. For surface seawater samples, an electric pump with a flow of approximately 200 L/h was used at a depth of approximately 1 meter. Approximately 400 L of seawater was collected in containers and transported to the shore for pretreatment.

The surface sediment sample was collected (0–10 cm in depth) in the same position using a sampling bucket with a sample weight of approximately 5 kg. The collected samples were then placed into polyethylene bags and transferred to the laboratory.

2.2.2. Sample Pretreatment for Seawater Samples. After being brought to shore, the seawater samples were placed in separate plastic containers to pretreatment for Cs-137 (200 L) and Sr-90 (100 L). The pretreatment separated Sr and Cs from large volumes of seawater. The pretreatment of sediment was conducted in the laboratory.

For pretreatment of Cs-137 in seawater samples, a 200 L sample with 7 M HNO₃ was acidified to have a pH of 1–2. A micropipette took 500 µL of Cs-134 solution (1.00 ± 0.05 Bq/mL) and added it to the seawater sample in order to calculate the absorption efficiency. The solution was stirred for 5–10 minutes. Next, 10 mL of CsCl solution (100 mg/mL) was added and stirred for 5–10 minutes and then added slowly to a 60 mL sample of NiCl₂ solution (100 mg/mL) and stirring continued. The solution pH (~5) was adjusted with 5 M NaOH solution into the sample, and 3.5 L of K₂Fe(CN)₆ solution (100 mg/mL) was added and stirred for approximately 3 minutes to readjust the pH (~5). Settling was allowed to proceed for approximately 8 h. The supernatant clear water was pumped away. The remaining ferrocyanide (III) precipitate (Cs₂Ni[Fe(CN)₆]₃, approximately 10 L) was transferred to a polyethylene can and brought to the laboratory.

For pretreatment of Sr-90 in seawater, a 100 L sample was acidified with 7 M HNO₃ to pH = 2. A micropipette was used to take 200 µL of Sr-85 solution (the Sr-85 radioactivity was 5.00 ± 0.14 Bq/mL) and place it into the sample for calculation of the chemical recovery. The solution was stirred for 5–10 minutes. Then, 20 mL of SrCl₂ solution (100 mg/mL) was added to the sample and mixed well. The sample was adjusted to pH = 8 with 6 M NH₄OH solution. This was added slowly to the sample of 1000 g Na₂CO₃ (predissolved with distilled water) and stirred for approximately 3 minutes. The precipitate was allowed to settle completely for approximately 8 hours. Finally, the supernatant was pumped away, and the remaining 30 L of precipitate was placed in a polyethylene can and transferred to the laboratory for chemical separation.

For pretreatment of Sr-90 in sediment samples, the sample was brought to the laboratory, and approximately 150 g of air-dried samples was placed into porcelain cups and heated at 450°C for approximately 24 hours. After cooling to room temperature, the sample was transferred into a 2-liter beaker glass. Next, 200 µL of Sr-85 solution was micropipetted (the radioactivity of Sr-85 was 4.96 ± 0.14 Bq/mL), and 10 mL of SrCl₂ (10 mg/mL) was added to the sample. Then, 1 liter of HCl acid was added to the sample, which was covered with a glass lid. This was boiled and stirred thoroughly for approximately 3 hours. After that, the solution was filtered with a glass filter and rinsed 3 times with hot water. The sample was diluted to approximately 4 liters with distilled water. Next, 150 g Na₂CO₃ was added and boiled for 20 minutes. The pH was adjusted to 9 by adding 10 M NaOH solution to completely precipitate. After settling at room temperature, the sample was ready for chemical separation.

2.3. Chemical Separation

2.3.1. Chemical Separation of Sr. In the laboratory, the precipitate from the sample (~30 L for seawater and ~1 L for sediment) after pretreatment was filtered through 10 µm filter paper. The precipitate was rinsed with distilled water, filtered (2 times), and then dissolved completely in 12 M HCl solution. Next, 200 g H₂C₂O₄·2H₂O was added to the solution, which was heated at 100°C to dissolve completely. The solution was boiled with a 10 M NaOH solution. The solution pH was adjusted to 4.0–4.2 with 10 M NH₄OH solution. The solution was heated for 1 h to completely precipitate and cool at room temperature.
Table 1: Sampling locations in the Gulf of Tonkin.

