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Study of the Naturally Occurring Radionuclide Concentrations and the Estimation of Dose Rates for the Samples Collected from the St. Martin's Island, Chittagong, Bangladesh.

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Abstract. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K radionuclides have been determined for Sediment, Fish and Coral Samples collected from the St. Martin's Island, Chittagong, Bangladesh. A total of 7 Samples (4 sediments, 2 fishes and 1 coral) from 3 different kinds were analyzed by using a High Purity Germanium (HPGe) detector of relative efficiency of 38%. For sediment samples, the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K have been found 17.243±2.14, 25.207±2.302 and 460.802±50.957 Bq/Kg, respectively. The mean activity concentrations of fish samples for the corresponding radionuclide were 10.86±16.615, 10.555±13.51 and 364.92±361.345 Bq/Kg respectively. Also for coral sample, the mean activity concentrations for the corresponding radionuclide were13.06±2.1, 16.57±2.65 and 84.96±43.29 Bq/kg, respectively. The radionuclide ^{137}Cs has not been detected in any of the samples. For sediment samples, the mean radiological hazard parameter values of outdoor absorbed dose rate, indoor absorbed dose rate, external radiation hazard, internal radiation hazard, annual effective dose equivalent, radium equivalent activity and representative level index were 43.863±2.877, 52.636±3.452 nGy.h^{-1}, 0.239±0.015, 0.286±0.018, 53.793±3.525 µSvy^{-1}, 88.695±5.672 Bq/kg, and 0.673±.04375 respectively. For fish samples, the mean values of the corresponding radiological indices were 27.368±19.606, 32.841±23.52 nGy.h^{-1}, 0.146±0.103, 0.175±0.129, 33.563±24.227µSvy^{-1}, 54.052±38.446 Bq/kg, and 0.421±0.302, respectively. For the case of coral sample, the values of the corresponding radiological indices were 20.183±7.36, 24.219±8.832 nGy.h^{-1},0.117±0.014, 0.152±0.017, 24.752±9.02 µSvy^{-1}, 43.29±5.466 Bq/kg, and 0.309±0.041 respectively. The obtained results of this study show that most of the sample's values are lower and the remaining are higher than the World average values. The mean representative index value is less than unity which confirms that the St. Martin area is safe for the inhabitants and the tourists. The results will be used as a baseline data for further researchers.

Keywords: Activity concentration, hazard parameters, effective dose, absorbed dose.

1 Introduction

Saint Martin's Island is the only coral island in Bangladesh. It is situated in the north-eastern part of the Bay of Bengal at a distance of about 9 km from the Cox's Bazar-Teknaf peninsula tip of Bangladesh. Its size is about 8 km^2 [(1)]. The population is about 3,700 and most of them are fishermen [(2)]. It is an important tourist place in Bangladesh. Every day hundreds of people visit this island.

The present research was aimed to carry out the radioactivity levels associated with sediment and other's Biota samples collected from the Saint Martin's Island to assess the radiological hazards to the tourists as well as the population nearby the Island. The concentrations of radionuclide of the samples under study were done by using a high resolution gamma spectrometry of High Purity Germanium detector (HPGe) at the Atomic Energy Centre, Chittagong, Bangladesh Atomic Energy Commission, Chittagong, Bangladesh.

Since the people of the country are not conscious about the radiation contamination and its effect on human body. In consideration of all perspectives, it is needed to find the distribution of various radionuclides present in different environmental samples. It is also necessary to explore and protect the different factors that influence the uptake of these radionuclides from environment to human food chain.

A previous work was carried out only on soil samples [(3)] of the same area. In the present study sediments, fishes and coral samples were collected for measuring the naturally occurring radioactivity and radiological hazard parameters from St. Martin's Island. As St. Martin's is a tourist place where tourists are gathered from home and abroad, it is necessary to check whether the area is safe or not for the tourists as well as for the inhabitants who are living nearby. In addition to this, it is important to create a public awareness about the radiation hazards to the studied place.

The present work was also initiated to assess the radiological hazards for St. Martin's Island, with the ultimate aim of establishing a baseline data for the concentrations of radionuclides in the island environment.

