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Radionuclides in *Scylla serrata* (Forsk., 1775) of Chakaria Sundarban Area, Bangladesh

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Abstract

The ^{226}Ra , ^{228}Ra , ^{232}Th , ^4K and ^{137}Cs contents were determined for one year in edible and inedible parts of *Scylla serrata* from Chakaria Sundarban area of Bangladesh coastal water. One factor ANOVA showed the significant differences of radionuclide concentrations in edible and inedible parts of *S. serrata* at 1% level (diff. for ^{226}Ra , $F = 66.524$, $df = 23$, $P = 0.0001$; ^{228}Ra , $F = 86.44$, $df = 23$, $P = 0.0001$; ^{232}Th , $F = 80.01$, $df = 23$, $P = 0.0001$ and ^4K , $F = 21.593$, $df = 23$, $P = 0.0001$). The ^{226}Ra concentration varied from 1.49 ± 0.09 to 3.69 ± 0.18 Bq kg^{-1} in edible portions of *S. serrata* in fresh weight (fw), with highest value recorded in November and the lowest in April. The ^{226}Ra activities of *S. serrata* in inedible parts ranged from 4.19 ± 0.41 to 12.50 ± 0.95 Bq kg^{-1} fw and the maximum value was found in June, minimum in April. The ^{228}Ra concentration ranged between 2.45 ± 0.11 and 5.76 ± 0.25 Bq kg^{-1} fw and the maximum concentration in edible parts was in January and minimum in June. The concentrations of ^{228}Ra ranged between 7.99 ± 0.54 and 18.77 ± 0.96 Bq kg^{-1} fw in the inedible parts, the maximum concentration in January and minimum in June. The activity of ^{232}Th varied between 2.34 ± 0.10 and 6.40 ± 0.25 Bq kg^{-1} fw as observed highest in January and lowest range during July in edible parts. The ^{232}Th varied between 9.96 ± 0.87 and 27.21 ± 1.21 Bq kg^{-1} fw in the inedible parts and highest was found in January and lowest in July. The ^4K content from the edible portion was found 65.35 ± 2.04 to 164.40 ± 3.91 Bq kg^{-1} fw, the highest value was recorded in July and lowest was in January. The ^4K activity was found 115.02 ± 2.99 to 287.37 ± 5.06 Bq kg^{-1} fw in inedible parts of *S. serrata* and highest concentration was found in July and lowest in January. The concentration of ^{137}Cs was below detection

level in both edible and inedible parts. Correlation analyzes showed a positive relationship between ^{228}Ra and ^{232}Th ($r = 0.821$, $P = 0.01$, $N = 10$); and negative correlation with ^{228}Ra and ^4K ($r = -0.646$, $P = 0.01$, $N = 10$) and as well as ^{232}Th and ^4K ($r = -0.574$, $P = 0.01$, $N = 10$). The total annual effective dose equivalent was $39.72 \text{ mSv yr}^{-1}$. This study is the first on radionuclide concentrations in *S. serrata* in Asian countries. Therefore, this result is a useful information regarding human consumption of mud crab.

Introduction

Mud crab (*Scylla serrata*, Portunidae) is the most valuable species among the identified crabs of Bangladesh. This species has a high demand as a human food in the world market due to its palatability and nutritive value. It is also used for poultry and aquaculture feed (Islam 1976; Zafar and Siddiqui 2000). Crab meat contains all the essential amino acids and vitamins necessary to man. Crab meat contains various types of minerals, such as sodium, calcium, and iron (Khan 1992). The demand of *S. serrata* is increasing in Bangladesh day by day. Mud crab can be cultivated through various methods in many countries of Indo-West Pacific region. It is a flourishing industry in the Indo-Pacific regions of the world. Due to the importance of *S. serrata*, works on taxonomy, biology, culture and fattening, biochemical analysis and industrial processing have been done by different researchers (Islam 1976; Khan 1992; Obayed 1998), but study on radionuclides concentration of *S. serrata* is totally absent in the country and as well as in other parts of the world.

