RADIOLOGICAL ASSESSMENT OF THE SURFACE SOIL OF BANGLADESH

by

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The specific activities of ^{232}Th, ^{238}U, ^{40}K, and ^{137}Cs in undisturbed surface soil at 56 locations of Bangladesh, covering its entire area, were measured using high resolution HPGe detector. The mean specific activity concentrations of the mentioned radionuclides were respectively found to be 83.56 17.96 Bq/kg, 44.35 12.65 Bq/kg, 630.89 173.85 Bq/kg, and 5.37 4.87 Bq/kg. A good correlation between the activities of ^{232}Th and ^{238}U was found. The radiological parameters namely radium equivalent activity, representative level index, external hazard index, internal hazard index, absorbed dose rate, and effective annual outdoor dose rate due to the natural radionuclides were also calculated. The average values of the mentioned parameters were found to be 212.26 43.93 Bq/kg, 1.55 0.32 Bq/kg, 0.29 0.06, 0.69 0.15, 97.27 20.03 nGy/h, and 119.37 24.58 μSv/y, respectively. The radiation dose levels at the points of sample collection were also measured by a portable radiation dose rate-meter. The average value of the outdoor dose rate was found to be 0.20 0.07 μSv/h ranging from 0.16 0.02 μSv/h to 0.28 0.04 μSv/h. The distributions of natural radionuclides were found to be normal. The concentration levels of different radionuclides were comparable to the corresponding reported values of the soil of different countries.

Key words: surface soil, Bangladesh, gamma spectrometry, radiation hazard index, dose rate

INTRODUCTION

The main sources of natural background radiation are the naturally occurring radionuclides in the ground (^{232}Th and ^{238}U and their decay products; ^{40}K *etc.*). High natural radiation levels have been found in some of the countries of the world like Brazil, China, and India [1, 2]. Moreover, enhanced radioactivity due to natural oil and gas production was also observed in a North German oil field [3]. The average annual effective dose to adults from natural sources of ionizing radiation is 2.4 mSv [4]. In addition to this, people are also exposed to artificial sources of radiation. Artificial radioactive isotopes (such as ^{137}Cs, ^{90}Sr, ^{239}Pu, *etc.*) from fallout, operation of nuclear facilities, *etc.* if introduced into the environment, will eventually reach humans *via* the food chain. Amongst the artificial sources, ^{137}Cs is considered one of the most hazardous radionuclides. The Chernobyl accident on April 26, 1986, in Ukraine became an unprecedented source of radionuclides input to the environment, which released about 3.7 10^7 GBq of ^{137}Cs to the environment [5], the total amount of discharged radionuclides was ~3.6 EBq (100 MCi) [6].

Bangladesh is a riparian country located at 88°01'E – 92°41'E longitude and at latitude 20°34'N – 26°38'N having comparatively high population density. It also has natural gas and oil. It is geographically quite downward to India. Most of the rivers of Bangladesh originate from the hill tracts of India, Nepal, and Bhutan; and flow through the land of India. Therefore its land might be contaminated by radioactive sources from the upstream and from sea and river water (through cyclone and flood).

As an aftermath of Chernobyl accident, the radioactivity levels in soil have been measured in different countries of the world [7]. In Bangladesh, no systematic data are available in this regard. It is therefore necessary to know the present level of radioactivity in soil samples of Bangladesh. In this context, a program has been undertaken to measure the radioactivity levels due to fallout and natural origin in soil samples of different locations of Bangladesh.