2 Materials and Methods

2.1 Study Area

For determining the radioactivity levels of naturally occurring and anthropogenic radionuclides and for finding the radiological dose rates in the sediment and other's Biota of the St. Martin's Island, a total number of 7 samples were collected from the place. Among 7 samples 4 were Sediment samples, 2 were fish samples, 1 was coral sample. The geographical location of all sediment's sampling points was recorded by using GPS of model **GPS Map 76CSx, GARMIN,** and other samples were taken randomly. A map of Bangladesh, location of St. Martin Island and the sampling locations in the island is shown in Figure 1.

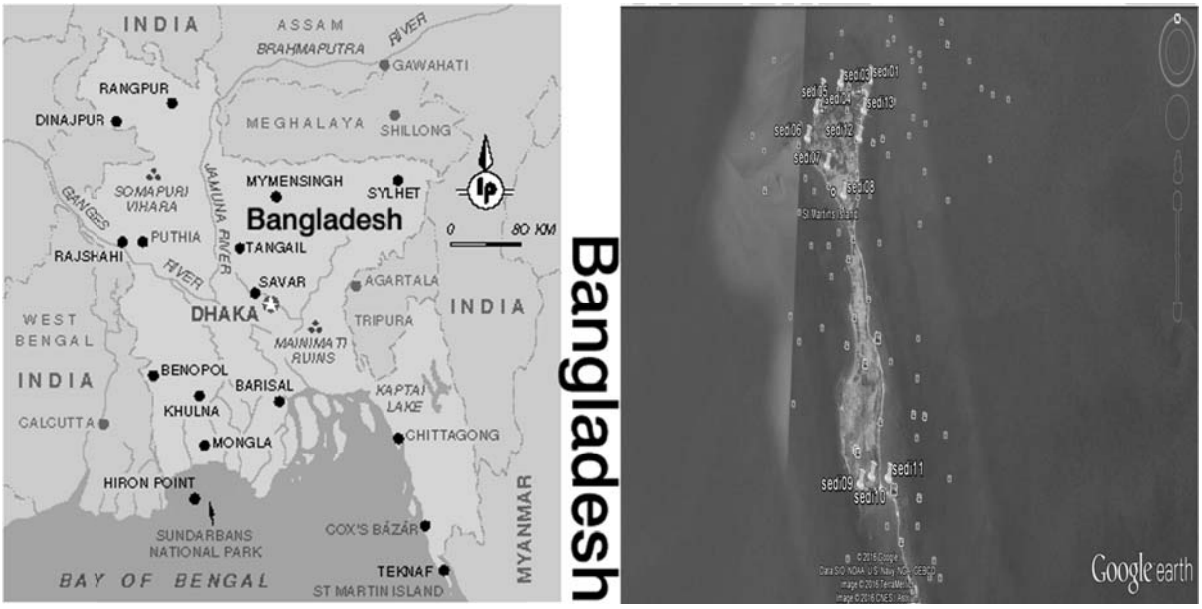


Figure 1: A map of Bangladesh with the location of St. Martin and the sampling locations for the same area.

2.2 Sample Collection

All the samples were collected using a cylindrical plastic container within a day on 17.10.15 in order to avoid the atmospheric variability and the outdoor radiation exposure rates of the individual sampling points were recorded by using a $\beta-\gamma$ survey meter (**Model-3 survey meter # 80162, LUDLUM 44-9**). Each of the collected samples was stored into a sealed plastic bag individually and marked with sample identification number.

2.3 Sample Preparation

Each of the samples was kept under the direct sunlight for several days to evaporate the water contents. Then the samples were dried in an electric oven at temperature of 105°C available at BAEC,Chittagong, Bangladesh.For making small grain size each of the samples was ground with agitate mortar and pestle and sieved by using 0.395mm diameter mesh size. The samples weight was taken by an electric digital balance and the range of samples weight were between (156-160gm) for sediment, for fishes(16-22gm) and for coral 156gm. The final weighted samples were kept for 4 weeks in order to be equilibrium. The size of the studied sample container was of the same size as the reference sample container.