Radionuclides are toxic elements and harmful for biotic organisms. They are concentrated by biota, physical and chemical adsorption, ion-exchange, co-precipitation, flocculation, sedimentation and uptake directly from water and through the passage of food web (Rice and Duke 1969). Aquatic organisms concentrate more radionuclides both naturally and artificially by direct absorption from the water and feeding (Polikarpov 1966). Bottom dwellers are the greatest accumulator of radionuclides than that of pelagic species because of adsorption of radionuclides to particles that accumulate on seabed (IAEA 1986). Radionuclides enter the human body through the consumption of seafood (fish, shrimp crab etc.). *S. serrata* may be contaminated by natural and artificial radioactivity in aquatic environment. So, if the mud crab contains radioactivity it may be a subject of internal and external radiation exposure to the human body. Thus, it is essential to measure the level of radioactive contamination. The natural radionuclides, ^{226}Ra , ^{228}Ra , ^{232}Th , and ^4K in the marine environment are obtained from the weathering and recycling of terrestrial minerals and rocks (Clark 1989). Military activities, dumping and sedimentation may be the major sources of radionuclides in the investigated area of Chakaria Sundarban.

The present work determines the concentration of radionuclides ^{226}Ra , ^{228}Ra , ^{232}Th ; and ^4K and ^{137}Cs in the mud crab of Chakaria Sundarban area of Bangladesh coastal water. These findings will help to develop a baseline data on radionuclides concentration of *S. serrata* for the coastal and marine environments.

Materials and Methods

Description of sampling station

Samples were collected from the Chakaria Sundarban area. The geographical location of the sampling area is 21°35' N latitude and 92°10' E longitude (Fig. 1). Sampling was carried out for one year from August 1999 to July 2000. The Chakaria Sundarban is an important area for fish and shrimps spawning and nursery ground (Mahmood 1990). This area is situated in the delta of Mathamuhuri river and the second largest mangrove area of Bangladesh (Karim and Khan 1992). The mangrove area of Chakaria Sundarban was entirely for expansion of shrimp farming (Mahmood 1995). This area is used for salt extraction during pre-monsoon and for aquaculture during the monsoon.

Collection of sample

Year round collection of *S. serrata* sample was carried out monthly using behundi jal (set-bag net) and chai (collapsible bamboo trap). A total of 20 marketable size of mud crab (7.40 – 10.50 cm in shell length, 10.40 – 13.50 cm in shell width and 180.00 – 650.00 gm in body weight of individuals) were collected every month from the Chakaria Sundarban area of Bangladesh. Collected samples were washed with clean water to remove mud, sands, small weeds and gravels; and then chilling method was performed to reduce spoilage.

Preparation of samples

After defrosting *S. serrata* were washed and then separated the edible or soft parts (i.e. body portion and meat of claws) from the inedible parts (i.e. legs, body shells and shells of claws). The separated parts were blotted for removed water and weighed to within ± 0.50 gm for fresh weight (fw). Then weighed samples were chopped and dried at 105 °C in an electrical oven at an anhydrous

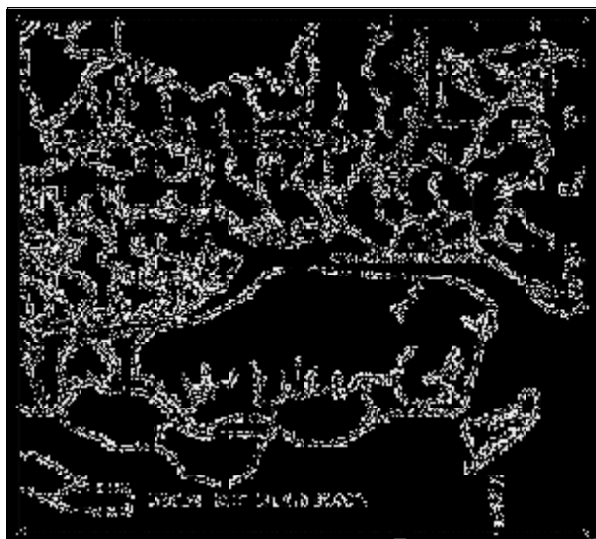


Fig. 1. Inset Bangladesh. Geographical location of the Chakaria Sundarban has been enlarged to show the exact locations of sampling stations (S)

state until a constant weight was obtained. The dried samples were ground to a fine powder and passed through a 2 mm mesh sieve, weighed and then transferred to airtight 150 ml (diameter: 5 cm and height: 7 cm) cylindrical plastic screw-capped containers and finally sealed. The radioactivity measurement of each sample was made minimum one month later to assure secular equilibrium between the ^{226}Ra and ^{232}Th series and most of their respective progeny/daughter (Schotzing and Debertin 1983; El-Tahaway et al. 1994; Ibrahim et al. 1995). The fresh weight and dried weight of samples were about 700 gm and 95 gm for edible parts and 600 gm and 110 gm for inedible parts respectively.

