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Activity of radioactive isotopes in the offshore seawater at vicinity areas of Phu Quy, Phu Quoc and Bach Long Vy islands

Nguyen Thi Thanh Nga[1,*], Le Nhu Sieu[1], Nguyen Dinh Tung[1], Nguyen Van Phu[1], Tran Dinh Khoa[1], VuongThi Thu Hang[1], Nguyen Trong Ngo[1]

[1] *Nuclear Research Institute, 01 Nguyen Tu Luc, Dalat, Lam Dong*

[*] *Corresponding author: thanhngadhsply@gmai.com*

Abstract: In this work, activity of radioactive isotopes in some offshore seawater samples at vicinity areas of three islands has been investigated Phu Quy (Binh Thuan province), Phu Quoc (Kien Giang province), and Bach Long Vy (Hai Phong province). The ranges of radioactivity ^{226}Ra, ^{238}U, ^{137}Cs, ^{90}Sr, and ^{239,240}Pu in seawater at three offshore islands are 2.09 ÷ 4.77 mBq/L, 2.33 ÷ 4.95 mBq/L, 0.98 ÷ 1.45 mBq/L, 1.22 ÷ 1.49 mBq/L, and 0.0045 ÷ 0.0066 mBq/L, respectively. These values show that the radioactivities of the above-mentioned radionuclides differ between monitoring periods and between three offshore islands of Vietnam, but which is equal to or lower than the corresponding value according to others seas in Vietnam and Asia – Pacific. The correlation between radioisotopes and properties of seawater (pH, conductance, and salinity) is also shown in this study to assess their impact on the fluctuations of the above radinuclides.

Keywords: *Radioactivity in seawater, monitoring, offshore island.*

I. INTRODUCTION

Natural radionuclides are sourced from the Earth's crust, the most common being ^{40}K, the radioactive chains of ^{238}U, ^{235}U, ^{232}Th, and some of the less common radionuclides such as ^{50}V, ^{87}Rb, ^{113}Cd, ^{138}La, etc [1].Moreover, to ensure national energy security and socio-economic development, countries around the world continue to build and operate nuclear power plants (NPP) due to traditional sources of energy have been saturated or exhausted. As a result, unfortunate accidents occurred such as leaked sealed radioactive sources from nuclear weapons tests (according to statistics from 1945 to 1980, there were more than 500 nuclear weapons tests in the atmosphere and hydrosphere) or accidents from nuclear power plants such as Chernobyl NPP (1986), Fukushima I - Japan NPP (2011) [2,3]. The result is a series of radioactive isotopes released into the atmosphere such as ^{3}H, ^{90}Sr, ^{14}C, ^{239,240}Pu, ^{137}Cs, ^{131}I, ... For this reason, people are worried about radioactive material released due to nuclear incidents.

In recent years, China has built a lot of NPP and mainly concentrated in the southeast coastal area of China, such as Fangchenggang NPP is about 50 km from Mong Cai city, Quang Ninh, Changjiang NPP on Hai Nam island is 185 km away from Van Don island district, Quang Ninh ...[4]. The operation of NPPs near the border of our country will be potentially hiding many risks of releasing radioactive substances into the marine environment, affecting Vietnam and other countries in the region, especially when incidents occur.

Phu Quy islocated at the coordinates of 10°29′–10°31′ north latitude and 108°55′–

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108°59′ east longitude. Phu Quy is an island district in the offshore island system in Binh Thuan province, consisting of 10 small islands with a natural area of approximately 30 km^2. It is adjacent to inland and international seaways and has an important transshipment position between Vung Tau, Phan Thiet, and Truong Sa archipelagos [5].

Phu Quoc is stretched from 9°53′ to 10°28′ north latitude and 103°49′–104°05′east longitude (Kien Giang Province), length nearly 50km. It is the largest island (an area of 593 km^2), which is in the South Pole of Vietnam with a triangle shape. Phu Quoc is located at a low latitude and deep into Thailand Bay, so the weather is cool with a tropical monsoon climate, accompanied by a dense river system with a potential groundwater reserve, the water quality is quite good and can be used for supplying water domestic water use and developing tourism and sightseeing activities at the beach [6].