2.4 Standard Gamma Sources

To measure the counting efficiency of the detector,reference sources that have similar chemical composition, concentration, geometry and counting configuration as the studied samples. In the present study, International Atomic Energy Agency (IAEA) recommended reference samples were used. The reference samples are (1) IAEA/RGU-1: Uranium ore in silica powder containing radionuclides or components U, Th, K; (2) IAEA/RGTh-1: Thorium ore in silicapowder containing radionuclides or components Th, U, K; (3) IAEA/RGK-1: Extra pure Potassium sulphate containing radionuclides or components U, Th, K.. The cap of the container was tightly sealed with plastic tap to ensure its air tightness.

2.5 Data Acquisition and Analysis of Gamma Spectra by Using Genie – 2000

In the present research work,a **HPGe Detector of CANBERRA**(Model No. BE3820, Serial No. 09078305, Active area 3800mm^2, Thickness 20mm, Relative Efficiency of 38%,Resolution of 1.9 keV(FWHM) for the peak of 1332 keV of ^{60}Co)was used to carry out the whole measurements. Each of the reference sources was placed on the top of the detector within the shielding arrangement taking a counting time for 20,000 seconds. The most prominent gamma ray energy peaks were of 238.63 keV (due to ^{212}Pb); 727.17 keV (due to ^{212}Bi); 241.98 keV, 295.21 keV& 351.92 keV (due to ^{214}Pb); 338.40 keV, 911.07 & 969.11 keV (due to ^{228}Ac); 510.57 keV, 583.19 keV& 2614.53 keV (due to ^{208}Tl); 609.31 keV, 1120.29 keV& 1764.49 keV (due to ^{214}Bi); 1460.75 keV (due to ^{40}K) and 661.66 keV (due to ^{137}Cs).The gamma ray emitting radionuclides were identified by γ-ray spectral analysis. The peak area of each γ-energy point was determined by using software genie-2000.

2.6 Sample Spectrum

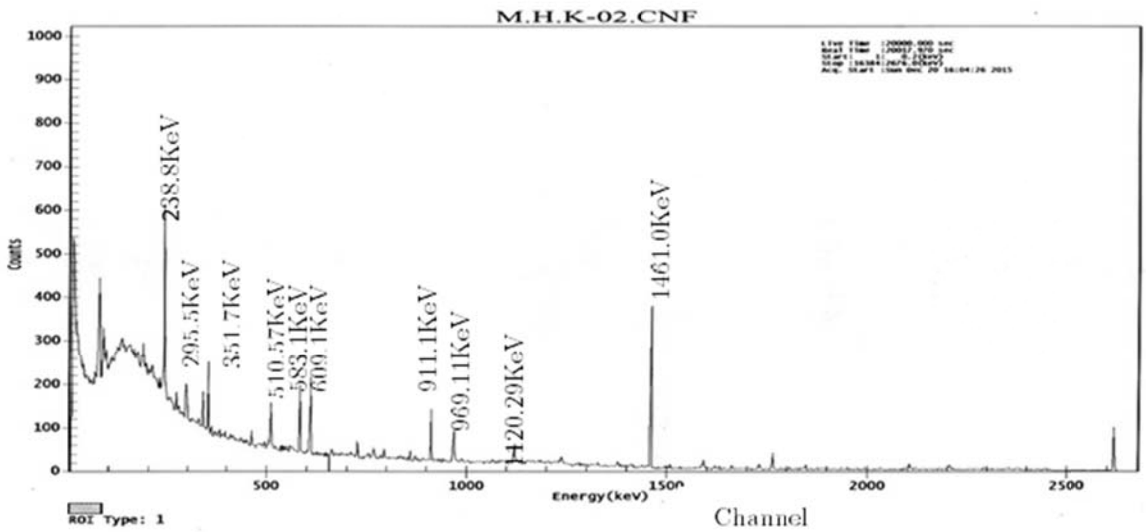


Figure 2: Counting gamma spectrum of the sample MHK-02 (Sedi-02)

2.7 Efficiency Calibration Curve

The counting efficiency of the (HPGe) detector was calculated by using the following formula:

$$\% \text{ Efficiency} = \frac{CPS \times 100}{Activity \times Intensity}$$

$$\text{or}, \varepsilon_f(\%) = \frac{CPS \times 100}{A_C \times I_\gamma}$$

where, CPS = Net count per second (i.e, Gross Counts– Background Counts)
Activity, A_C = Standard source activity for the respective energy peak.
Intensity, I_γ = Intensities of gamma energies.