MATERIALS AND METHODS

Study area, sampling and preparation

In order to assess the concentration levels of the radionuclides ^{232}Th, ^{238}U, ^{40}K, and ^{137}Cs in the soil of Bangladesh, 56 location as shown in fig. 1, were selected all over Bangladesh by considering the population density, area, and the communication system(s)

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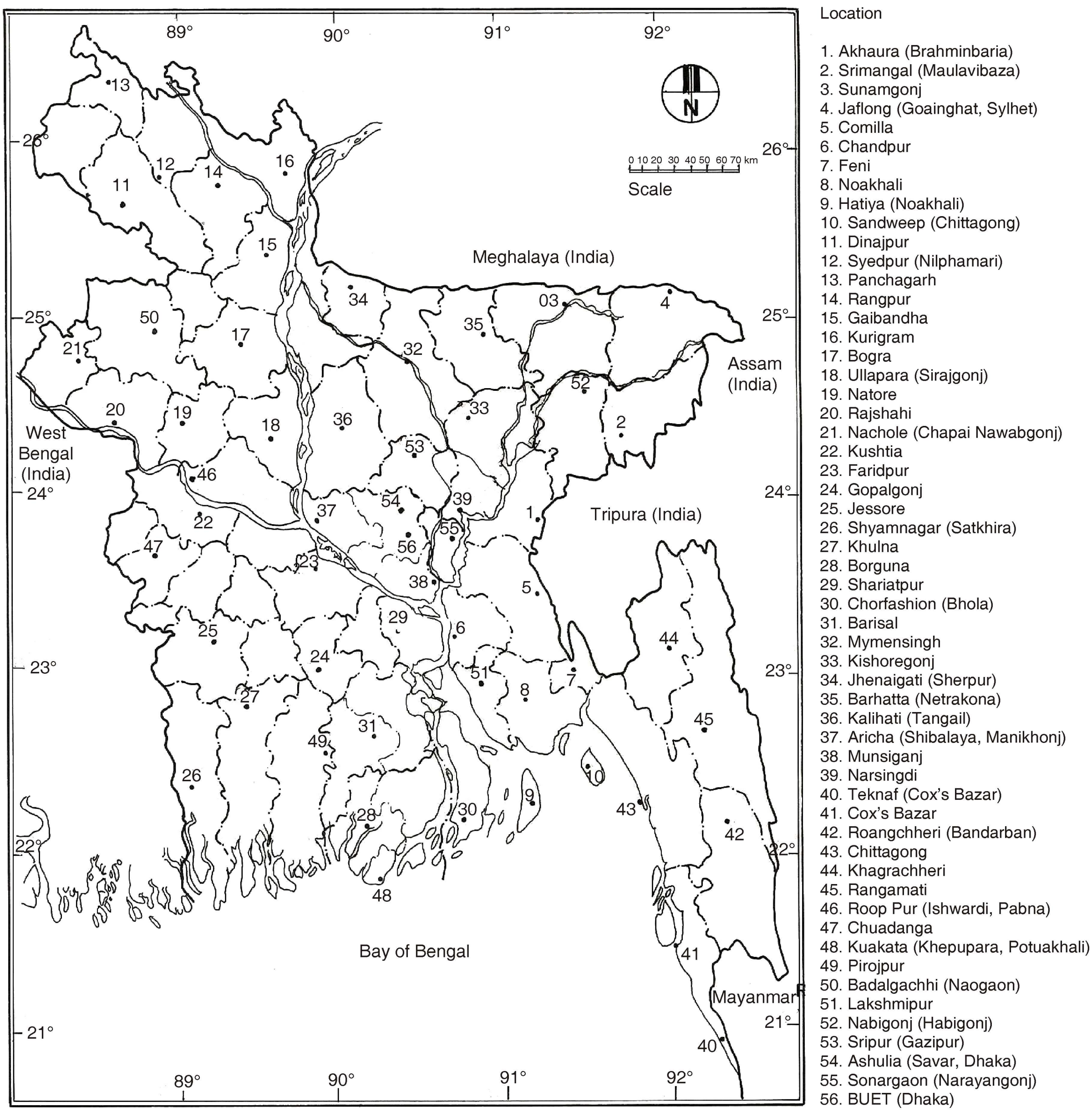