Bach Long Vi island is located at 20°07′–20°08′ north latitudeand 107°42″–107°44′east longitude in the center of Gulf of Tonkin. The island is an offshore district of Haiphong city, with a circumference 6.5 km, width of approximately 1.5 km and length of approximately 3 km.

This study was provided some data on the current state of natural and artificial radioactive pollution in three adjacent offshore areas and compared the radioactivity between the nearshore and offshore areas. Besides, the assessment of the correlation between the concentration of radioactive substances in the offshore sea was also conducted. The collected data will be useful in assesing the impact of current and future applications of atomic energy on Vietnam's marine environment.

II. SAMPLING AND RESEARCH METHODS

A. Sampling

In this report, seawater samples were collected at a depth of 1 m at locations 2 - 120 km from the shore of Phu Quy, Phu Quoc, and Bach Long Vi islands (Fig. 1). The collection position is shown in Table 1. The seawater samples were collected by using an electrical pump with a flow of approximately 200-500 L/h. The sample volume, which was used to determine the radioactive isotopes, is about 5 L for ^{40}K; 200 L to analysis isotopes: ^{137}Cs, ^{226}Ra, ^{238}U, and ^{90}Sr and 400 L for $^{239,240}Pu$. The samples after collection were preliminarily processed by the enrichment method on the island and then transferred to the laboratory for further analysis[7–10].

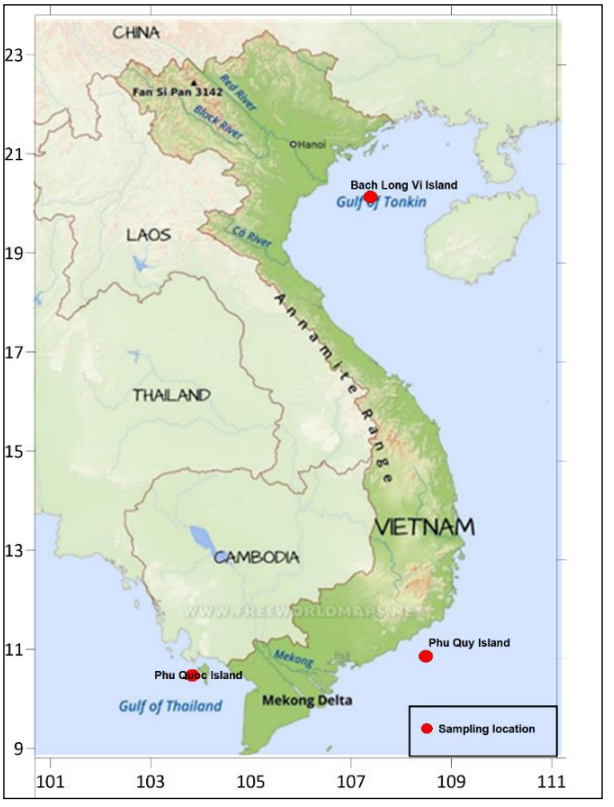

Fig. 1. Sampling locations in Bach Long Vi, Phu Quy, and Phu Quoc islands in Viet Nam

B. Method of analysis

In this study, the samples were measured on a low background gamma spectrometer equipped with a HPGe detector model GX-3019 having a relative efficiency of 30%, Peak/Compton ratio of 56:1, an energy resolution of 1.90 keV at 1332 keV of ^{60}Co, and sensitivity volume of 138 cm^3. The detector was placed in a chamber made oflead, the integral background from 100 - 2000 keV about 2.1 pulses/second. The spectrometer was calibrated by using the soil reference materials IAEA-CU-2006-03 and IAEA-CU-2009-03, which including radioactive isotopes: ^{54}Mn, ^{60}Co, ^{65}Zn, ^{109}Cd, ^{134}Cs, ^{137}Cs, ^{210}Pb, ^{241}Am, ^{40}K, ^{226}Ra, ^{234}U, ^{238}U, ^{228}Ac, ^{208}Tl, ^{214}Bi, with a measurement time of about 90,000 s to obtain good counting statistic. The gamma spectral are stored on PC and processed by common standard software such as MAESTRO®-32, SPEDAC, GAMMAW - specialized IAEA software for gamma spectrum analysis.