Thus, from the measured count rates and known activities, the counting efficiencies at various gamma energy points for the corresponding source were calculated by the above equation.

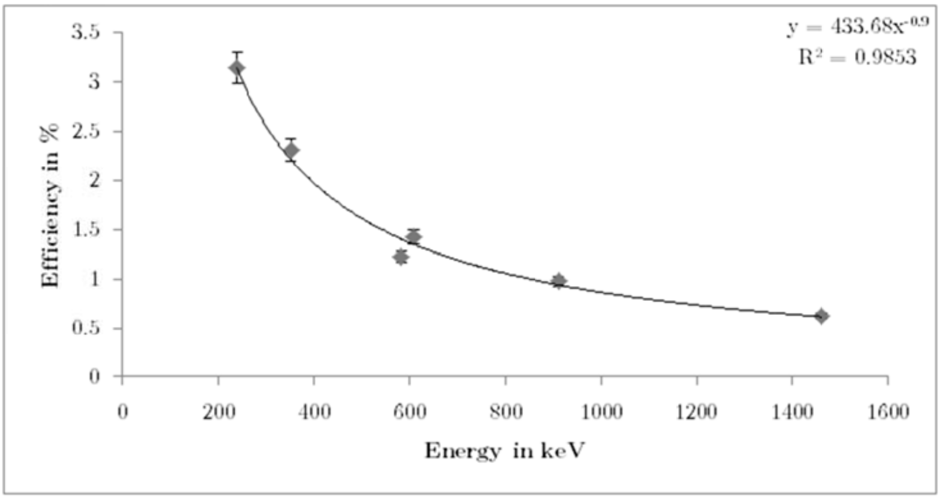


Figure 3: Counting efficiency curve of the (HPGe) Detector (For standard sample)

2.8 Measurement of Radionuclides and Activity Concentration Calculation

The samples after pretreatment, preparation and packing in the air tight sealed containers were stored for 4 weeks to reach secular equilibrium between the ^{238}U and ^{232}Th series and their respective progeny before their measurements [4,5,6]. It is assumed that ^{222}Rn and ^{220}Rn could not escape from the sealed containers from thin closure. The gamma ray activities of the Sediment and other's Biota samples were determined by the same calibrated detector coupled with Digital Spectrum Analyzer-1000 (DSA-1000). The most prominent gamma ray energies of ^{212}Pb (238.63 keV), ^{208}Tl (583.19 keV) and ^{228}Ac (911.07 keV) and energies of ^{214}Pb (351.92 keV) &^{214}Bi (609.31 keV) were used to determine the activity concentration of ^{232}Th and ^{238}U respectively. The ^{40}K and ^{137}Cs radionuclides were measured from their respective γ-ray energies 1460.75 keV and 661.66 keV, respectively [7,8,9,10].

Before calculating the net counts, region of interest (ROI) was taken for every sample from the spectrum. In data analysis, the net count of the sample was brought about by subtracting a linear background distribution of the pulse height spectra from the corresponding peak energy area. Activities of the natural radionuclides presented in the sediment and other's biota samples were calculated by using the following formula [9]:

$$\text{Activity} = \frac{CPS \times 100 \times 1000}{\varepsilon_f(\%) . \times I_\gamma \times w_s(gm)}$$

where, CPS = Net counts per second (i.e., CPS for sample – CPS for background)

ε_f = Counting gamma energy efficiency of the detector.

I_γ = Intensity of the gamma ray.

W_s = Weight of the sample.

The error of the measurements was expressed in terms of standard deviation of 1σ level.

2.9 Calculation of Radiological Hazard Parameters

The outdoor absorbed dose rate in air at 1 m above the ground surface (in $nGy.h^{-1}$) using the conversion factors given in the UNSCEAR 2000 report[(12)] is

$$D_{outdoor} = \left(0.427C_{Ra} + 0.66C_{Th} + 0.0432C_{K}\right) \quad (1)$$

where, C_{Ra}, C_{Th} and C_K are average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K respectively in sediment samples.