Figure 1. Map of Bangladesh showing different locations of sample collection

for sample collection. These 56 locations covered the entire geographical area of Bangladesh. From each of the locations, undisturbed soil samples were collected for measuring the radioactivity. At each of the pre-selected 56 sampling stations, approximately 1.5 kg of soil was taken from a 20 cm 20 cm wide and approximately 3 cm deep surface area of undisturbed land (the land which was kept in natural condition *i. e.*, the land which has never been ploughed, cultivated, dug, fertilized, or landfilled during the last 20 years period, ignoring the natural flow/drainage of rain water and normal land erosion; so that it may be considered as the representative land of the sampling station). The soil samples were packaged tightly in polythene bags in such a way that no fraction of the collected soil could normally escape the polythene bag. Individual identification marks were given to each of the soil sample packets with marker pens. The outdoor radiation dose levels at the points of sample collection and nearby places were measured several times at each point by a portable radiation dose rate-meter "PDR 1Sv" and the means were calculated for each sampling point.

The soil samples were crushed into powder individually and then sieved by a 1.0 mm sieve. All of the samples were in dry condition as these were collected during the dry season. Then 1 kg of pure soil from each of the crushed and sieved sample was measured individually by a sensitive balance. The powdered 1kg samples were then poured into Marinelli beakers carefully and sealed air-tightly. The sealed soil samples were allowed to attain the radioactive secular equilibrium between ^{226}Ra and its progenies by preserving them in an airtight condition individually for 28 days [8].

Gamma-spectrometry and analytical procedure

The radioactivity was measured by the aid of a high purity germanium detector interfaced with necessary electronic accessories. The high purity germanium "closed-end-coaxial p-type dipstick" radiation detector "Silena" was employed in the present study for measurement of γ-activity in soil samples collected from different locations in Bangladesh. The efficiencies of the detector employed at different energies were determined by employing standard procedure [9]. The most gamma energy peaks at 238.63 keV, 351.92 keV, 583.19 keV, 609.31 keV, 911.07 keV, 1120.29 keV, and 1460.75 keV were clearly identified. For most of the samples a gamma energy peak at 661.66 keV was also identified. These energy peaks were used for the estimation of the corresponding radionuclides. The counting period for each of the sample was 20 000 seconds.

The equilibriums between ^{232}Th and its progenies and ^{238}U and its progenies had been considered and therefore the specific activities of the radionuclei ^{232}Th and ^{238}U were calculated with the corrections on the respective decay schemes. For ^{232}Th, the activities of ^{212}Pb, ^{208}Tl, and ^{228}Ac had been considered; for ^{238}U, the activities of ^{214}Pb and ^{214}Bi had been measured; however, the specific activities of ^{40}K and ^{137}Cs had been determined directly by single channel energy counts and by using the following formula [9]

$$A \quad \frac{C}{\varepsilon(E) I_{\gamma} W} \tag{1}$$

where A [Bqkg^{-1}] is the activity of the sample, C [s^{-1}] – the peak area counts, $\varepsilon(E)$ [keV] – the efficiency of the detector at energy E, I_{γ} [keV] – the photon emission probability at energy E, and W [kg] – the mass of the soil samples.

The statistical errors were considered in calculating the radioactive concentration levels of the samples by employing standard mathematical formula [10]. In the present study, 1σ of all the measurements were considered as it covers 68.27% of most probable values.

Outdoor radiation dose level

Radiation dose levels at 56 stations covering entire area of Bangladesh were measured by a routinely calibrated (at the Secondary Standard Dosimetry Laboratory, Institute of Nuclear Science and Technology, Atomic Energy Research Establishment, Bangladesh Atomic Energy Commission) low level portable radiation dose rate survey meter "PDR 1Sv" (NE Technology Limited, England, 1990). With each dose measurement, the dose level was observed for at least two minutes in the display-scale of the "PDR 1Sv" at one meter above the ground (gonad level). Several pairs of readings were taken from each of the spots and surroundings. All measurements taken included the minimum dose level, the maximum dose level, and the trend (average) dose level and they were noted down, individually. Finally, the average trend dose levels were calculated for each site.

