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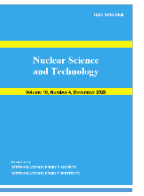

Radioactivity of some natural and artificial radionuclides in coastal seawater at Ninh Thuan, Ba Ria - Vung Tau and Ca Mau provinces in 2018

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Abstract: Radioactivities of some natural (Ra-226, Th-232, U-238, Po-210) and artificial (Cs-137, Sr-90 and Pu-239,240) radionuclides in coastal seawater at monitoring points of Ninh Thuan, Ba Ria - Vung Tau and Ca Mau provinces were analyzed quarterly in the year of 2018. Radioactivities of Ra-226, Th-232, U-238, and Cs-137 had been determined by simultaneous precipitation method and measured on low-level background gamma spectrometer; the radioactivities of Po-210 and Pu-239,240 had been analyzed by radiochemical separation method and measured on alpha spectrometer; the radioactivity of Sr-90 had been analyzed by radiochemical separation method and measured on low-level background beta counting system. The ranges of radioactivities of Ra-226, Th-232, U-238, Po-210, Cs-137, Sr-90, and Pu-239,240 were 2.71 ÷ 15.91, 3.45 ÷ 35.93, 3.02 ÷ 21.47, 1.51÷ 6.74, 0.88 ÷ 1.68, 1.08 ÷ 1.86 and 0.0026 ÷ 0.0062 mBq/L, with the average values of 6.37, 12.67, 8.34, 3.29, 1.36, 1.46 and 0.0043 mBq/L, respectively. The results shown that the radioactivities of the above-mentioned radionuclides varied between the seawaters at Ninh Thuan, Ba Ria - Vung Tau and Ca Mau seawater. Additionally, physicochemical parameters in seawater were also surveyed to assess their impact on the fluctuations of the above radionuclides.

Keywords: *Natural radionuclide, artificial radionuclide, radioactivity in coastal seawate.*

I. INTRODUCTION

Natural and artificial radionuclides are always present in the soil, water, and the air. Natural radionuclides are sourced from the Earth's crust, the most common being K-40, the radioactive chains of U-238, U-235, Th-232, and some of the less common radionuclides such as V-50, Rb-87, Cd-113, In-115, Te-123, La-138, Ce-142, etc. In addition, cosmogenic radionuclides are formed continually in the atmosphere due to cosmic radiation from outer space as H-3, Be-7 and C-14 are some important cosmogenic ones. Artificial radionuclides have been formed from the products of nuclear weapon tests in the atmosphere and water, routine releases of under-controlled low-level radioactive wastes of nuclear facilities (nuclear power plants, fuel reprocessing facilities, etc.) [1, 2, 3], radioactive waste dumping under the sea in the year 60s of last century, and nuclear incidents/accidents of nuclear power plants, nuclear submarines, incidents occurring

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during the receiving, using, and transporting of radioactive materials [1, 4, 5, 6,].

The principal objectives of the environmental marine radioactivity monitoring programme are: (1) survey and obtain the environmental quality changes for major radionuclides in coastal seawater at Ninh Thuan, Ba Ria - Vung Tau and Ca Mau in the year of 2018; (2) detect and issue monitoring about abnormal radioactive fluctuations of industrial activities as well as the use of nuclear energy of countries in the region that may affect Vietnam's marine environment.

II. MATERIAL AND METHOD

Coastal seawater samples were collected in February, May, August and November in the year of 2018 at three monitoring points (Ninh Thuan, Ba Ria - Vung Tau and Ca Mau provinces) and shown on Fig. 1. The samples were collectted at about 2-5 km from the coast with a volume of 200 L to analyze radionuclides of Ra-226, Th-232, U-238, Cs-137 [7]; 100 L for Po-210 analysis [8]; 100 L for Sr-90 analysis [9]; and 400 L for Pu-239,240 analysis [10]. The obtained samples were pretreated in the field and transferred to the laboratory to determine the radioactivity of natural and artificial radionuclides according to the procedures established by Center for Environment Research and Monitoring, Nuclear Research Institute that have been evaluated and accredited to be conformable to the ISO/IEC 17025:2017 with the national code of VILAS 525 [7-10].

Fig. 1. Three sampling monitoring points for coastal seawater at Ninh Thuan, Ba Ria-Vung Tau, and Ca Mau provinces of Vietnam

The radionuclides of Ra-226, Th-232, U-238, Cs-137 were enriched by simultaneous precipitation method, with recovery efficiency from 90 to 95%. They were then determined on the low-level background gamma spectrometer with the HpGe detector model GX3019, which had a relative efficiency of 30%, and an energy resolution of 1.90 keV at 1333 keV and a peak-to-Compton ratio of 56:1. Computer software MAESTRO-32 was used to process obtained spectrums [7].