The ^{137}Cs isotope was determined directly through the peak 661.7 keV (85.1%); ^{238}U was quantified based on photo peak of their progenies ^{234}Th at 63.3 keV (3.8%) [7].

The ^{90}Sr radionuclide was analyzed by radiochemical separation technique to enrich, remove influencing elements. After chemical separation of Sr, the sample was waited in 14 days to meet the ^{90}Sr-^{90}Y activity equilibrium. We then conducted chemical separation and measurement ^{90}Y on a low background beta activity counting system with measuring time from 0.5 - 1 h. The low alpha/beta counting system MPC 9300 uses an airflow rate meter with anti-coincidence and shielded lead layer to reduce the cosmic background to 0.02 counts/second for beta background and 0.001 counts/second for the alpha background. It has 64% efficiency for beta and 31% efficiency for alpha with a mixed source in 25.4 mm diameter [10].

For the ^{239,240}Pu isotopes, the sample also was separated from radioactive by electrolysis method to concentrate the sample on a stainless-steel plate and then conduct the measurement on alpha Ensemble-2 spectrometer with counting time from 24 to 48 hours [8].

C. Quality Assurance and Quality Control

The precision and accuracy of the analysis were performed by analyzing international comparison samples held by IAEA-RML-2018-01 for Cs-134, Cs-137 [11]. The results of the analysis of reference samples correspond to the certified values. The QC results in Table II show that the bias is lower than 10% for all samples except for Sr-90 activity in the seawater sample.

Table I. Sampling locations

Sampling station	Sampling date	Latitude	Longitude	Water depth (m)	Number of samlpes	Remarks
Phu Quy island	05/2019 08/2019	10°30′ N	108°57′ E	1	2 1	2 km from the shore
Phu Quoc island	05/2019 08/2019	10°02′ N	103°59′ E	1	1 2	2 km from the shore
Bach Long Vy island	06/2020	20°07′ N	107°43′ E	1	2	~ 5 km from the island, ~ 120 km from shore

Table II. Results of reference materials for QC of seawater sample.

Sample	Isotope	Lab value ± uncertainty (Bq/kg)	Certified/reference value ± uncertainty (Bq/kg)	Bias (%)
IAEA-RML-2018-01 (Seawater)	Cs-134	0.295 ± 0.022	0.3125 ± 0.0031	-5.6
	Cs-137	0.251 ± 0.018	0.2597 ± 0.0026	-3.4
IAEA-368 (Sediment)	Po-210	21.56 ± 1.14	23.02 ± 3.70	-6.3
IAEA-156 (Clover)	Sr-90	14.27 ± 1.15	14.8 ± 1.45	

III. RESULTS AND DISCUSSION

The results of activity values of natural and artificial radioactive isotopes in seawater collected offshore of at Phu Quy – Binh Thuan, Phu Quoc – Kien Giang, and Bach Long Vi – Hai Phong islands were shown in Table III. The physical and chemical parameters of seawater are presented in Table IV and measured on HI98194 instrument. Measure is as follows: put the template into the clean-up, then insert the probe of the measuring instrument into the sample. After a few minutes, the results displayed on the screen. Depending on that purpose (conductivity, pH,…), we set the measurement parameter at the beginning.

Table III. Radioactive activity in offshore seawater at three islands location.