Cumulative frequency plot

In order to determine the Geometric Mean and Geometric Standard Deviation of data, the net readings were evaluated with the help of the Cumulative Frequency Plot. The cumulative frequency plot or simply the probability plot of a series of entries in data set is a tool for determining its geometric mean (GM) and geometric standard deviation (GSD). To draw the cumulative frequency plot (probability plot) for data containing a number of entries, the entries were ranked at first by arranging them in ascending order and then the cumulative percents were calculated by using the formula [11]

$$\text{Cumulative percent} \quad \frac{100(i \quad 0.5)}{n} \tag{2}$$

where i is the serial position and n – the total number of entries. In the present study, cumulative frequency plots were drawn for finding out the geometric average concentrations and standard deviations of the radionuclides ^{232}Th, ^{238}U, ^{40}K, and, ^{137}Cs found in soil collected from different locations of Bangladesh and the consequent radiation dose levels; and for the outdoor dose levels measured instantly by a survey meter.

Frequency distribution of the concentrations of radionuclides

Frequency distribution of the concentrations of the radionuclides ^{232}Th, ^{238}U, ^{40}K, and ^{137}Cs were also done. The distributions were plotted by classes of the concentrations of a radionuclide as abscissa and the corresponding frequencies (in percent) as ordinate [12].

Radiation hazard indices

Radiation hazards due to the concentrations of natural radioisotopes (^{232}Th, ^{238}U, and ^{40}K) found in soil samples were calculated. Six indices were used in this regard, namely: radium equivalent activity (Ra_{eq}), representative level index (I_r), external hazard index (H_{ex}), internal hazard index (H_{in}), absorbed dose rate

(D), and the annual effective outdoor radiation dose rate (E). These indices simply denote the levels of hazard due to natural radioactive nuclei and help to compare the hazard levels of different samples.

Radium equivalent activity: Ra_{eq} of the activities of the natural radionuclides in individual samples was calculated by using standard formula [13]

$$Ra_{\mathrm{eq}} \quad C_{\mathrm{Ra}} \quad \frac{10}{7} C_{\mathrm{Th}} \quad \frac{10}{130} C_{\mathrm{K}} \tag{3}$$

where C_{Ra}, C_{Th}, and C_K [Bqkg^{-1}] are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Since ^{226}Ra and ^{238}U were in secular equilibrium during reading out period, the activity of ^{238}U was assumed to be equal to the activity of ^{226}Ra in the present study.

Representative level index: $I_{\gamma r}$ of the natural radionuclides in individual samples was calculated by employing the standard formula [14-16]

$$I_{\gamma r} \quad \frac{1}{150} C_{\mathrm{Ra}} \quad \frac{1}{100} C_{\mathrm{Th}} \quad \frac{1}{1500} C_{\mathrm{K}} \tag{4}$$

where C_{Ra}, C_{Th}, and C_K are the same as defined earlier.

External hazard index: External hazard indices due to the natural radionuclides were calculated by employing the following formula [17]

$$H_{\mathrm{ex}} \quad \frac{C_{\mathrm{Ra}}}{740} \quad \frac{C_{\mathrm{Th}}}{520} \quad \frac{C_{\mathrm{K}}}{9620} \tag{5}$$

where C_{Ra}, C_{Th}, and C_K [Bqkg^{-1}] are the specific activities of the radionuclides ^{226}Ra (^{238}U in the present study), ^{232}Th and ^{40}K respectively.

Internal hazard index: Internal hazard indices due to the mentioned natural radionuclides were also calculated by using the following formula [18]

$$H_{\mathrm{in}} \quad \frac{A_{\mathrm{Ra}}}{185} \quad \frac{A_{\mathrm{Th}}}{259} \quad \frac{A_{\mathrm{K}}}{4810} \tag{6}$$

where A_{Ra}, A_{Th}, and A_K [Bqkg^{-1}] are the specific activities of the radionuclides ^{226}Ra (^{238}U in the present study), ^{232}Th, and ^{40}K, respectively.