| Sampling station       | Sampling date            | Latitude  | Longitude | Water depth (m) | Remarks                  |
|------------------------|--------------------------|-----------|-----------|-----------------|--------------------------|
| Tra Co town            | December 2018, April 2019, June 2019, October 2019 | 21°25'37" N | 108°02'19" E | 1               | ~5 km from the shore     |
| Bach Long Vi Island    | December 2018, April 2019, June 2019, October 2019 | 20°07'15" N | 107°42'54" E | 1               | ~5 km from the island, ~120 km from shore |
| Ky Anh town            | December 2018, April 2019, June 2019, October 2019 | 18°08'52" N | 106°23'13" E | 1               | ~5 km from the shore     |

Figure 1: Sampling locations in the Gulf of Tonkin.
The solution was filtered again into a separate solution and precipitated to different cups. The filtering solution was acidified with 12M HCl. Precipitation was conducted one more time as in the first step, and the 2 parts of the precipitate were combined for further treatment. The precipitate was rinsed with distilled water (3 times) and filtered. The precipitate was calcined for 5 hours at a temperature of 600–700°C.

The precipitate was dissolved with 10M HCl, and the sample was filtered through filter paper and rinsed with hot water. Next, 2 mL of FeCl₃ solution (5 mg/mL) was added to the sample, and the solution was boiled. Then, the solution was adjusted to pH = 8 by slowly adding NH₄OH without CO₂. The solution was boiled for 5 minutes to precipitate. The precipitate was filtered from the solution through filter paper when the solution was still hot. The solution part was retained, and the precipitate was removed.

Next, 5 mL of BaCl₂ (10 mg/mL) was added to the solution, along with 10 mL of 6M CH₃COONH₄. The pH was adjusted to 5.0–5.5 by adding 5M CH₃COOH and 6M CH₃COONH₄ buffer solutions and then heating at 100°C. Then, 5 mL of 10% K₂Cr₂O₇ was added to the solution. Heating was continued for complete precipitation and then cooled to room temperature.

The sample was filtered with fine filter paper (0.45 μm) and rinsed with a small amount of 0.01 M CH₃COONH₄. The solution part was retained, and the precipitate was removed. The solution was adjusted to pH = 9 by slowly adding 6M NH₄OH solution. The sample solution was heated, and (NH₄)₂CO₃ saturated solution was added slowly to produce SrCO₃ precipitation. The precipitate was filtered with filter paper and rinsed with 0.01 M NH₄OH solution. The solution part was removed from the precipitate.

The precipitate was dissolved again with 10M HNO₃. Then, 2 mL FeCl₃ (5 mg/mL) and 1 g of NH₄NO₃ were added, and the solution was boiled. The solution was adjusted to pH = 8 by slowly adding NH₂OH without CO₂ to precipitate Fe(OH)₃, and boiling was continued for 5 minutes. The time was noted to calculate the enrichment time of Sr-90/Y-90.

The solution was filtered through filter paper while the sample was still hot. The precipitate was rinsed with 0.01 M NH₂OH. The solution was retained, and the precipitate part was removed. The solution was dried until the volume was approximately 100 mL, cooled, and 5 mL of 10M HNO₃ was added to the solution. The sample was transferred to a measuring box (diameter 43 mm, height 25 mm; add distilled water to achieve the volume of geometry), and the sample was measured on a gamma spectrometer to determine the Sr-85 activity for calculating the Sr chemical recovery. After that, the solution was stored for 14 days for the Y-90 separation.

2.3.2. Chemical Separation of Y. After 14 days, the samples were used in the next procedure (described as follows) for Y-90 separation.

Then, 2 mL YCl₃ (10 mg/mL) was added to the sample in the solution. The solution was boiled, and NH₂OH was slowly added without CO₂ to obtain the precipitate Y(OH)₃ (at pH = 8). The solution was boiled for 5 minutes. Note this time for correction decay factor between initial precipitation and the time of counting beta spectra.

The sample solution was filtered through filter paper when the solution was hot. The precipitate was rinsed in the filter with hot water (2 times). The precipitate was retained and removed from the sample solution. Next, the precipitate was completely dissolved with 10M HCl, the filter was rinsed with hot distilled water, and the solution was heated. Then, 2 g of H₂C₂O₄·2H₂O was added, and the pH was adjusted with 10M NH₄OH 10M to precipitate Y₃(FeO₄)₅ at 1.5–2. The solution was heated and then cooled to room temperature.