The γ -ray activity measurement

The detection and measurement of γ -ray activities (^{226}Ra , ^{228}Ra , ^{232}Th , ^4K and ^{137}Cs) in both edible and inedible parts of *S. serrata* were carried out by g-spectrometry using a 111 cm³ intrinsic coaxial high resolution HPGe detector with a relative efficiency of 23% and a resolution of 1.80 keV (FWHM) for the peak of the 1332 keV of ^{60}Co . The detector was coupled with 4096-channel computer analyzer. The relative uncertainties of the activities were within $\pm 5\%$. All the samples were counted at a period of 50 kilosecs. To reduce γ -ray background the detector was shielded by a cylindrical thick lead shield 5.08 cm with a fixed bottom and moving cover following Islam et al. (1990). Background spectra for the 50 kilosecs were collected every week and the background counts were subtracted from the respective region of interests (Debertin and Helmer 1980). The energies of 295.21 keV (^{214}Pb), 351.92 keV (^{214}Pb), 609.31 keV (^{214}Bi) and 1120.39 keV (^{214}Bi) were used to determine ^{226}Ra and 911.08 keV (^{228}Ac) and 969.11 keV (^{228}Ac) were used to determine ^{228}Ra activity. The energy of 583.19 keV (^{208}Tl) was used to determine ^{232}Th activity. The energy of 1460.38 keV γ -ray was used for ^4K . The 661.66 keV γ -ray was used to determine ^{137}Cs (IAEA 1989; ICRP 1983; Alam et al. 1995). Only peak γ -ray was observed to have standard deviation of $< 5\%$ during the efficiency calibration used to calculate the specific activity of ^{226}Ra , ^{228}Ra , ^{232}Th , ^4K and ^{137}Cs .

Assessment of effective dose equivalent

Only edible parts of crab calculated based on local knowledge of consumption habit and data was supplied by the Chakaria Fish Businessmen Association, Cox's Bazar. The annual consumption rate per person (adult) of the mud crab is about 0.90 kg•person•yr. in the coastal area of Chakaria. The effective dose equivalent to an individual from an intake of a radionuclide via ingestion of one type of food is calculated using the following formula (Miller et al. 1990; UNCEAR 1988) – $D_{\text{ing.}} = C_{\text{R}} \cdot I_{\text{F}} \cdot E_{\text{D}}$, where, $D_{\text{ing.}}$ is the annual effective dose to an individual due to ingestion of radionuclide (Sv yr⁻¹); C_{R} is the concentration of radionuclide in ingested food (Bq kg⁻¹); I_{F} is the annual intake of food containing radionuclide (kg yr⁻¹) and E_{D} is the ingestion dose conversion factor for radionuclide (Sv Bq⁻¹). E_{D} values

used in the present work are taken from NRPB-R-245 (Phipps et al. 1991) and ICRP-67 (1994).

Therefore, the total dose via, ingestion is calculated using this formula,

$$D_{\text{ing.}} = C_{\text{R}} (^{226}\text{Ra}) \cdot I_{\text{F}} \cdot E_{\text{D}} + C_{\text{R}} (^{228}\text{Ra}) \cdot I_{\text{F}} \cdot E_{\text{D}} + C_{\text{R}} (^{232}\text{Th}) \cdot I_{\text{F}} \cdot E_{\text{D}} + \dots$$

Results

The concentrations of different radioisotopes in this investigation are:

²²⁶Ra

The concentration of ²²⁶Ra ranged from 1.49 ± 0.09 to 3.68 ± 0.18 Bq kg⁻¹ fw (fresh weight) of *S. serrata*, maximum value was found in November and minimum in April in edible parts (Table 1). In the inedible parts ²²⁶Ra activities ranged from 4.19 ± 0.41 to 12.50 ± 0.95 Bq kg⁻¹ fw (Table 2). The maximum value was found in June and the minimum in April. The yearly mean of ²²⁶Ra concentration in edible parts of *S. serrata* was 2.77 ± 0.17 Bq kg⁻¹ (Table 1). The yearly mean of ²²⁶Ra concentration in edible parts of *S. serrata* was 2.77 ± 0.17 Bq kg⁻¹ and 8.81 ± 0.71 Bq kg⁻¹ in inedible parts (Table 2). The annual effective dose of ²²⁶Ra was $6.59 \mu\text{Sv yr}^{-1}$ (Table 3).