The indoor absorbed dose rate is 1.2 times higher than the outdoor dose given by(13)

$$D_{indoor} = D_{outdoor} \times 1.2\left(nGyh^{-1}\right) \quad (2)$$

The annual effective dose equivalent Deff from outdoor terrestrial gamma radiation is (14)

$$D_{eff} = D_{outdoor}\left(nGyh^{-1}\right) \times 0.7\left(Sv.Gy^{-1}\right) \times 8,760\left(hy^{-1}\right) \times 0.2 \quad (3)$$

where 0.2 is the outdoor occupancy factor and 0.7 $Sv.Gy^{-1}$ is the quotient of effective dose equivalent rate to absorbed dose rate in air.

For indoor radiation exposure, the annual effective dose equivalent was calculated by using an occupancy factor of 0.8 (14) as:

$$D_{eff} = D_{indoor}\left(nGyh^{-1}\right) \times 0.7\left(Sv.Gy^{-1}\right) \times 8,760\left(hy^{-1}\right) \times 0.8 \quad (4)$$

The total annual effective dose equivalent from terrestrial radiation is the sum of outdoor and indoor annual effective dose equivalent.

The external radiation hazard, Hext and internal radiation hazard, Hint are calculated as follows:

$$H_{ext.} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \quad (5)$$

$$H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \quad (6)$$

where the numerical quantities of equations (5) and (6) are in units of $Bq.kg^{-1}$ and A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K respectively.

The formulas for calculating the radium equivalent activity (15), Raeq and the representative level index (14,15, 16), I γ r in the present research are as follows:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k \quad (7)$$

$$I_{\gamma r} = \left(\frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_k}{1500}\right) \quad (8)$$

where, A_{Ra}, A_{Th} and A_K in equation (7) and C_{Ra}, C_{Th} and C_K in equation (8) are activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively in sediment samples.

3 Results and Discussion

The concentrations for the corresponding gamma emitting radionuclides of all samples, collected from the St. Martin's island, Chittagong, Bangladesh were measured by using the High Purity Germanium detector. In the present study, the activity of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs of all samples were measured within counting error of 1σ level. The activity concentrations for three different types of samples are shown below:

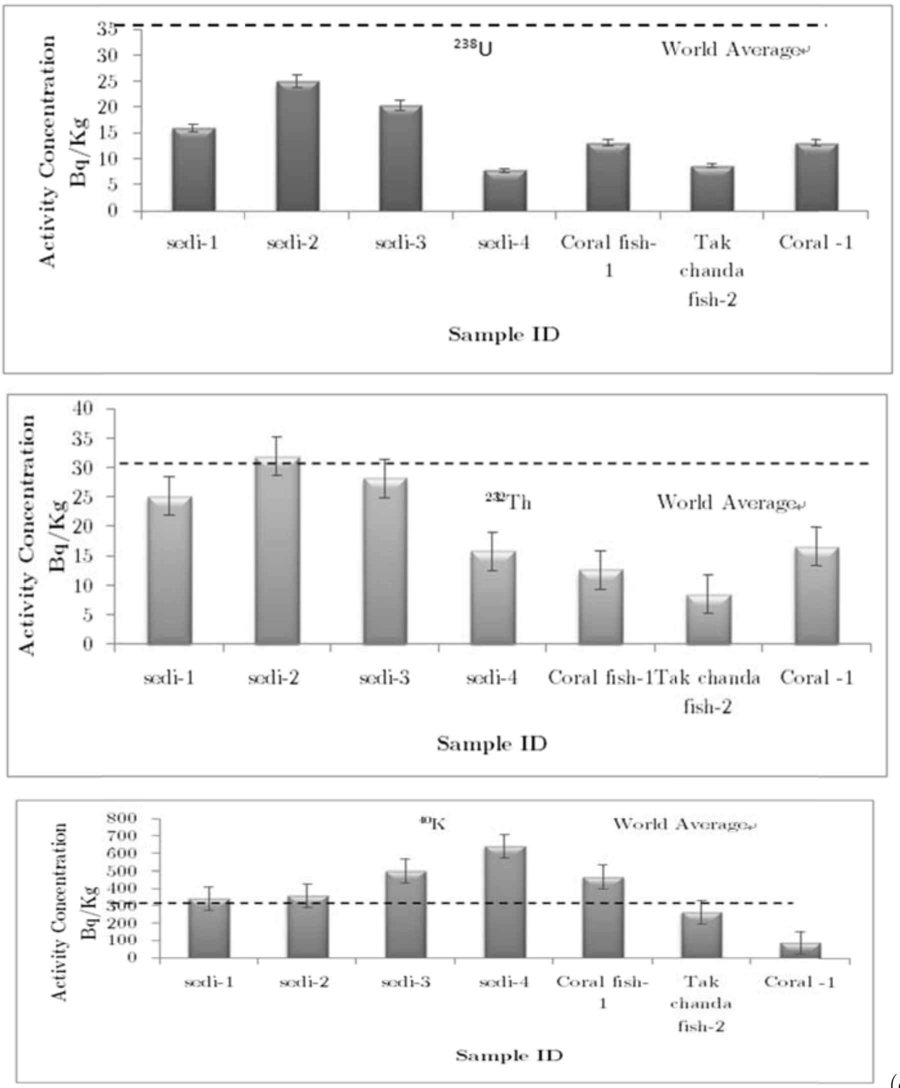
3.1 Activity Concentrations and Radiological Hazard Parameters of all Samples