The sample was filtered through fine filter paper of 0.45 μm. The precipitate was rinsed in the filter with distilled water (2 times) and then rinsed again with C₂H₅OH. The filter containing the precipitate was dried with an IR and weighed to obtain the mass of precipitate. Then, the precipitate was measured on a low-background beta counting system (MP-9300).

2.4. Analysis. A low-background alpha/beta total counting system (Model: MPC 9300) was used for counting beta spectra from Y-90 after separation. The efficiency of the beta detector was calculated by a 100 μL ⁹⁰Sr/⁹⁰Y solution standard (36.45 ± 0.32 Bq/ml), and the background was approximately 0.8 cpm. The absolute efficiency of the counting system is about 39.48%.

A High Purity Germanium (HPGe) gamma detector system was used to determine the gamma spectra from Sr-85, Cs-134, and Cs-137. The model of HPGe detector used in this study was the GX-3019, which has a 30% relative efficiency, resolution of 1.90 keV at 1332 keV of Co-60, peak-to-Compton ratio of 56, and sensitivity volume of 138 cm³. The integral background in the energy range of 100–2000 keV was approximately 2.1 cps. Computer software MAESTRO-32 was used to process the obtained spectra. The absolute efficiency of the HPGe detector was determined using IAEA reference material (IAEA-447 Moss soil, IAEA-375 Russian soil).

2.4.1. Analysis of Sr-90. After chemical separation, the Sr-90 concentration was determined by quantifying the Y-90 level using a low-background beta counter after a radioactive equilibrium between Sr-90 and Y-90 was established. Each sample was measured in 5400 s in the beta counting system. The activity of Sr-90 in the sample is calculated by the following formula:

$$A_{\text{Sr-90}} = \frac{S \times D_1 \times D_2}{E \times H_{\text{Sr}} \times H_{\text{Y}} \times M}$$

where $A_{\text{Sr-90}}$ is the activity of Sr-90 (Bq/m³ or Bq/kg), $S$ is the counting rate of the sample (counts/second), $E$ is the efficiency of the counting system (%), $H_{\text{Sr}} = (A_1/A_2) \times 100$ is the chemical recovery of Sr (%), $A_1$ is activity of Sr-85 in the sample before the chemical separation of Sr (Bq), $A_2$ is the activity of Sr-85 in the sample after chemical separation.
of Sr (Bq), \( H_Y = \left( W_2/W_1 \right) \times 100 \) is the chemical separation efficiency of \( Y \) (%), \( W_1 \) is the weight of \( Y^{3+} \) before the chemical separation of \( Y \) (g), \( M \) is the sample volume or sample weight (m\(^3\) or kg), \( D_1 = e^{[(ln2)/T_{1/2}] \times t_1/2]} \) is the correction decay factor from the chemical separation of \( Y \) to counting, \( D_2 = 1/1 - e^{[(ln2)/T_{1/2}] \times t_2]} \) is the correction decay factor during the measurement period, \( t_1 \) is the time from chemical separation of \( Y \) to counting (hours), \( t_2 \) is the measurement time (hours), and \( T_{1/2} \) is the half-life of Y-90 (64.2 hours).

The uncertainty of Sr-90 activity was calculated from the following equation:

\[
\sigma_{A_{Sr-90}} = A_{Sr-90} \sqrt{\left( \sigma\left( N_{Y-90} \right) / N_{Y-90} \right)^2 + \left( \frac{\sigma_M}{M} \right)^2 + \left( \frac{\sigma_H}{H_Y} \right)^2 + \left( \frac{\sigma_{HY}}{H_{Sr}} \right)^2},
\]

where \( \sigma_{A_{Sr-90}} \) is the uncertainty of Sr-90 activity, \( N_{Y-90} \) is the count of Y-90, \( \sigma_{N_{Y-90}} \) is the uncertainty of count of Y-90, \( \sigma_M \) is the uncertainty of sample volume, \( \sigma_E \) is the uncertainty of efficiency of the alpha/beta counting system, \( \sigma_{HY} \) is the uncertainty of chemical recovery of \( Y \), and \( \sigma_{H_{Sr}} \) is the uncertainty of chemical recovery of Sr.