Absorbed dose rate: The absorbed dose rates due to gamma radiations in air at 1 m above the ground surface for the naturally occurring radionuclides ^{238}U, ^{232}Th, and ^{40}K were calculated by [18]

$$D \quad 0.462 C_{\mathrm{Ra}} \quad 0.604 C_{\mathrm{Th}} \quad 0.0417 C_{\mathrm{K}} \tag{7}$$

where D [nGyh^{-1}], and C_{Ra}, C_{Th}, and C_K [Bqkg^{-1}] are the activity concentrations of ^{226}Ra (^{238}U in the present study), ^{232}Th, and ^{40}K, respectively.

Annual effective radiation dose rate: The annual effective outdoor dose rate E [µSvy^{-1}] was calculated by employing the formula [18]

E [µSvy^{-1}] = dose rate [nGyh^{-1}] × 24 hour × 365.25 day × 0.2 (occupancy factor) × 0.7 Sv/Gy (conversion coefficient) × 0.001 (8)

RESULTS AND DISCUSSION

Concentration of natural and anthropogenic radionuclides in surface soil

The measured specific activities of the radionuclides ^{232}Th, ^{238}U, ^{40}K, and ^{137}Cs in soil samples of different regions of Bangladesh are given in tab. 1 along with the other radiological parameters. Following are the short descriptions of those.

Thorium-232: The average concentration of ^{232}Th in soil samples was found to be 83.56 17.96 Bq/kg ranging from 39.27 7.74 to 128.21 7.83 Bq/kg. The lowest activity was found in the soil sample collected from Nabigonj (in Habigonj district) and the highest activity was found in the soil sample of Nachole (in Chapai Nawabgonj district) [tab. 2]. From the corresponding cumulative frequency plot, it was found that the geometric mean of ^{232}Th in soil samples is 83.50 Bq/kg which is approximately equal to the arithmetic mean value; indicating the normal distribution. The geometric means and geometric standard deviations of the concentrations of radionuclides ^{232}Th, ^{238}U, ^{40}K, and ^{137}Cs along with the arithmetic means and standard deviations are shown in tab. 2. The frequency distribution of ^{232}Th concentrations (as shown in fig. 2) also reveals that the distribution is normal.

The average level of radioactivity of ^{232}Th in the present study was found to be higher than those of the other regional studies of Bangladesh [19, 20, 22, 23] except for the one conducted in Northern Districts [21], Gudalore (India) [25], Pakistan [30-36], Sri Lanka [17], Sichuan (China) [37], Taiwan [39], Songkhla (Thailand) [40], in one study of Malaysia [42], Vietnam [44], Juban (Yemen) [45], Tafila (Jordan) [47], Saudi Arabia [48-52], Mazandaran (Iran) [53], four local studies of Turkey (Marmara, Kirklareli, Izmir, and Bursa) [56-59], Tripoli (Libya) [60], Egypt [61-64], Nigeria [66], Serbia and Montenegro [68], Ireland [69], Caceres (Spain) [70], Hungary [71], Cyprus [72], Louisiana (USA) [73] and also the world average [18] value; lower than those of Ooty (India) [24], three South Indian studies [26-28], China [37], two studies of Malaysia [41, 43], Kestanbol (Turkey) [55], and South Cameroon [67]; and similar to that of Northern India [29]; which are shown in tab. 4.

Uranium-238: Average specific activity of ^{238}U was found to be 44.35 12.65 Bq/kg with a range 17.84 6.21 to 76.06 7.58 Bq/kg. The lowest concentration of ^{238}U was found in the sample of Nabigonj while the highest concentration was found in

Table 1. Specific activity of radionuclides in soil samples collected from different locations in Bangladesh along with some consequent radiological indices and annual outdoor dose rate measured by portable radiation dose rate-meter