1. The activity concentrations of radionuclides of all samples have been shown in table 1 and the graphical representation for the radionuclides have been shown in Figure 4.
2. The radiological hazard parameters of all samples have been shown in Table 2.

Table 1. The activity concentrations of the radionuclides, ^{238}U, ^{232}Th, ^{40}K and of all samples.

Sample Type	Sample id	^{238}U	^{232}Th	^{40}K
	sedi-1	15.93±2.1	25.09±2.72	341.77±47.2
Sediment	sedi-2	25±2.17	31.85±2.77	358.32±47.1
	sedi-3	20.29±2.25	28.12±2.89	498.34±53.3
	sedi-4	7.75±2.04	15.77+0.83	640.84±56.23
	Mean	17.243±2.14	25.207±2.302	460.802±50.957
	Coral f-1	13.09±14.04	12.64±17.44	464.74±305.61
Fish	Tak chandaf-2	8.63±19.19	8.47±9.58	265.1±417.08
	Mean	**10.86±16.615**	10.555±13.51	364.92±361.345
Coral	Coral -1	**13.06±2.1**	16.57±2.65	84.96±43.29

3.2 Activity Concentrations of 238U, 232Th and 40K for All Samples with the World Values



(a) (b) (c)

Figure 4: Comparison of the activity concentrations for all samples- (a) for ^{238}U (b) for ^{232}Th (c) for ^{40}K .

3.3 The Activity Concentration of 238U, 232Th and 40K for All Samples

Activity of ^{238}U: The activity concentrations of ^{238}U of the sediment samples have been found in the range of 7.754±2.044 to 25.005±2.176 Bq/kg with an average value of 17.243±2.14 Bq/kg. The values of fish have been found in the range 8.639±19.19 to 13.094±14.036, an average value of 10.86±16.615 Bq/kg. The value of coral is 13.06±2.1; whereas the world average value is 35 Bq/kg. So, no activity concentration value is found above the world average value except coral.

Activity **of ^{232}Th**: The activity concentrations of **^{232}Th** of the sediment samples have been in the range of 15.769±0.827 to 31.852±2.773 Bq/kg with an average of 25.207±2.302 Bq/kg. The values of fish have been found in the range of 8.471±9.58 to 12.641±17.44 Bq/kg; an average value of is 10.555±13.515 Bq/kg. The value of coral is 16.57±2.65. Whereas the world average value is 30 Bq/kg. The average activity concentration value is found below the world average value.

Activity **of ^{40}K:** The activity concentrations of **^{40}K** of the sediment samples have been in the range of341.769±47.2006 to 640.841±56.237 Bq/kg with an average of 460.802±50.957 Bq/kg. The values of fish have been found in the range 265.1±417.08 to 464.742±305.61 Bq/kg with an average of 364.92±361.345Bq/kg. The value of coral is 84.96±43.29; whereas the world average value is 400 Bq/kg which shows that the average sediment value is higher than the world average value. For the case of fish samples the observed peak was distorted due to low sample weight and the short counting time.

3.4 Calculation of Radiation Hazard Parameters:

The radiological hazard parameters for all samples are shown in Table (2)

Table 2. The Comparison of the radiological hazard parameters among all type of samples.

