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Assessing the radiological risks associated with primarily natural radioactivities of coastal seawater in northen Vietnam using the erica software

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Abstract: The activity concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th (^{228}Ac), and ^{238}U (^{214}Bi) were determined in marine sediments, seawaters and seafood along the Gulf of Vietnam to establish baseline data for future environmental monitoring at a surface water depth of 0–3 cm. The concentration of uranium, thorium and radium were determined using a low background gamma spectrum as well as activity of ^{238}U(^{214}Bi), ^{232}Th(^{228}Ac) and ^{226}Ra. The mean radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{238}U were found to be 8.59 ± 0.54, 1.31 ± 0.15, and 6.91 ± 0.64 Bq m^{-3}, respectively, in seawater samples and 32.96 ± 1.90, 37.64 ± 1.91, and 39.28 ± 1.96 Bq kg−1, respectively, in marine sediments, 0.21 ± 0.03, 0.69 ± 0.11, and 0.19 ± 0.03 Bq kg−1, respectively, in fish samples and 0.23 ± 0.041, 0.41 ± 0.06, and 0.31 ± 0.06 Bq kg−1, respectively, in clam samples. The radioactivity concentrations in seawater are higher than those in sediment and compared with those reported in other countries. The mean values of distribution coefficient (L/kg) is 0.53, 0.13, and 0.23, respectively, in fish samples and 0.19, 0.16 and 0.13, respectively, in clam samples at Hai Phong, Quang Ninh and Ha Tinh. Moreover, the ecological dose at Hai Phong, Quang Ninh and Ha Tinh are 0.03, 0.02 and 0.02 µGy h−1, respectively, in fish and 0.02, 0.03, and 0.03 µGy h−1, respectively, in clams and the mean human's seafood consumers dose rate is 1.13×10^{-6} Sv/yrs. Results were discussed and compared with those reported in similar studies and with internationally recommended values within limits recommended by UNSCEAR.

Keywords: *Radioactivity concentrations, distribution coefficient, Erica software, dose rate, human's seafood consumers dose.*

I. INTRODUCTION

In the marine environment, the sea is a complex system containing different components (water column, suspended particulates, colloids, sediments, and organic matter) and inhabited by life forms at multiple scales (from plankton to large mammals), undergoing complex interactions. With the arrival of nuclear technology in the late 1940s, a variety of man-made radionuclides have entered the marine environment, either as a result of military operations, industrial discharges, medical releases, or nuclear accidents. This has resulted in their widespread distribution, cycling across the sea and uptake by biota, both locally (in the vicinity of discharge points) and globally [1]. Consequently, the marine lives for example fish, shellfish, etc. are contaminated. To assess the dispersion, transport and lifetime as the retention

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time of a pollutant in an ocean area, it is common to use general ocean cycle models in combination with the diffusion model with the input data source such as wind direction, wind speed, regional currents, tides, topography, etc. [2-3].

In recent years, the study of radioactivity in the marine environment has been of great interest because this is a flexible environment, spreading without borders, so it has received profound attention at the national, regional and international levels. The concentration of these radioactive isotopes depends on the geographical area, so every country should calculate their dose rate on living organisms, seafood and minerals which affects people in the area [4-6]. Naturally occurring radioisotopes in the marine environment of interest are ^{238}U, ^{232}Th and ^{226}Ra. In the world and neighboring countries, the study of these natural isotopes in the marine environment has been receiving great attention. Many authors have developed different methods and techniques for radioactivity determination in water - sediment - seafood [4, 7-8]. Research on evaluation and determination of radioactive concentration in seawater, sediments, and fish [8-11]. Furthermore, the bioaccumulation and dose rate of marine organisms and sediments have been studied, and calculated in different studies [8-13]. Models and methodologies have been developed to assess the impact of radioactivity from nuclear facilities, particularly radioactivity on marine environments and marine life [14-15, 19]. However, deep-sea radioactivity and radiation exposure of these radionuclides on marine organisms have not been extensively studied. The exposure of humans to marine radioactivity by consuming marine products in the long term could lead to potential health risks. Nowadays, many scientists have acknowledged the stochastic effects of radiation on the human body from a low dose rate where the effect is proportional to the dose [17]. Fish is considered to be the most radiation-sensitive aquatic organism. Many researchers studied the biological effects of radiation on fish after a nuclear power plant accident [14-19].

In Vietnam, the application of radioisotope analysis techniques in the general environment and particularly seawater is an important topic. In this study, radioactivity of seawater, seafood, sediment and effective dose rate for local people in the different regions of Vietnam such as Quang Ninh, Hai Phong and Tra Co over different periods are calculated to monitor the environment. The studied regions are the vital socio-economic geographical positions of Vietnam. In addition, these areas also have the flow of fresh water from the Red River and the western East Sea system to Vinh gate, passing Quynh Chau which is a strait located between Loi Chau peninsula and Hainan island.