Location	**Sampling Time**	**Radioactive activity**				
		^{226}Ra (mBq/L)	**^{238}U (mBq/L)**	**^{137}Cs (mBq/L)**	**^{90}Sr (mBq/L)**	**^{239,240}Pu (10^{-3} mBq/L)**
Phu Quy island-Binh Thuan (Offshore)	05/2019	2.09 ± 0.15	2.51 ± 0.33	1.45 ± 0.29	1.33 ± 0.26	6.6 ± 0.8
	08/2019	4.32 ± 0.39	4.95 ± 0.43	1.12 ± 0.25	1.49 ± 0.32	4.5 ± 1.0
Phu Quoc island-Kien Giang (Offshore)	05/2019	2.88 ± 0.28	2.33 ± 0.40	1.06 ± 0.16	1.22 ± 0.25	5.1 ± 1.0
	08/2019	4.77 ± 0.71	4.83 ± 0.84	0.98 ± 0.17	1.41 ± 0.30	4.5 ± 0.7
Bach Long Vi island-Hai Phong (Offshore)	06/2020	10.07 ±1.52	8.04 ± 1.08	0.95 ± 0.18	1.20 ± 0.24	3.3 ± 0.3
Vinh Hai-Ninh Thuan (Near shore) [12]	(2015-2019)	0.91 ÷ 4.14	1.61 ÷ 7.69	1.14 ÷ 1.44	1.08 ÷ 2.43	1.0 ÷ 6.4
Asia – Pacificregion [15]		0.50 ÷ 4.14	1.00 ÷ 5.29	0.13 ÷ 2.95	0.67 ÷ 2.01	1.5 ÷ 9.2

Table IV. Physical and chemical parameters of seawater at Phu Quy – Binh Thuan, Phu Quoc – Kien Giang and Bach Long Vy – Hai Phong islands

Parameters	Phu Quy island		Phu Quoc island		Bach Long Vi island
	05/2019	08/2019	05/2019	08/2019	06/2020
pH	7.85	7.79	7.92	7.78	8.18
Conductance (mS/cm)	46.90	46.80	45.90	44.90	49.0
Salinity (‰)	33.80	32.80	32.30	31.20	35.1

The results have shown that the radioactivity of radioactive isotopes ^{226}Ra, ^{238}U, ^{137}Cs, ^{90}Sr, and ^{239,240}Pu in seawater at offshore of three islands are 2.09 ÷ 10.07mBq/L; 2.33 ÷ 8.04 mBq/L; 0.95 ÷ 1.45 mBq/L; 1.20 ÷ 1.49 mBq/L and 0.0033 ÷ 0.0066 mBq/L, respectively.

From the results presented in Table 3, the values of the radioactivity of natural radioisotopes determined at three islands are higher in comparison with the respective data from other seas in Vietnam but it is still in the range of radioactive isotopes in Asia - Pacific.

The ^{226}Ra isotope has a half-life of 1,600 years. In the marine environment, ^{226}Ra is easily absorbed into suspended particles and enters marine sediments. The average activity of ^{226}Ra in seawater at the monitoring points of Binh Thuan and Kien Giang provinces in August is about 1.5 to 2.0 times higher than in May, but the average value is in the range of the recorded values of the other areas of Vietnam and Asia - Pacific region [13-15]. The highest concentration of ^{226}Ra comes from Bach Long Vi island due to the high total supended solids (TSS) – about 15 mg/l. This island has the longest distance from shore (~140 km from shore) comparing with two other islands.

The ^{238}U is a naturally occurring radioisotope with a half-life of 4.47 billion years. Compared with other isotopes,^{238}U is more soluble in seawater. In 2019, we found that the value of ^{238}U individual activities of the two seas at Binh Thuan and Kien Giang in August was about 2 times higher than in May, and their activity range was 2.51 ÷ 4.95 mBq/L and 2.33 ÷ 4.83 mBq/L, respectively, but it is still within the range of ^{238}U studies of other seas in Vietnam [13,14] and Asia - Pacific [15]. This is likely because the total supended solids in seawater is much larger than the others samples (about 12.4/3.8). The ^{238}U isotopes have the same trend of ^{226}Ra isotope, the concentration of ^{238}U in Bach Long Vi island has the highest value in three locations.

The ^{137}Cs are artificial radioactive isotopes with a half-life of 30 years. Activity value range of ^{137}Cs in seawater samples at 03 monitoring points from 0.95 to 1.45 mBq/L. These values have no significant changes and are within the range of values recorded in the nearshore and offshore seas of Vietnam and the Asia - Pacific region [12-15].