Sample ID	Outdoor absorbed Dose rate in nGy.h^{-1}	Indoor absorbed dose rate in nGy.h^{-1}	The external Radiation hazard, H_{ext}	The internal radiation hazard, H_{int}	The annual Effective dose equivalent, D_{eff} (μSvy^{-1})	The representative level index, Iγr	The radium activity, Ra_{eq}, (Bq.Kg^{-1})
Sedi-01	38.124±2.839	45.748±3.406	0.210+0.015	0.254±0.018	46.755±3.481	0.584±0.043	78.124±5.72
Sedi-02	47.175±2.890	56.61±3.468	0.265+0.016	0.332±0.018	57.855±3.54	0.724±0.044	98.136±5.79
Sedi-03	48.751±3.144	58.5012±3.772	0.267+0.016	0.321±0.019	59.788±3.85	0.748±0.048	98.873±6.24
Sedi-04	41.401±2.638	49.681±3.165	0.215+0.0132	0.236±0.016	50.774±3.235	0.636±0.040	79.645±4.93
Mean	43.863±2.877	52.636±3.452	0.239+0.015	0.286±0.018	53.793±3.526	0.673±0.043	88.696±5.672
Coral fish-01	34.008±18.512	40.809±22.214	0.180+0.099	0.216±0.119	41.707±22.703	0.523±0.284	66.950±37.05
TakChanda fish-02	20.727±20.7	24.872±24.84	0.112+0.107	0.134±0.140	25.419±25.39	0.318±.320	41.154±39.84
Mean	27.368±19.606	32.841±23.527	0.146+0.103	0.175±0.129	33.563±24.046	0.421±0.302	54.052±38.446
Coral-01	20.183±7.360	24.219±8.832	0.117+0.0147	0.152±0.017	24.752±9.02	0.309±0.041	43.297±5.47
World Average	60	72	1	1	80	0.66	370

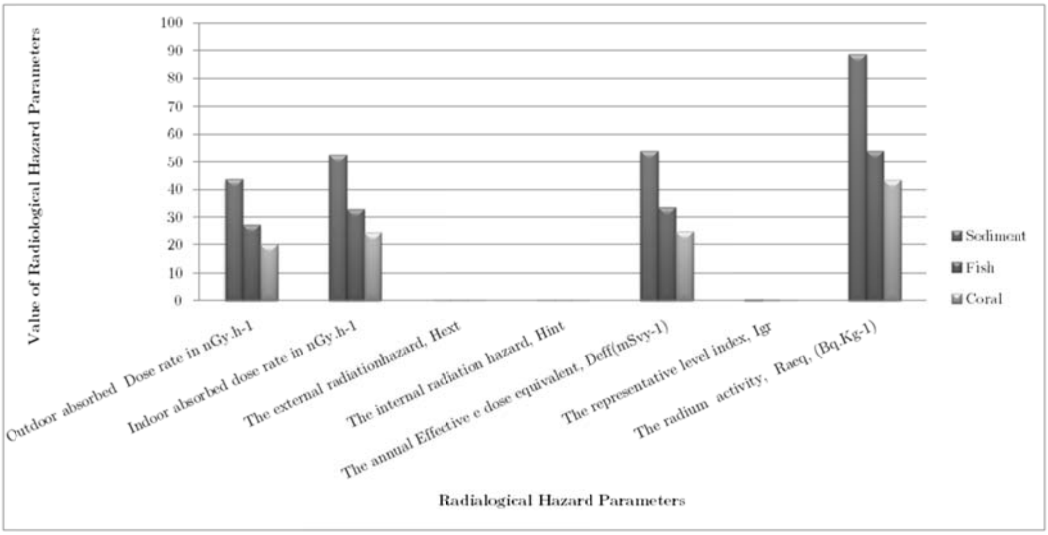


Figure 5: The Comparison of the radiological hazard parameters among all type of samples.

3.5 The Radiological Hazard Parameters for All Samples

A comparison of radiological hazard parameters for all samples has been shown in Table 2 and the graphical representations in Figure 5, respectively.

3.5.1 The Outdoor Absorbed Dose Rate:

The outdoor absorbed dose rate in air at 1m above from the ground surface in the sediment samples have been found in the range of 38.124±2.839 nGy.h^{-1} to 48.751±3.144 nGy.h^{-1} with the mean value of 43.863±2.877 nGy.h^{-1}. The values of fish have been found between 20.727±20.7 and 34.008±18.512; an average value of 27.368±19.606 nGy.h^{-1}. The value of coral is 20.183±7.360; whereas the world average value is 60 nGy.h^{-1}. So, no outdoor absorbed dose rate's value is found above the world average.