The Po-210 and Pu-239,240 radionuclides were enriched by selective precipitation, using the radiochemical separation method and then deposited on silver or stainlesssteel discs by electrolysis technique.

The recovery efficiency for Po-210 was from 85 to 90% determined by the Po-209 yield tracer, and for Pu-239,240 from 75 to 80% determined by the Pu-242 yield tracer. The enriched samples were measured on alpha spectrometer having a energy resolution of ≤ 25 keV, a counting efficiency of ≥ 25%, and background in the energy range greater than 3 MeV of ≤ 1 count/hour [8, 9].

The Sr-90 radionuclide was analyzed by radiochemical separation method to enrich, remove influencing elements, wait for 14 days (to reach the radioactive equilibrium between Sr-90 and Y-90), separate the radionulide of Y-90, and then measure on a low-level background beta counter. The recovery efficiency of the separation process was about 75%. The low-level background alpha/beta counting system (MPC 9300) with gas - flow proportional detector has beta background of 0.02 counts/s, beta counting efficiency of 37% for mixed standard source Sr-90/Y-90 [10].

Quality control programme for the measurements was conducted by using the IAEA standard reference materials (SRM) samples, and by IAEA proficiency tests to determine Cs-134, Cs-137, Sr-90, Po-210 and Pu-239,240 radionuclides.

III. RESULTS AND DISCUSSION

Results of radioactivities in coastal seawater at monitoring points of Ninh Thuan *(NT)*, Vung Tau *(VT)* and Ca Mau *(CM)* provinces during the survey in the year of 2018 are presented in Table I and shown on Figs. 2 and 3.

Table I. The radioactivities of radionuclides in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau provinces during the survey in the year of 2018

Lab. code	***Radionuclide***						
	Ra-226, mBq/L	***Th-232, mBq/L***	***U-238, mBq/L***	***Po-210, mBq/L***	***Cs-137, mBq/L***	***Sr-90, mBq/L***	***Pu-239,240, µBq/L***
NT 02-18	3.58 ± 0.64	5.75 ± 2.80	6.37 ± 2.48	1.51 ± 0.08	1.57 ± 0.42	1.31 ± 0.33	3.7± 0.6
NT 05-18	3.60 ± 0.49	3.45 ± 0.71	3.24 ± 0.72	1.87 ± 0.12	1.05 ± 0.37	1.38 ± 0.25	4.4 ± 0.7
NT 08-18	4.14 ± 0.76	4.78 ± 1.55	5.29 ± 0.69	1.53 ± 0.03	1.36 ± 0.40	1.32 ± 0.33	5.3 ± 1.1
NT 11-18	3.71 ± 0.65	5.98 ± 0.64	4.36 ± 0.65	1.86 ± 0.10	0.88 ± 0.27	1.08 ± 0.27	4.5 ± 1.0
VT 02-18	2.71 ± 0.94	5.04 ± 1.28	3.02 ± 0.45	2.20 ± 0.11	1.41 ± 0.37	1.18 ± 0.28	5.4 ± 0.9
VT 05-18	5.08 ± 1.18	6.63 ± 0.94	5.36 ± 1.00	2.53 ± 0.12	1.16 ± 0.19	1.45 ± 0.44	3.4 ± 0.8
VT 08-18	6.00 ± 1.37	8.50 ± 2.36	6.98 ± 1.48	1.61 ± 0.05	1.68 ± 0.38	1.86 ± 0.55	3.2 ± 0.7
VT 11-18	5.30 ± 0.70	9.07 ± 1.06	6.00 ± 0.70	2.46 ± 0.13	1.29 ± 0.33	1.63 ± 0.30	4.8 ± 1.0
CM 02-18	15.91 ± 2.39	35.93± 5.17	20.93± 2.74	7.64 ± 0.23	1.70 ± 0.44	1.65 ± 0.37	6.2 ± 0.9
CM 05-18	5.06 ± 1.02	15.77± 3.10	7.88± 1.78	4.23 ± 0.20	1.20 ± 0.41	1.58 ± 0.24	2.6 ± 0.6
CM 08-18	6.61 ± 1.01	18.72± 2.94	9.18± 1.89	5.30 ± 0.21	1.37 ± 0.19	1.27 ± 0.30	3.2 ± 0.8
CM 11-18	14.73 ± 2.31	33.47± 3.76	21.47± 2.18	6.74 ± 0.23	1.68 ± 0.44	1.77 ± 0.37	5.1 ± 0.9