²²⁸Ra

The ²²⁸Ra concentration ranged between 2.45 ± 0.11 and 5.76 ± 0.25 Bq kg⁻¹ fw in edible parts (Table 1). The maximum concentration was found in January and the minimum during June in edible parts. In the inedible parts it ranged between 7.99 ± 0.54 and 18.77 ± 0.96 Bq kg⁻¹ fw (Table 2). The

Table 1. Monthly variation of ²²⁶Ra, ²²⁸Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs concentrations of edible parts of *S. serrata*.

Months	Activity in fresh weight (Bq kg ⁻¹ ± 1σ)				
	²²⁶ Ra	²²⁸ Ra	²³² Th	⁴⁰ K	¹³⁷ Cs
August	1.97±0.17	3.76±0.20	3.51±0.19	150.69±3.57	BDL
September	2.49±0.14	4.23±0.23	4.64±0.22	125.08±2.99	BDL
October	3.51±0.13	3.63±0.21	2.37±0.12	104.58±3.07	BDL
November	3.68±0.18	2.97±0.12	3.16±0.18	90.39±3.05	BDL
December	2.75±0.11	4.13±0.25	4.26±0.21	78.25±2.62	BDL
January	3.39±0.19	5.76±0.25	6.40±0.25	65.35±2.04	BDL
February	2.65±0.13	4.07±0.27	3.98±0.20	83.21±2.17	BDL
March	2.43±0.12	5.01±0.22	4.12±0.21	96.01±2.49	BDL
April	1.49±0.09	4.81±0.24	5.01±0.23	100.41±3.01	BDL
May	3.40±0.19	3.70±0.18	3.96±0.18	120.57±2.81	BDL
June	3.57±0.10	2.45±0.11	3.57±0.12	135.99±3.42	BDL
July	2.04±0.10	2.63±0.09	2.34±0.10	164.40±3.91	BDL
Mean ± 1s	2.77±0.17	3.93±0.22	3.98±0.26	109.58±3.65	---

BDL = Below Detection Limit.

maximum concentration occurred in January and the minimum was in June. The yearly mean of ^{226}Ra concentration in edible and inedible parts of *S. serrata* were $3.93 \pm 0.22 \text{ Bq kg}^{-1}$ and $12.78 \pm 0.82 \text{ Bq kg}^{-1}$ respectively. The annual effective dose of ^{228}Ra was $11.46 \mu\text{Sv yr}^{-1}$.

^{232}Th

The activity of ^{232}Th in edible parts varied between 2.34 ± 0.10 and $6.40 \pm 0.25 \text{ Bq kg}^{-1}$ fw, maximum was observed in January and lowest during July (Table 1). The ^{232}Th concentration varied between 9.96 ± 0.87 and $27.21 \pm 1.21 \text{ Bq kg}^{-1}$ fw in the inedible parts, the highest ^{232}Th activity was found in January and lowest in July (Table 2). Correlation analysis showed a positive significant relation between the ^{228}Ra and ^{232}Th concentrations [$r = 0.821$, $P = 0.01$, $N = 10$]. The annual effective dose of ^{232}Th was $15.77 \mu\text{Sv yr}^{-1}$ (Table 3).

Table 2. Monthly variation of ^{226}Ra , ^{228}Ra , ^{232}Th , ^{40}K and ^{137}Cs concentrations of inedible parts of *S. serrata*.