II. MATERIALS AND METHODS

A. Description of the study site

The Gulf of Tonkin is a saltwater bay located between Vietnam and China. With an area of about 126,250 km², the Gulf of Tonkin is the northwestern branch of the East Sea and part of the Pacific Ocean. The bay has two estuaries including the Quynh Chau strait with 35.2 km wide between the Loi Chau peninsula and Hai Nam island in China. The main gate of the bay is identified as a straight line from Con Co island, Quang Tri province, Vietnam and Oanh Ca Cape, Hai Nam, China, 110 nautical miles (about 200 km) wide.

The Gulf of Tonkin (Quang Ninh, Hai Phong and Ha Tinh) are the upstream locations of ocean currents that move towards Vietnam, especially in the Northeast season. Seawater, sediment, fish and clam samples were collected along the Gulf of Tonkin at Quang Ninh, Hai Phong and Ha Tinh 4 times: December 2018, February 2019, June 2019 and October 2019.

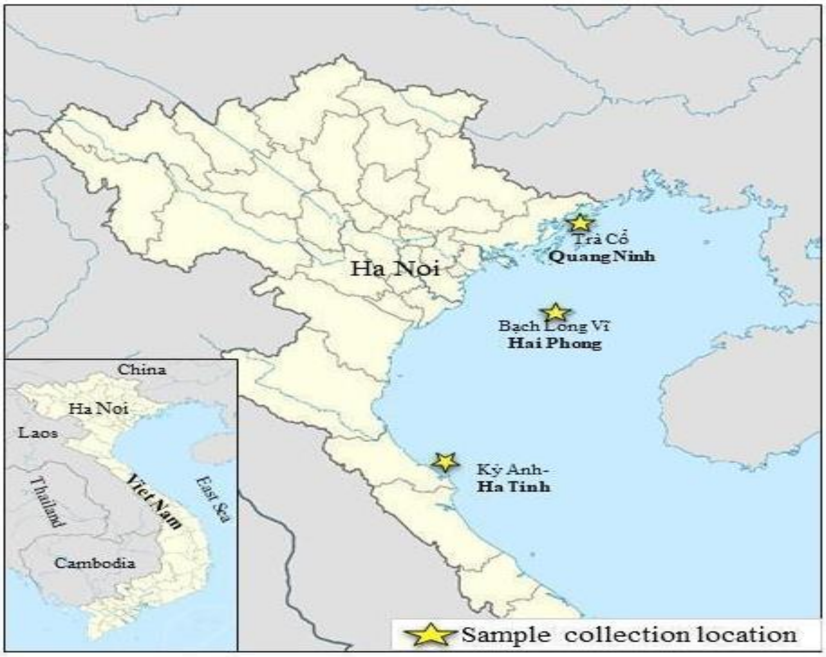

Fig. 1. Locations of 3 selected sampling areas

B. Methods of determination

Radioactivity analysis with low-background gamma spectrometry is the traditional method to determine the low activity concentration of environmental samples. Samples were sealed in about 30 days to reach the radioactive equilibrium between Ra and Rn. The specific activity of seawater, sediment, fish and clam samples was measured by a low-background gamma measurement system (model GX3019) with a relative detector efficiency of 30% and the resolution of 1.90 keV at 1332 keV of ^{60}Co. The integral background of the 100-2000 keV region is about 2 pulses/second. The specific activity of ^{238}U were determined based on the gamma-ray peaks of its daughters: 63 keV of ^{234}Th and 186 keV of ^{235}U and ^{226}Ra. The specific activity of ^{232}Th were determined based on the 911 keV peak of ^{228}Ac. The specific activity of ^{226}Ra were determined based on the gamma-ray peaks of its daughters: 352 keV of ^{214}Pb, and 609 and 1764 keV of ^{214}Bi. Samples were measured on the HPGE detector about 24 hours to ensure statistical counts. The measurement efficiency of the HPGE detector was determined by IAEA standards. Quality assurance and quality control in the laboratory is a time/month according to sample analysis as the Vietnam requirements of ISO/IEC 17025:2005; Annually year, the accuracy and precision method are evaluated by the analysis of international comparative samples organized by the IAEA-RML (e.g. IAEA 375 Russian soil, IAEA CU 2009 03 Moss soil) [9, 21].

C. Distribution coefficient

For aquatic ecosystems (Kd is the distribution coefficient used to describe the ratio of radionuclides concentrations in sediments, fish, shellfish and in water).

Distribution coefficient [8]:

$$Kd = \frac{A_S}{A_n} \quad (1)$$

Where As - The radioactivity of marine organisms (Bq/kg fresh);

An - The radioactivity of seawater (Bq/L).