The Yttrium recovery was calculated from the weight of \( Y^{3+} \). Y-90 activity was calculated by using measured efficiency and recovery value. The Sr-90 activity will be approximately equal to Y-90 activity as ~97.5% of secular equilibrium will be reached after 14 days. The detection limits of Sr-90 in seawater and sediment samples were 0.1 Bq/m\(^3\) and 0.08 Bq/kg at a 95% confidence level, respectively.

2.4.2. Analysis of Cs-137. For the seawater sample, the Cs\(_2\)Ni[Fe(CN)\(_6\)] precipitate from pretreatment on the shore was brought to the laboratory (pH = 5). The precipitate was filtered through filter paper to completely remove the water and then dried at 100°C. The dried precipitate was placed in a plastic box (diameter 43 mm) and tracked with the HPGe detector for 24 hours to determine the activity of Cs-137. The activity of Cs-134 was also tracked and compared with the activity of Cs-134 in the pretreatment process from above in order to calculate the chemical separation recovery. The results of the absorption efficiency are listed in Table 2.

The sediment samples were brought back to the laboratory and dried at 80°C. The sample was finely ground to a particle size of less than 0.15 mm and placed in a cylindrical plastic box (diameter 60 mm, height 40 mm). The sediment sample was tracked with the HPGe detector for 24 hours to determine the activity of Cs-137.

The activity of Cs-137 in the sample is calculated by the following formula:

\[
A_{Cs-137} = \frac{N_p \times \epsilon \times t_1 \times \gamma \times M}{H_{Cs}},
\]

where \( A_{Cs-137} \) is the activity of Cs-137 in sample (Bq/m\(^3\) or Bq/kg), \( N_p \) is the number of counts from peak 661.6 keV of Cs-137, \( \epsilon \) is the efficiency at energy 661.6 keV of HPGe detector, \( \gamma \) is the gamma yield, \( M \) is the volume or mass of sample (m\(^3\) or kg), \( H_{Cs} \) is the chemical recovery of Cs (%).

The detection limits of Cs-137 in seawater and sediment samples were 0.005 Bq/m\(^3\) and 0.01 Bq/kg at a 95% confidence level, respectively.

The chemical recoveries of Sr and Cs were calculated from the activity of Sr-85 and Cs-134 before and after chemical separation. The chemical recovery of Y was calculated from the weight of Y\((\text{g})\) from YCl\(_3\) (before chemical separation) and Y\((\text{g})\) from Y\(_2\)(C\(_2\)O\(_4\))\(_3\) (after chemical separation). The results are listed in Table 3.

2.5. Quality Assurance and Quality Control. The precision and accuracy of the seawater samples were performed by analyzing international comparison samples held by IAEA-RML 2017–01 for Cs-134, Cs-137, and Sr-90 [5]. The results of the analysis of reference samples correspond to the validation values. For sediment samples, IAEA reference materials were used to QC the procedure of Sr-90 and Cs-137. The QC results in the Table 4 show that the bias is lower than 10% for all samples except for Sr-90 activity in the seawater sample.

3. Results and Discussion

The activity of the radioactive isotopes Sr-90 and Cs-137 in seawater and sediments from the Tra Co, Bach Long Vi, and Ky Anh areas of the Gulf of Tonkin in 2018–2019 is presented in Table 2. Along with the determination of the radioactive isotopes of Sr-90 and Cs-137 in seawater and sediments, the organic carbon content in marine sediment samples was determined (Table 5). The relationship between the radioactive activity of Sr-90 and Cs-137 with the organic carbon content in sediment is shown in Figure 2.

The radioactive activity of Sr-90 in seawater during 2018 and 2019 (min to max value) for Tra Co, Bach Long Vi, and Ky Anh ranged from 0.78 to 1.08 Bq/m\(^3\), 1.11 to 1.24 Bq/m\(^3\), and 0.81 to 1.23 Bq/m\(^3\), respectively. The activity of Cs-137 in seawaters of Tra Co, Bach Long Vi, and Ky Anh ranged from 1.05 to 1.33 Bq/m\(^3\), 1.23 to 1.62 Bq/m\(^3\), and 1.02 to 1.66 Bq/m\(^3\), respectively. The average Sr-90 radioactivity is 0.88, 1.17, and 1.09 Bq/m\(^3\), and the average Cs-137 radioactivity is 1.22, 1.43, and 1.33 Bq/m\(^3\), respectively. The variation of activity

\[
\text{(4)}
\]
of Sr-90 and Cs-137 in seawater for Tra Co, Bach Long Vi, and Ky Anh was also observed without significant fluctuations in general.