Months	Activity in fresh weight ($\text{Bq kg}^{-1} \pm 1\sigma$)				
	^{226}Ra	^{228}Ra	^{232}Th	^{40}K	^{137}Cs
August	6.50±0.43	12.25±0.85	15.05±1.04	259.18±4.26	BDL
September	7.99±0.50	13.55±0.92	20.72±0.99	218.89±4.01	BDL
October	10.32±0.77	11.84±0.94	10.69±0.85	178.93±3.99	BDL
November	11.40±0.74	9.71±0.61	13.47±0.80	155.18±3.20	BDL
December	8.69±0.56	13.46±0.84	18.32±0.10	136.15±3.19	BDL
January	11.10±0.53	18.77±0.96	27.21±1.21	115.02±2.99	BDL
February	7.13±0.80	13.16±0.73	16.99±0.97	147.25±3.13	BDL
March	6.90±0.39	16.33±0.90	18.37±0.91	168.97±3.67	BDL
April	4.19±0.41	15.58±0.83	24.01±1.15	174.21±3.48	BDL
May	10.88±0.75	12.08±0.86	17.22±1.01	208.59±5.46	BDL
June	12.50±0.95	7.99±0.54	15.17±0.91	238.42±4.86	BDL
July	8.16±0.65	8.59±0.68	9.96±0.87	287.37±5.06	BDL
Mean ± 1s	8.81±0.71	12.78±0.82	17.27±0.95	190.68±4.51	---

BDL - Below Detection Limit.

Table 3. An assessment of radiation dose effective from ingestion of *S. serrata* in human body

Months	Effective dose ($\mu\text{Sv yr}^{-1}$)				
	^{226}Ra	^{228}Ra	^{232}Th	^{40}K	Monthly total
August	0.39	0.91	1.17	0.68	3.15
September	0.49	1.03	1.55	0.56	3.63
October	0.69	0.88	0.79	0.47	2.83
November	0.73	0.72	1.05	0.41	2.91
December	0.54	1.00	1.42	0.35	3.31
January	0.67	1.40	2.13	0.29	4.49
February	0.52	0.99	1.33	0.37	3.21
March	0.48	1.22	1.37	0.43	3.50
April	0.30	1.17	1.67	0.45	3.59
May	0.67	0.90	1.32	0.54	3.43
June	0.71	0.60	1.19	0.61	3.11
July	0.40	0.64	0.78	0.74	2.56
Yearly total	6.59	11.46	15.77	5.90	39.72

⁴⁰K

The ⁴⁰K content was found 65.35 ± 2.04 to 164.40 ± 3.91 Bq kg⁻¹ fw, the highest value was recorded in July and lowest was in January from the edible parts of *S. serrata* (Table 1). The ⁴⁰K activity varied 115.02 ± 2.99 to 287.37 ± 5.06 Bq kg⁻¹ fw. in the inedible parts, the highest concentration was also found in July and lowest in January (Table 1). The yearly mean of ⁴⁰K concentration in edible and inedible parts of *S. serrata* were 109.58 ± 3.65 Bq kg⁻¹ and 190.68 ± 4.51 Bq kg⁻¹ respectively. The ⁴⁰K shows a negative correlation with ²²⁸Ra, [r = - 0.646, P = 0.01, N = 10] and as well as ²³²Th [r = - 0.574, P = 0.01, N = 10]. The annual effective dose of ⁴⁰K was 5.90 μSv yr⁻¹ (Table 3).

¹³⁷Cs

¹³⁷Cs concentration was found below detection level in both edible and inedible parts of *S. serrata*. Therefore, the artificial sources of radionuclides in the study area were not frequent.

Discussion

Alam et al. (1996) reported that the radionuclides concentrations were ²²⁶Ra 0.24 ± 0.09 Bq kg⁻¹ fw; ²³²Th 0.28 ± 0.08 Bq kg⁻¹ and ⁴⁰K 18.65 ± 2.03 Bq kg⁻¹ in *Peneaus indicus* and ²²⁶Ra 0.23 ± 0.06 Bq kg⁻¹; ²³²Th 0.42 ± 0.13 Bq kg⁻¹ and ⁴⁰K 13.36 ± 2.03 Bq kg⁻¹ in *Metapeneaus brevicornis*.

Alam et al. (2002) stated that higher vales of radionuclides concentrations were found in offal part than edible portion of five Cynoglossids (flat fish) in the adjacent area of Chakaria Sundarban at the Kutubdia Channel. They recorded ²²⁶Ra 14.00 ± 6.00 Bq kg⁻¹; ²³²Th 14.00 ± 3.00 Bq kg⁻¹, and ⁴⁰K 192.00 ± 9.00 Bq kg⁻¹ in offal part and ²²⁶Ra 10.00 ± 3.00 Bq kg⁻¹; ²³²Th 11.00 ± 4.00 Bq kg⁻¹, and ⁴⁰K 178.00 ± 18.00 Bq kg⁻¹ in edible portion of *Cynoglossus lingua*.