D. ERICA software

ERICA software was used to assess the radionuclide's impact on the marine environment, in this case are the 3 areas of the Gulf of Tonkin. ERICA is a flexible computational software according to the ERICA integrated approach to biological risk assessment in organisms, the basis for the ERICA integrated approach is the concept of reference organism, exposure dose, effect dose, and animal groups compatible with ICRP's assessment [13, 20-22].

In Erica software, there are two basic calculation steps: estimating the activity concentration of radionuclides and calculating the dose rate in organisms. The dose rate value of organisms were calculated based on the Internal Dose Rate D_{int}^{b} and the External Dose Rate $\mathrm{D}_{\mathrm{ext}}^{\mathrm{b}}$ by ERICA.

E. Human seafood consumer

In this study, we chose fish and clams to represent marine life of the areas for input data because the natural radionuclides inside the human body are from mainly food and drinking water sources. The study organisms are the main food source of the local population as well as the other surrounding areas.

According to FAO in Vietnam, the average seafood consumption is of 22.6 kg/year [23]. The dose rate calculations are the Erica model as the following function:

$$D = A_B.Intake.DCF \quad (2)$$

Where:

D is consumer dose rate (Sv/year);

A_B is the activity concentrations in consumable biota (Bq/kg);

Intake is Consumption rates (kg/year);

DCF is dose conversion factor from ICRP 119 or equivalent or use model (Sv/Bq) [15, 24].

III. RESULT

A. Radioactivity results

The activities of ^{238}U, ^{232}Th and ^{226}Ra isotopes in 4 different sample types in the Gulf of Tonkin over time periods are given in Table I. Generally, the table shows that the survey data is inconstant in different times of survey. However, the gamma spectrometry technique might not give the exact results of these radionuclides in seawater and marine creatures because the disequilibrium between ^{238}U, ^{232}Th, ^{226}Ra and their daughters is existing in the marine environment.

B. Assessment of distribution coefficient

The Table II shows the distribution coefficient of natural radioactive isotopes ^{226}Ra, ^{238}U and ^{232}Th in fish, clams in Quang Binh, Hai Phong and Ha Tinh of Vietnam.

C. Ecological dose calculation results from Erica software

^{226}Ra, ^{238}U and ^{232}Th activities in the 3 areas as well as environmental parameters (distribution coefficient, concentration factors) were used as input data to the software of the Erica program. We calculated biological doses in Fish and Clams. The results are showed in the below Table III.

D. Consumable biota dose rate in a year

The assessment of dose rate and human consumption of marine organisms within 1 year using the Erica tool is extremely useful because it has established a method to estimate the dose rate for different species of organisms with the distinct ecological environments of the other regions. Especially, the background radiation data of Vietnam's marine environment is combined with the other values of artificial radioactive elements that will help scientists having a basis to further study the effects of radiation on the surrounding environment.

Table I. Radioactive activities of ^{226}Ra, ^{238}U and ^{232}Th in seawater, sediment and seafood samples in Quang Ninh, Hai Phong, Ha Tinh from December 2018 to October 2019

Time	Activity											
	Quang Ninh				**Hai Phong**				**Ha Tinh**			
	Water ***(mBq/L)***	Sediment ***(Bq/kg)***	Fish ***(Bq/kg)***	Clams **(Bq/kg)**	Water **(mBq/L)**	Sediment **(Bq/kg)**	Fish **(Bq/kg)**	Clams **(Bq/kg)**	Water **(mBq/L)**	Sediment **(Bq/kg)**	Fish **(Bq/kg)**	Clams **(Bq/kg)**
	^{226}Ra											
Dec-18	7.03	29.20	0.09	0.37	4.51	33.45	0.28	0.30	7.22	30.43	0.37	0.55
Feb-19	8.05	28.42	0.22	0.45	7.10	38.65	0.23	0.12	6.47	31.24	0.20	0.23
Jun-19	11.3	26.19	0.12	0.15	7.86	32.35	0.19	0.15	7.44	23.35	0.15	0.06
Oct-19	8.95	35.26	0.17	0.25	7.01	49.75	0.97	0.12	11.44	20.61	0.13	0.05
Average	**8.83±1.03**	**29.76±3.83**	**0.15±0.03**	**0.31±0.06**	**6.62±0.63**	**38.55±3.23**	**0.42±0.07**	**0.17±0.03**	**8.14±0.92**	**26.41±3.36**	**0.21±0.06**	**0.22±0.08**
	^{238}U											
Dec-18	8.67	40.60	0.19	0.16	7.05	42.27	0.27	0.54	7.69	35.68	0.09	0.31
Feb-19	4.79	37.08	0.12	0.93	4.62	43.81	0.14	0.03	7.97	33.27	0.26	0.72
Jun-19	5.15	35.08	0.20	0.19	2.85	40.26	0.10	0.10	5.58	26.51	0.04	0.07
Oct-19	4.53	35.62	0.12	0.18	4.22	49.75	0.76	0.08	12.04	23.81	0.09	0.05
Average	**5.79±1.56**	**37.10±6.57**	**0.16±0.05**	**0.37±0.07**	**4.68±0.76**	**44.02±9.45**	**0.32±0.09**	**0.19±0.05**	**8.32±1.55**	**29.82±6.41**	**0.12±0.03**	**0.29±0.12**
	^{232}Th											
Dec-18	1.79	39.46	0.26	0.94	0.49	37.00	1.49	0.27	1.16	35.82	1.45	0.47
Feb-19	2.32	39.03	1.33	0.75	0.66	46.10	0.38	0.22	1.00	41.89	0.61	0.61
Jun-19	2.30	36.20	0.30	0.33	0.83	27.10	0.71	0.39	0.91	37.43	0.10	0.19
Oct-19	1.62	44.21	0.77	0.75	0.80	32.24	1.06	0.53	1.22	27.51	0.72	0.09
Average	**2.01±0.24**	**39.72±4.45**	**0.67±0.17**	**0.69±0.10**	**0.66±0.12**	**35.61±4.96**	**0.91±0.12**	**0.35±0.09**	**1.07±0.18**	**35.66±4.53**	**0.72±0.17**	**0.34±0.07**