For the sediment samples, the radioactivity of Sr-90 during 2018 and 2019 for Tra Co, Bach Long Vi, and Ky Anh ranged from 0.37 to 0.59 Bq/kg dry, 0.41 to 0.54 Bq/kg dry, and 0.34 to 0.56 Bq/kg dry. The radioactivity of Cs-137 in sediments ranged from 0.44 to 1.05 Bq/kg dry, 0.82 to 1.17 Bq/kg dry, and 0.67 to 1.00 Bq/kg dry, respectively. The average activity of Sr-90 is 0.49, 0.49, and 0.43 Bq/kg dry, and the average activity of Cs-137 is 0.74, 1.01, and 0.81 Bq/kg dry, respectively.

The ratio of Cs-137/Sr-90 helps to determine the origins of artificial radioisotopes. Here, the average ratios of Cs-137/Sr-90 in surface seawater at Tra Co, Bach Long Vi, and Ky Anh are 1.42, 1.22, and 1.22, respectively, which is somewhat low compared to the global fallout ratio of 1.6–1.8 [2, 6, 7].

### Table 2: Radioactivity concentrations of Sr-90 and Cs-137 in surface seawater and sediment in the Gulf of Tonkin.

| Sampling location | Sampling date | Surface seawater (Bq/m³) | Sediment (Bq/kg, dry) |
|-------------------|---------------|--------------------------|-----------------------|
|                   |               | Cs-137                  | Sr-90                 | Cs-137/Sr-90 | Cs-137                  | Sr-90 | Cs-137/Sr-90 |
| Tra Co            | 1 Dec. 2018   | 1.05 ± 0.16*             | 1.08 ± 0.20           | 0.97         | 0.97 ± 0.17             | 0.44 ± 0.08 | 2.20 |
|                   | 1 Feb. 2019   | 1.21 ± 0.21              | 0.80 ± 0.15           | 1.51         | 0.44 ± 0.08             | 0.54 ± 0.13 | 0.81 |
|                   | 1 June 2019   | 1.33 ± 0.18              | 0.78 ± 0.16           | 1.71         | 0.51 ± 0.10             | 0.37 ± 0.09 | 1.38 |
|                   | 1 Oct. 2019   | 1.27 ± 0.17              | 0.85 ± 0.14           | 1.49         | 1.05 ± 0.18             | 0.59 ± 0.14 | 1.78 |
|                   | **Average ± SD** | **1.22 ± 0.12** | **0.88 ± 0.14** | **1.42 ± 0.31** | **0.74 ± 0.31** | **0.49 ± 0.10** | **1.54 ± 0.59** |
| Bach Long Vi      | 1 Dec. 2018   | 1.27 ± 0.22              | 1.18 ± 0.24           | 1.08         | 1.17 ± 0.19             | 0.48 ± 0.08 | 2.44 |
|                   | 1 Feb. 2019   | 1.62 ± 0.25              | 1.11 ± 0.20           | 1.46         | 1.08 ± 0.17             | 0.41 ± 0.07 | 2.63 |
|                   | 1 June 2019   | 1.23 ± 0.13              | 1.24 ± 0.25           | 0.99         | 0.82 ± 0.13             | 0.54 ± 0.14 | 1.52 |
|                   | 1 Oct. 2019   | 1.58 ± 0.23              | 1.14 ± 0.20           | 1.39         | 0.98 ± 0.14             | 0.52 ± 0.12 | 1.88 |
|                   | **Average ± SD** | **1.43 ± 0.20** | **1.17 ± 0.06** | **1.22 ± 0.23** | **1.01 ± 0.15** | **0.49 ± 0.06** | **2.12 ± 0.51** |
| Ky Anh            | 1 Dec. 2018   | 1.02 ± 0.11              | 0.81 ± 0.15           | 1.26         | 0.88 ± 0.15             | 0.56 ± 0.13 | 1.57 |
|                   | 1 Feb. 2019   | 1.66 ± 0.25              | 1.19 ± 0.24           | 1.39         | 0.69 ± 0.12             | 0.34 ± 0.08 | 2.03 |
|                   | 1 June 2019   | 1.29 ± 0.24              | 1.13 ± 0.21           | 1.15         | 1.00 ± 0.19             | 0.39 ± 0.08 | 2.56 |
|                   | 1 Oct. 2019   | 1.33 ± 0.17              | 1.23 ± 0.25           | 1.09         | 0.67 ± 0.08             | 0.42 ± 0.09 | 1.60 |
|                   | **Average ± SD** | **1.33 ± 0.26** | **1.09 ± 0.19** | **1.22 ± 0.14** | **0.81 ± 0.16** | **0.43 ± 0.09** | **1.94 ± 0.47** |