Higher concentrations of radionuclide of *Crassostrea sp.* in shell portion (²²⁶Ra 11.50 ± 2.00 Bq kg⁻¹ and ²³²Th 7.70 ± 1.50 Bq kg⁻¹) than in the soft tissue (²²⁶Ra 4.20 ± 1.00 Bq kg⁻¹ and ²³²Th 3.20 ± 0.70 Bq kg⁻¹) were recorded by Chowdhury et al. (2003).

Alam et al. (1999) found higher concentrations of different radioisotopes in shell part (²²⁶Ra 14.10 ± 7.40 Bq kg⁻¹; ²³²Th 8.50 ± 2.10 Bq kg⁻¹ and ⁴⁰K 137.00 ± 34.00 Bq kg⁻¹) than whole soft tissue (²²⁶Ra 4.30 ± 1.90 Bq kg⁻¹; ²³²Th 2.60 ± 1.90 Bq kg⁻¹ and ⁴⁰K 80.00 ± 16.50 Bq kg⁻¹) of *Perna viridis* in the Moheshekhali channel (adjacent with study area) and the Karnafully river.

Radionuclide elements in water (²²⁶Ra 3.19 ± 0.83 Bq l⁻¹; ²³²Th 2.91 ± 0.89 Bq l⁻¹ and ⁴⁰K 242.22 ± 48.88 Bq l⁻¹) and soil (²²⁶Ra 14.78 ± 4.44 Bq kg⁻¹; ²³²Th 26.03 ± 3.41 Bq kg⁻¹ and ⁴⁰K 489.80 ± 43.17 Bq kg⁻¹) were reported by Alam et al. (1998) from the present study area in salt pans.

Radium is one of the major sources of the radioactivity found in water and food (Kuo et al. 1997). In the present study major concentrations of ^{226}Ra were also available in the mud crab.

Alam et al. (1995) reported that ^{232}Th content in five small species of shrimps ranged from 0.36 ± 0.10 to 0.78 ± 0.23 Bq kg^{-1} fw.

The ^{40}K concentration was the top most radioisotopes element in *S. serrata*. The ^{40}K represents more than 80% in aquatic components (Lambrechts et al. 1992). Sharifuzzaman (1998) reported that ^{40}K content was higher in the rainy season in flat fish. Here, ^{40}K was higher in monsoon time (June - August).

Williams and Swanson (1958) reported that concentrations of ^{137}Cs depend on cesium-potassium ratio and they also mentioned that high concentrations of potassium reduce the absorption of ^{137}Cs . The present investigation indicates high concentration of ^{40}K , and nondetectable amount of ^{137}Cs . So, in the present investigation ^{137}Cs was not found or it is below detection level.

There is no published report on radionuclide concentrations in *S. serrata* or related other species in the country as well as in other parts of the world.

Alam et al. (1999) reported high concentrations of radionuclides in shell parts than meat parts of *P. viridis*. In the present investigation higher concentrations of radionuclides were found in inedible parts than edible parts of *S. serrata*.

Conclusion

The concentrations of radionuclides (^{226}Ra , ^{228}Ra , ^{232}Th and ^{40}K) in inedible parts were higher than edible parts. One factor ANOVA showed the significant difference ($F = 66.524$, $df = 23$, $P = 0.0001$ for ^{226}Ra ; $F = 86.44$, $df = 23$, $P = 0.0001$ for ^{228}Ra ; $F = 80.01$, $df = 23$, $P = 0.0001$ for ^{232}Th and $F = 21.593$, $df = 23$, $P = 0.0001$ for ^{40}K) of different radionuclide concentrations between edible and inedible parts of *S. serrata*. The present investigation indicated that target species *S. serrata* is not contaminated by anthropogenic radionuclide ^{137}Cs . In the mud crab the total annual effective dose for human health was found $39.72 \mu\text{Sv yr}^{-1}$, which is lower than the ICRP - 60 (1990) recommended standard value (1 mSv yr^{-1}). The natural radionuclides (^{226}Ra , ^{228}Ra , ^{232}Th and ^{40}K) were not in dangerous level. Therefore, the mud crab (*S. serrata*) is a highly acceptable food item for human as can be seen in the present study.

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