Map. Loc. No.	Location	Activity [$Bqkg^{-1}$] ^{232}Th		^{238}U		^{40}K		^{137}Cs		Radium equivalent [$Bqkg^{-1}$]	Represent. level index [$Bqkg^{-1}$]	External hazard index	Internal hazard index	Dose rate [$nGyh^{-1}$]	Annual effective outdoor dose [μSv]	Annual outdoor dose rate [mSv]
01	Akhaura	81.63	7.92	49.11	8.61	373.83	61.83	4.04	1.62	194.48	1.39	0.26	0.66	87.58	107.48	1.58
02	Srimangal	74.13	6.03	35.61	7.61	333.31	56.53	Below MDA		167.15	1.20	0.23	0.55	75.13	92.20	1.49
03	Sunamgonj	104.82	7.30	45.08	9.04	915.66	72.65	9.72	1.98	265.26	1.96	0.36	0.84	122.32	150.12	1.66
04	Jaflong	83.34	7.97	42.07	7.80	480.36	58.49	26.79	2.23	198.08	1.43	0.27	0.65	89.80	110.21	1.58
05	Comilla	65.94	6.46	34.99	8.24	636.23	65.42	4.67	1.59	178.13	1.32	0.24	0.58	82.52	101.28	1.84
06	Chandpur	65.24	6.45	29.73	8.27	467.29	63.26	Below MDA		158.88	1.16	0.21	0.51	72.63	89.13	1.66
07	Feni	74.00	9.27	46.20	9.45	729.69	72.97	10.56	2.03	208.04	1.53	0.28	0.69	96.47	118.39	1.50
08	Noakhali	76.49	6.80	37.48	8.69	632.79	67.77	7.61	1.79	195.43	1.44	0.26	0.63	89.90	110.33	1.31
09	Hatiya	81.11	8.65	37.36	8.44	672.18	67.22	4.98	1.62	204.94	1.51	0.28	0.65	94.28	115.71	1.66
10	Sandweep	92.26	9.56	46.23	9.38	758.84	73.49	6.03	1.80	236.40	1.74	0.32	0.76	108.73	133.43	1.58
11	Dinajpur	104.98	9.07	74.28	9.19	848.31	69.02	7.93	1.83	289.51	2.11	0.39	0.98	133.10	163.35	1.84
12	Syedpur	96.51	8.88	54.51	8.67	564.34	64.70	5.09	1.67	235.79	1.70	0.32	0.78	107.01	131.33	2.10
13	Panchagarh	110.95	9.08	73.52	9.33	815.96	67.94	3.44	1.62	294.79	2.14	0.40	1.00	135.01	165.68	1.75
14	Rangpur	123.99	8.72	64.51	8.41	869.87	64.34	12.31	1.85	308.55	2.25	0.42	1.01	140.97	173.00	2.54
15	Gaibandha	81.20	8.51	43.54	8.60	718.91	66.86	7.07	1.72	214.84	1.58	0.29	0.70	99.14	121.67	1.40
16	Kurigram	124.59	9.35	62.95	9.13	740.47	67.58	17.00	2.03	297.89	2.16	0.40	0.98	135.21	165.94	2.10
17	Bogra	101.03	8.94	55.88	8.77	664.99	66.50	5.66	1.67	251.36	1.83	0.34	0.83	114.57	140.60	1.66
18	Ullapara	93.64	8.91	61.10	8.99	812.37	67.94	16.74	2.09	257.36	1.89	0.35	0.86	118.66	145.63	1.75
19	Natore	71.48	8.41	43.47	8.30	715.31	66.50	2.76	1.51	200.61	1.48	0.27	0.66	93.09	114.24	1.58
20	Rajshahi	92.85	8.80	54.08	8.85	657.80	66.14	6.42	1.67	237.32	1.73	0.32	0.79	108.50	133.15	1.93
21	Nachole	128.21	7.83	76.06	7.58	402.59	53.56	5.97	1.33	290.19	2.06	0.39	0.99	129.37	158.76	2.19
22	Kushtia	83.47	9.39	56.23	9.65	611.07	71.17	3.81	1.70	222.48	1.62	0.30	0.75	101.88	125.03	1.84
23	Faridpur	70.83	8.31	40.54	8.50	744.06	66.86	5.58	1.64	198.96	1.47	0.27	0.65	92.54	113.57	1.75
24	Gopalgonj	76.08	8.54	45.91	8.56	553.56	64.70	Below MDA		197.17	1.44	0.27	0.66	90.25	110.75	1.84
25	Jessore	80.88	8.59	50.35	8.69	751.26	67.22	4.38	1.64	223.68	1.65	0.30	0.74	103.44	126.95	1.75
26	Shyamnagar	81.33	7.99	44.50	7.90	747.66	62.19	6.68	1.41	218.20	1.61	0.29	0.71	100.86	123.78	2.10
27	Khulna	85.69	8.62	48.92	8.67	923.79	69.02	3.49	1.54	242.40	1.80	0.33	0.79	112.88	138.53	2.01