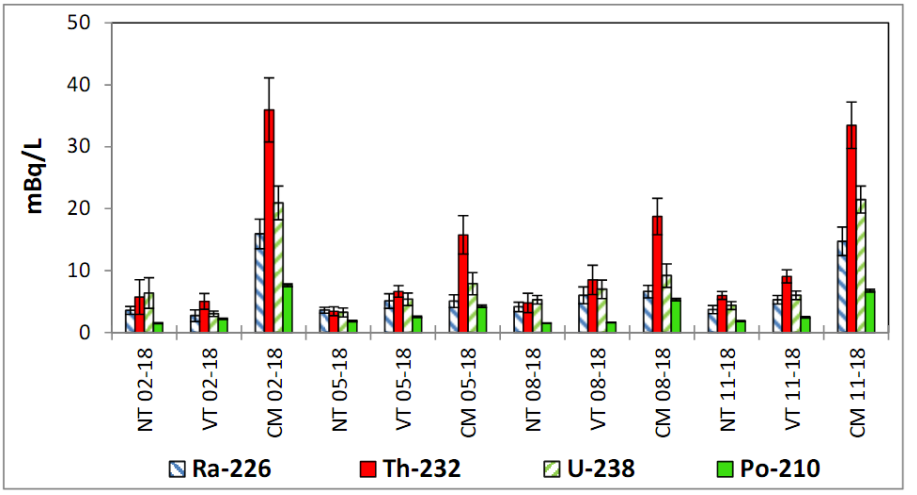

Fig. 2. Variability of radioactivity of Ra-226, Th-232, U-238 and Po-210 in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau provinces

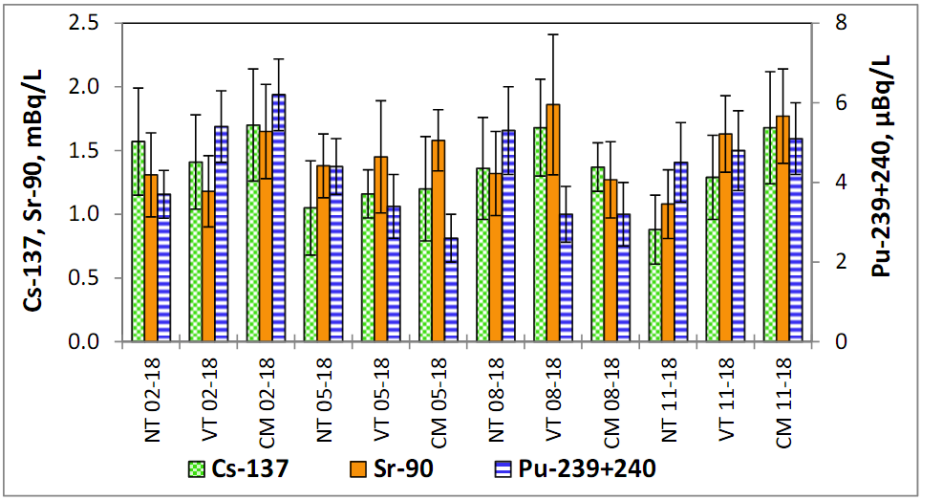

Fig. 3. Variability of radioactivity of Cs-137, Sr-90 and Pu-239,240 in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau provinces

Some physicochemical parameters such as pH, total suspended solids (TSS), conductivity, etc., were measured along with the radioactivity of radionuclides in coastal seawater samples. The results of physicochemical parameters in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau in the year of 2018 are presented in Table II.

The obtained data at monitoring point of Ninh Thuan show that the physicochemical parameters such as pH, TSS, conductivity, and salinity of seawater were not much changed in 2018. At monitoring point of Vung Tau, the TSS parameter was a tendency to increase from February to November. At monitoring point of Ca Mau, the TSS parameter exceeded the permissible limit about 10 times in February and November (QCVN 10-MT: 2015/BTNMT) and had a wide range of fluctuations. Other indicators at the both monitoring points of Vung Tau and Ca Mau had not much fluctuation range.