Table II. Distribution coefficient (L/kg) of naturally radioactive isotope ^{226}Ra, ^{238}U and ^{232}Th in biological samples in Quang Ninh, Hai Phong and Ha Tinh

Time	Quang Ninh		Hai Phong		Ha Tinh	
	Fish	***Clams***	**Fish**	***Clams***	**Fish**	***Clams***
			^{226}Ra			
Dec-18	0.013	0.053	0.062	0.067	0.051	0.076
Feb-19	0.027	0.056	0.032	0.017	0.031	0.036
Jun-19	0.011	0.013	0.024	0.019	0.020	0.008
Oct-19	0.019	0.028	0.138	0.017	0.011	0.004
Average	**0.017**	**0.037**	**0.064**	**0.030**	**0.028**	**0.031**
			^{238}U			
Dec-18	0.022	0.018	0.038	0.077	0.012	0.040
Feb-19	0.025	0.194	0.030	0.006	0.033	0.090
Jun-19	0.039	0.037	0.035	0.035	0.007	0.013
Oct-19	0.026	0.040	0.180	0.019	0.007	0.004
Average	**0.028**	**0.072**	**0.071**	**0.034**	**0.015**	**0.037**
			^{232}Th			
Dec-18	0.145	0.525	0.041	0.551	0.250	0.405
Feb-19	0.573	0.323	0.576	0.333	0.610	0.610
Jun-19	0.130	0.143	0.855	0.470	0.110	0.209
Oct-19	0.475	0.463	0.325	0.663	0.590	0.074
Average	**0.331**	**0.364**	**0.449**	**0.504**	**0.390**	**0.324**

Table III. Results of assessment of dose rate (μGy/h) in fish and clams at Quang Ninh, Hai Phong and Ha Tinh by Erica software

Organisms	**Fish**			**Clams**		
Isotopes	***^{226}Ra***	***^{232}Th***	***^{238}U***	***^{226}Ra***	***^{232}Th***	***^{238}U***
Quang Ninh	4.15×10^{-2}	1.5×10^{-2}	8.9×10^{-3}	5.8×10^{-2}	1.6×10^{-2}	8.9×10^{-3}
Hai Phong	5.62×10^{-2}	2.1×10^{-2}	7.7×10^{-3}	4.2×10^{-2}	8×10^{-3}	4.6×10^{-3}
Ha Tinh	2.8×10^{-2}	1.7×10^{-2}	2.9×10^{-3}	4.37×10^{-2}	7.8×10^{-1}	7×10^{-3}

Table IV. Population dose (Sv/year) for a person consuming fish and clams in 1 year

The areas	***Quang Ninh***	***Hai Phong***	***Ha Tinh***
Dose for a person when consuming seafood in 1 year	7×10^{-7}	7×10^{-7}	2×10^{-6}

These dose values are all less than the dose limit for the population of 1mSv/year according to UNSCEAR 2000.

IV. CONCLUSION

From the research results, activities of the isotope ^{226}Ra, ^{238}U and ^{232}Th were identified in different sample objects (sea water, sediment, fish and clams) in different geographical areas of the Gulf of Tonkin. These values are useful for future radioactive monitoring programs in Vietnam. Moreover, the dose values of a person consuming seafood in 1 year calculated by ERICA are all less than the dose limit for the population of 1mSv/year according to UNSCEAR 2000. Therefore, Erica ecosystem risk assessment model is suitable to annually monito and survey the radioactive background in Viet Nam and give warnings to managers on which can be taken appropriate treatment for the impact of the radio activities concentration on the environment.

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