28	Borguna	80.80	8.57	39.69	8.22	736.87	66.50	6.39	1.72	211.80	1.56	0.29	0.68	97.87	120.11	2.10
29	Shariatpur	92.36	8.83	46.12	8.63	830.34	68.30	Below MDA		241.94	1.78	0.33	0.78	111.72	137.10	2.28
30	Chorfashion	87.49	8.68	44.29	8.59	693.75	66.14	Below MDA		222.64	1.63	0.30	0.72	102.24	125.47	1.93
31	Barisal	81.50	8.62	43.87	8.63	873.47	69.02	4.36	1.62	227.49	1.69	0.31	0.73	105.92	129.99	2.01
32	Mymensingh	93.35	8.41	50.48	8.31	704.53	63.98	7.02	1.64	238.03	1.74	0.32	0.78	109.08	133.87	2.37
33	Kishoregonj	85.62	8.62	40.78	8.24	733.29	67.22	6.81	1.67	219.50	1.62	0.30	0.70	101.13	124.11	1.84
34	Jhenaigati	58.55	8.11	30.32	8.14	355.86	61.83	4.30	1.56	141.34	1.02	0.19	0.46	64.21	78.80	1.93
35	Barhatta	80.27	8.47	38.54	8.33	603.88	65.42	13.07	1.96	199.66	1.46	0.27	0.64	91.47	112.26	1.84
36	Kalihati	85.18	8.72	37.36	8.14	470.88	63.62	3.55	1.59	195.27	1.41	0.26	0.63	88.34	108.42	1.66
37	Aricha	69.79	8.34	36.50	8.39	664.99	65.78	4.04	1.64	187.35	1.38	0.25	0.61	86.75	106.46	1.75
38	Munsigonj	86.89	8.72	40.08	8.70	744.07	67.22	5.19	1.64	221.44	1.63	0.30	0.71	102.03	125.21	2.37
39	Narsingdi	79.47	8.53	37.07	8.33	657.80	65.78	8.24	1.72	201.20	1.48	0.27	0.64	92.56	113.59	2.01
40	Teknaf	105.59	9.02	45.01	8.49	524.80	64.34	8.58	1.83	236.22	1.71	0.32	0.76	106.46	130.65	1.93
41	Cox's Bazar	56.04	8.03	18.39	7.76	434.94	62.90	Below MDA		131.90	0.97	0.18	0.41	60.48	74.23	1.93
42	Roangchheri	58.28	7.73	23.71	7.49	492.45	60.39	3.34	1.43	144.85	1.07	0.20	0.46	66.69	81.85	1.84
43	Chittagong	65.07	8.32	32.00	8.29	726.10	66.86	6.26	1.70	180.81	1.35	0.24	0.58	84.36	103.54	1.84
44	Khagrachheri	68.47	8.36	30.20	8.13	727.75	62.90	Below MDA		184.00	1.37	0.25	0.58	85.66	105.12	1.58
45	Rangamati	45.28	7.86	22.23	6.72	373.83	62.54	3.86	1.59	115.67	0.85	0.16	0.37	53.21	65.30	1.66
46	Roop Pur	74.20	8.45	36.14	8.41	636.23	65.78	Below MDA		191.08	1.41	0.26	0.61	88.04	108.05	1.75
47	Chuadanga	73.44	8.42	40.16	8.30	593.10	65.06	4.15	1.56	190.70	1.40	0.26	0.62	87.64	107.56	2.01
48	Kuakata	101.33	9.01	58.75	8.71	442.13	63.26	4.04	1.64	237.52	1.70	0.32	0.80	106.78	131.05	1.84
49	Pirojpur	104.75	9.07	49.88	8.39	381.02	62.19	Below MDA		228.83	1.63	0.31	0.75	102.20	125.43	1.84
50	Badalgachhi	103.30	9.07	59.05	9.00	884.26	69.02	4.49	1.67	274.64	2.02	0.37	0.90	126.55	155.30	2.01
51	Lakshmipur	77.07	8.53	34.97	7.66	776.42	67.22	Below MDA		204.79	1.52	0.28	0.65	95.08	116.69	2.19
52	Nabigonj	39.27	7.74	17.84	6.21	276.78	61.47	4.22	1.59	95.23	0.70	0.13	0.31	43.50	53.39	1.49
53	Sripur	93.56	8.87	56.17	8.83	298.35	61.47	4.88	1.64	212.78	1.51	0.29	0.73	94.90	116.47	2.45
54	Ashulia	66.07	8.24	38.19	8.13	294.75	61.47	Below MDA		155.25	1.11	0.21	0.52	69.84	85.71	2.19
55	Sonargaon	77.98	8.59	41.82	8.46	740.47	66.86	Below MDA		210.18	1.55	0.28	0.68	97.30	119.41	2.19
56	BUET, Dhaka	81.94	8.60	39.77	8.23	514.01	64.34	5.22	1.70	196.37	1.43	0.26	0.64	89.30	109.59	1.84