Table II. The results of physicochemical parameters in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau provinces during the survey in the year of 2018

Monitoring point	*Lab. code*	*pH*	*TSS (mg/L)*	*Conductivity (mS/cm)*	*Salinity (‰)*
Ninh Thuan	NT 02-18	7.55	1.62	47.8	33.4
	NT 05-18	7.41	3.00	48.3	34.2
	NT 08-18	7.62	2.36	47.8	33.8
	NT 11-18	7.75	2.10	46.4	32.5
Ba Ria – Vung Tau	VT 02-18	7.78	8.08	48.2	34.0
	VT 05-18	7.97	10.50	45.7	32.3
	VT 08-18	7.54	21.02	44.7	31.4
	VT 11-18	7.52	36.66	47.9	33.4
Ca Mau	CM 02-18	7.45	513.26	36.8	25.2
	CM 05-18	7.54	56.80	43.5	30.6
	CM 08-18	7.44	90.54	42.0	29.3
	CM 11-18	7.32	481.34	41.4	28.5

At the monitoring point of Ninh Thuan, the radioactivity of Ra-226 did not have any significant changes, and the average radioactivity of Ra-226 in seawater was of 3.76 mBq/L. Meanwhile, the average radioactivity of Ra-226 in seawater in Ca Mau was of 10.58 mBq/L, and larger than in Ninh Thuan (4.77 mBq/L) and Vung Tau (3.76 mBq/L) about 2.8 and 2.2 times, respectively.

The average radioactivity of Th-232 at the monitoring point of Ninh Thuan was of 4.74 mBq/L. After 4 monitoring periods, it was found that the radioactivity of Th-232 tended to increase slightly from February to November. The average radioactivity value of Th-232 at the monitoring point of Ca Mau was of 25.97 mBq/L, which about 7 times higher than the monitoring points of Ninh Thuan and Vung Tau in February, November (dry season) and 4 times in May, August (rainy season).

The average radioactivity of U-238 was of 4.82 mBq/L at the monitoring point of Ninh Thuan and it had no significant change in comparison of previous surveys (from the year of 2014 to 2017) [11]. For the monitoring point of Vung Tau, the average radioactivity of U-238 was of 5.34 mBq/L and the range was of 3.02 ÷ 6.98 mBq/L. For the monitoring point of Ca Mau, the radioactivity of U-238 was quite wide, ranging from 7.88 to 21.47 mBq/L with the maximum value falling in February and November. For the monitoring point of Ca Mau, the radioactivity of U-238 was about from 3 to 7 times higher than those in Ninh Thuan and Vung Tau in February and November, and about from 2 to 3 times in May and August.

The radioactivity of Po-210 in coastal seawater at the monitoring point of Ninh Thuan shown that there was no significant change among the observations, and the range of radioactivity was within of 0.16 ÷ 2.05 mBq/L that was in correspondence with the value range of previously surveys from 2014 to 2017 [11]. The radioactivity of Po-210 in seawater at monitoring point of Vung

Tau was the same level as in Ninh Thuan. Particularly, the radioactivity of Po-210 in coastal seawater at the monitoring point of Ca Mau ranged from 4.23 ÷ 7.64 mBq/L that was higher than about 2 to 4 times in comparison of those of Vung Tau and Ninh Thuan. The variability of radioactivity of Po-210 was the same as the radioactivities of Ra-226, Th-232 and U-238.

The relationship between the radioactivities of Ra-226, Th-232, U-238, Po-210 and TSS in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau were shown on Fig. 4.

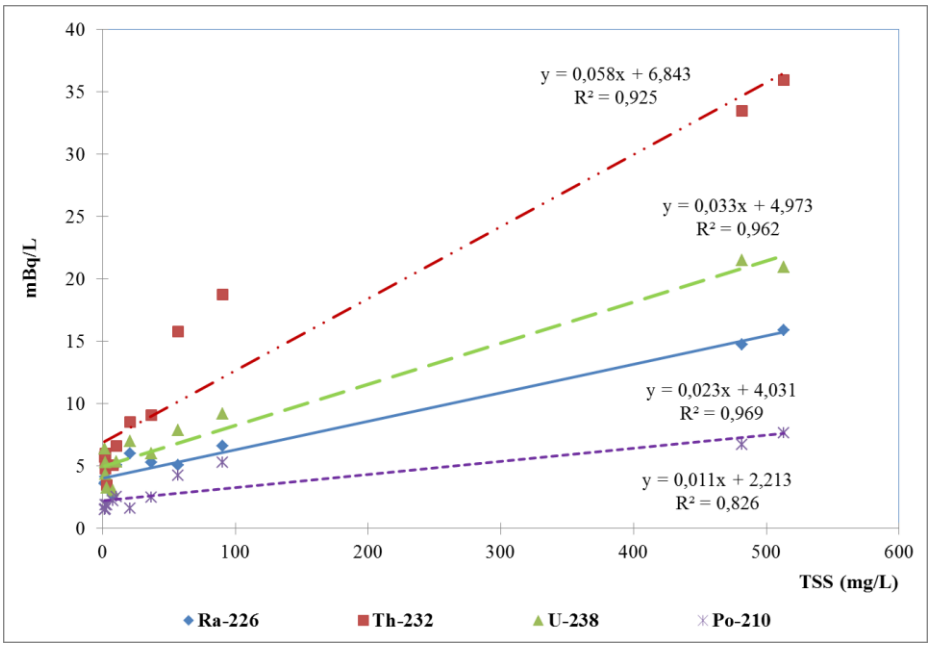

Fig. 4. The relationship between the radioactivities of Ra-226, Th-232, U-238, Po-210 and total suspended solids in coastal seawater at the monitoring points of Ninh Thuan, Vung Tau and Ca Mau