3.5.2 The Indoor Absorbed Dose Rate:

The indoor absorbed dose rate in air for the sediment samples have been found in the range of 45.748±3.4068 nGy.h-1 to 58.5012±3.772 nGy.h-1 with the mean value of 52.636±3.452 nGy.h-1. The values of fish have been found in the range 24.872±24.84 to 40.809±22.214 with an average value of 32.84±23.527 nGy.h-1. The value of coral is 24.219±8.832; and the world average value is 72 nGy.h-1(17). So, no indoor absorbed dose rate was found above the world average.

3.5.3 The External Radiation Hazard,(Hext):

The external radiation hazard values for the sediment samples have been found in the range of 0.210±0.015 to 0.267±0.016 with the mean value 0.239±0.015. The values of fish have been found in the range 0.112±0.107 to 0.180±0.099 with mean value of 0.146±0.103. The value of coral is 0.117±0.0147; whereas the world average value is 1(19).So the external radiation hazard's value is less than the world average. Therefore, the St. Martin Island is safe for the tourists and the inhabitants nearby.

3.5.4 The Internal Radiation Hazard,(Hint):

The internal radiation hazard for the sediment samples have been found in the range of 0.236±0.016 to 0.332±0.018 with the mean value being 0.286±0.018. The values of fish have been found in the range from 0.134±0.140 to 0.216±0.119; an average value of 0.175±0.129. The value of coral is 0.152±0.017; whereas the world standard value is 1(19).So The internal radiation hazard value is less than the world average.

3.5.5 The Annual Effective Dose Equivalent (Deff):

The annual effective dose equivalent for the sediment samples has been found in the range of 46.755±3.481 to 59.788±3.85 with the mean value being53.793±3.526. The values of fish have been found in the range of 25.419±25.39 to 41.707±22.703 with an average value of 33.563±24.046. The value of coral is 24.752±9.02; whereas the world average value is 80µSvy-1[(19)]. So, the annual effective dose equivalent value is less than the world average value.

3.5.6 The Representative Level Index (I γ r):

The representative level index for the sediment samples has been found in the range of 0.584±0.043 to 0.748±0.048 with the mean value being0.673±0.0437. The values of fish have been found in the range 0.318±0.320 to 0.523±0.284; an average value of 0.421±0.302. The value of coral is 0.309±0.041; whereas the world standard value is 0.66(19). So, the representative level index value for sediments is within the world average value.

3.5.7 The Radium Equivalent Activity,(Raeq):

The radium equivalent activity for the sediment samples has been found in the range of 78.124±5.72 Bq/kg to 98.873±6.24 Bq/kg with the mean value being 88.695±5.672 Bq/kg. The values of fish have been found in the range 41.154±39.84 Bq/kg to 66.950±37.05 Bq/kg. The average value of fish is 54.052±38.446 Bq/kg. The value of coral is 43.29±5.466 Bq/kg; whereas the world standard value is 370 Bq/kg (19). So, the radium equivalent activity value is less than the world average. In this graph, the values of the external radiation hazard,(Hext), the internal radiation hazard,(Hint) & the representative level index (I γ r) were very small and that is why it was not shown very clearly in the representation.

4 Conclusion

The detection of radionuclides, their activity concentrations and radiological hazard parameters of three different kinds of total seven (four sediment, two fish and one coral) samples collected from St. Martin's Island. Chittagong, Bangladesh were determined as a part of assessment of the radiological impact of St. Martin's island on the inhabitants and the tourists. The detection of natural radionuclides of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs and their activity concentrations were determined by using a calibrated High purity Germanium Detector (HPGe). There is no artificial radionuclide found in this study. The activity concentrations and radiological hazard parameters for all samples were determined individually. The activity concentration values for most of the samples were lower and for a few were higher than the world average values. The radiological hazard parameters of these samples were found within the acceptable limit set by the ICRP report which shows that there is no radiation hazard risk for the inhabitants and the tourists. Therefore, we can say that this island is safe for both inhabitants and tourists.

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