According to the monitoring results in 2019, the activity of ^{90}Sr (half-life of 29 years) in seawater at 03 monitoring points (2 times/year) shows that there is no really clear change between the time collected sample and previous years (activity range: 1.33 ÷ 1.49 mBq/L at Phu Quy island and 1.22 ÷ 1.41 mBq/L at Phu Quoc island, 1.20 mBq/L at Bach Long Vi island). Radioactivity of ^{90}Sr is lower than Ninh Thuan nearshore seas but still

is tantamount to the range of radioactivity in offshore waters.

The ^{239}Pu (half-life of 24,100 years) and ^{240}Pu (half-life of 6,500 years) are two isotopes that have close alpha energy, therefore the total activity of ^{239}Pu and ^{240}Pu is ^{239,240}Pu. Activity range^{239,240}Pu in seawater at the offshore monitoring points of Phu Quy, Binh Thuan (activity range 4.5 ÷ 6.6×10^{-3}mBq/L), Phu Quoc, Kien Giang (activity range 4.5 ÷ 5.1 ×10^{-3} mBq/L) and Bach Long Vi, Hai Phong (3.3×10^{-3} mBq/L) does not fluctuate significantly between monitoring periods and has the same activity level as that near the shore of the other seas of Vietnam(activity range 6.0 ÷ 6.4 ×10^{-3}mBq/L). The activity level of ^{239,240}Pu in seawater through observations during the year is within the activity range of Asia - Pacific region [13-15].

The data of artificial radioisotope concentrations obtained are tendentiously lower than or equal to the same data provided by other authors in the Asia-Pacific region.

Table V. Correlation matrix (Pearson) between radioisotope and physical and chemical parameters of seawater in the study areas

Variables	^{226}Ra	^{238}U	^{137}Cs	^{90}Sr	^{239,240}Pu	pH	Conductance (mS/cm)	Salinity (‰)
^{226}Ra	**1.00**	**0.97**	-0.67	-0.36	**-0.88**	0.79	0.69	0.56
^{238}U	**0.97**	**1.00**	-0.63	-0.11	**-0.88**	0.63	0.63	0.48
^{137}Cs	-0.67	-0.63	**1.00**	0.18	**0.91**	-0.37	-0.02	0.17
^{90}Sr	-0.36	-0.11	0.18	**1.00**	0.15	-0.81	-0.45	-0.51
^{239,240}Pu	**-0.88**	**-0.88**	**0.91**	0.15	**1.00**	-0.55	-0.39	-0.19
pH	0.79	0.63	-0.37	-0.81	-0.55	**1.00**	0.83	0.79
Conductance (mS/cm)	0.69	0.63	-0.02	-0.45	-0.39	0.83	**1.00**	**0.97**
Salinity (‰)	0.56	0.48	0.17	-0.51	-0.19	0.79	**0.97**	**1.00**

Values in bold are different from 0 with a significance level alpha = 0.05

The correlation matrix (Pearson) between radioisotope and physical and chemical properties parameters of seawater inthe study areas was shown in Table 5. The ^{226}Ra isotope correlated with ^{238}U (^{226}Ra is the daughter of ^{238}U) and ^{239,240}Pu. The ^{238}U correlated with ^{226}Ra, ^{239,240}Pu (negative). The ^{137}Cs correlated with ^{239,240}Pu. The ^{90}Sr has no correlation with other isotopes but a weak correlation with pH. The ^{239,240}Pu correlated with ^{226}Ra, ^{238}U (negative) and ^{137}Cs (positive).

IV. CONCLUSION

To sum up, the activity of natural radioisotopes of ^{226}Ra, ^{238}U, and artificial radioisotopes ^{137}Cs, ^{90}Sr, ^{239,240}Pu observed were not only not significant variation between periods and monitoring locations but also within the value of the Asia Pacific region. The maximum value of these isotopes is much lower than the value of Asia-Pacific and other seas of Vietnam recorded (Ninh Thuan Sea from 2015-2019) [16]. The correlation between radioisotopes and

properties of seawater has alsobeen shown in this study.

Through the monitoring program, the baseline data of the background of radioisotopes in the marine environment has been collected and contributed to the national and regional base data sets. However, to establish the rules of the evolution of radioactive isotopes in the marine environment components and to set up the radioactive database for environmental impact assessment, we need to continue research and expand the scope of monitoring some more offshore and coastal areas [17].

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