The average radioactivity of Cs-137 in coastal seawater at the monitoring point of Ninh Thuan was of 1.22 mBq/L and it had no significant change in comparison with the previous surveys [11]. The radioactivities of Cs-137 at monitoring points of Vung Tau and Ca Mau were the same level in comparison with those of the monitoring point of Ninh Thuan.

According to the monitoring results, the radioactivity of Sr-90 in coastal seawater at the monitoring point of Ninh Thuan was no change among observations and in correspondence with the value of previous surveys (from 2014 to 2017) [11]. The radioactivities of Sr-90 in coastal seawater at the monitoring points of Vung Tau and Ca Mau were the same level in comparerison of the monitoring point of Ninh Thuan, and the range of radioactivity of Sr-90 was of 1.08 ÷ 1.86 mBq/L.

The radioactivity of Pu-239,240 in coastal seawater at the monitoring point of Ninh Thuan was an average value of 0.0045 mBq/L. It was found that there was no significant change in comparison with the previous surveys (from 2014 to 2017) [11]. At the monitoring points of Vung Tau and Ca Mau, the levels of radioactivity of Pu-239,240 did not have significant fluctuations among the observations.

By analyzing the correlation between the radioactivity of natural radionuclides and TSS, it is shown that the higher of TSS in coastal seawater tended to the higher of radioctivities of Ra-226, Th-232, U-238 and Po-210 (Fig. 4). This explains that the radioactivity of Ra-226, Th-232, U-238 and Po-210 in coastal seawater at the monitoring point of Ca Mau was higher than at other monitoring points (Table I) [11, 12].

The ranges of radioactivity of natural and artificial radionuclides in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau provinces and the data of Asia - Pacific region in 2018 were presented in Table III.

Table III. Ranges of radioactivity of natural and artificial radionuclides in coastal seawater at monitoring points of Ninh Thuan, Vung Tau and Ca Mau provinces and the data of Asia - Pacific region in 2018

Radionuclide	***Ninh Thuan***	***Ba Ria - Vung Tau***	***Ca Mau***	***Asia - Pacific region*** [12]
Ra-226 *(mBq/L)*	3.58 ÷ 4.14	2.71 ÷ 6.00	5.06 ÷ 15.91	0.50 ÷ 4.14
Th-232 *(mBq/L)*	3.45 ÷ 5.75	5.04 ÷ 9.07	15.77 ÷ 35.93	5.27
U-238 *(mBq/L)*	3.24 ÷ 6.37	3.02 ÷ 6.98	7.88 ÷ 21.47	1.00 ÷ 5.29
Po-210 *(mBq/L)*	1.51 ÷ 1.87	1.61 ÷ 2.53	4.23 ÷ 7.64	0.20 ÷ 1.84
Cs-137 *(mBq/L)*	0.88 ÷ 1.57	1.18 ÷ 1.86	1.20 ÷ 1.70	0.13 ÷ 2.95
Sr-90 *(mBq/L)*	1.08 ÷ 1.38	1.16 ÷ 1.68	1.27 ÷ 1.77	0.67 ÷ 2.01
Pu-239,240 *(µBq/L)*	3.7 ÷ 5.3	3.2 ÷ 5.4	2.6 ÷ 6.2	5.3

The obtained results shown that the radioactivities of the above-mentioned radionuclides in coastal seawater at monitoring points of Ninh Thuan and Vung Tau was similar to those in the Asia-Pacific region. At monitoring points of Ca Mau, the radioactivities of natural radionuclides were higher in comparison with the published levels in the Asia-Pacific region, especially for Th-232 and U-238 due to the high TSS.

IV. CONCLUSIONS

The average value of radioactivity of concerned natural and artificial radionuclides in coastal seawater at the monitoring points of Ninh Thuan and Vung Tau was within the value of the Asia Pacific region. At the monitoring point of Ca Mau, the average radioactivities of the natural radionuclides was higher than the values at the monitoring points of Ninh Thuan and Vung Tau from 1.0 to 5.5 times. For artificial radionuclides, the variations of radioactivity were not significant among the monitoring observations as well as the monitoring points.

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