*Measurement uncertainty.

### Table 3: Organic carbon content (%) at study areas.

| Sample and collection time | Tra Co | Bach Long Vi | Ky Anh |
|----------------------------|--------|--------------|--------|
| 1 December 2018            | 3.32   | 7.3          | 1.51   |
| 1 February 2019            | 0.81   | 5.3          | 1.03   |
| 1 June 2019                | 1.5    | 2.01         | 1.5    |
| 1 October 2019             | 3.41   | 3.51         | 1.1    |
sediment, the ratios of Cs-137/Sr-90 at Tra Co, Bach Long Vi, and Ky Anh are 1.54, 2.12, and 1.94, respectively, which is equivalent to or slightly higher (by approximately 1.3 times) than the corresponding value (1.6 and 1.8) in the global fallout [7].

The radioactivity of Sr-90 and Cs-137 in surface seawater and sediments was previously determined at several locations in the Asia-Pacific regional seas. The average activity of Cs-137 in Pacific Ocean is about 2.5 Bq/m³ and 1.4 Bq/kg dry (corrected day to March 2001) in seawater and sediment, respectively [8]. The average activity of Sr-90 in the East China Sea and North Pacific (measured from 3–31 July 2000) is 0.9025 Bq/m³ [9]. In the study areas, the average activity values of Sr-90 in surface seawater and sediments are 1.05 Bq/m³ and 0.47 Bq/kg dry, respectively. For Cs-137, the activity is 1.33 Bq/m³ and 0.85 Bq/kg dry in seawater and sediment, respectively. This result shows that the activity of Sr-90 and Cs-137 in the surveyed areas of the Gulf of Tonkin is lower than or equal to the Asia-Pacific regional seas.

The determination of organic carbon content in sediments was evaluated with regard to the relationship between the organic carbon and radioactivity of Sr-90 and Cs-137 in sediments. From that, Table 5 shows the organic carbon content by the Walkley-Black method at Tra Co, Bach Long Vi, and Ky Anh. Figure 2 shows the correlation between the organic carbon content and radioactivity in the sediment sample at three locations. The results showed a high positive correlation coefficient for the organic carbon content with Cs-137. This also shows that Cs-137 is strongly associated with certain special organic components in sediments, leading to Cs-137 being trapped in sediments and limiting its movement [7, 10, 11]. By contrast, Sr-90 is almost independent of the composition of organic carbon, where the correlation between Sr-90 activity and organic carbon content is weak. The activity of Sr-90 also changes less seasonally, while the activity of organic carbon increases.

4. Conclusion

Activity concentrations of Sr-90 and Cs-137 in seawater and sediment were determined in the Gulf of Tonkin of Vietnam (Tra Co, Bach Long Vi, and Ky Anh) from December 2018 to October 2019. The obtained results determined the radioactivity of Sr-90 and Cs-137 in surface seawater and sediment at several locations in the Gulf of Tonkin at different times. A comparison was conducted using the results of some published authors regarding the Asia-Pacific region and the South China Sea. The ratios of Cs-137/Sr-90 in surface seawater at Tra Co, Bach Long Vi, and Ky Anh were lower than the global fallout ratio; while in sediment, the ratios of Cs-137/Sr-90 were
equivalent to or slightly higher (by approximately 1.3 times) than the corresponding value in the global fallout. The results for radioactivity and organic carbon content also showed that Cs-137 is strongly correlated with the organic carbon content in sediment. By contrast, Sr-90 is almost independent of the organic carbon content.

Data Availability
The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest
The authors declare that there are no conflicts of interest regarding the publication of this paper.

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