Table 2. The range and average activities of radionuclides (in $Bqkg^{-1}$ unit)

Name of radionuclide	Minimum activity 1 [location and no. in map]	Maximum activity 1 [location and no. in map]	(Arithmetic) Mean activity	Geometric*	
				Mean activity	Standard deviation (1)
^{232}Th	39.27 7.74 [Nabigonj, 52]	128.21 7.83 [Nachole, 21]	83.56 17.96	83.50	1.23
^{238}U	17.84 6.21 [Nabigonj, 52]	76.06 7.58 [Nachole, 21]	44.35 12.65	44.30	1.32
^{40}K	276.78 61.47 [Nabigonj, 52]	923.79 69.02 [Khulna, 27]	630.89 173.85	632.0	1.32
^{137}Cs	2.76 1.51 [Natore, 19]**	26.79 2.23 [Jaflong, 04]	5.37 4.87	5.30	1.96

* Geometric mean and standard deviation were found from the corresponding cumulative frequency plot (probability plot). Geometric standard deviation has no unit as it is simply a ratio between the corresponding values of 50% and 84.1% cumulative frequency.

** This minimum activity is the corresponding minimum activity detected above the MDC (ignoring all minimum activities). In calculating the average values, all minimum values were assumed to be zero.

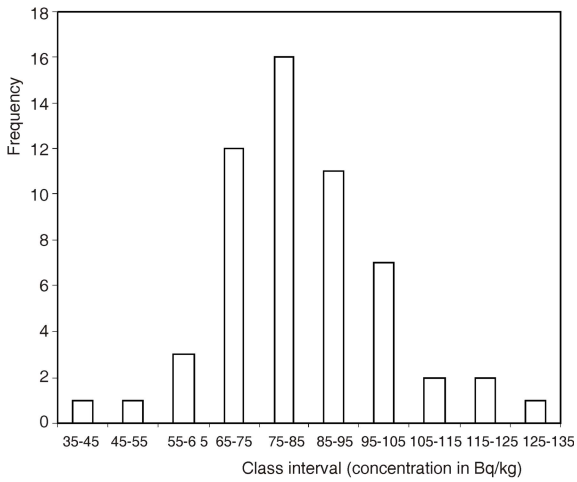

Figure 2. Frequency distribution of ^{232}Th

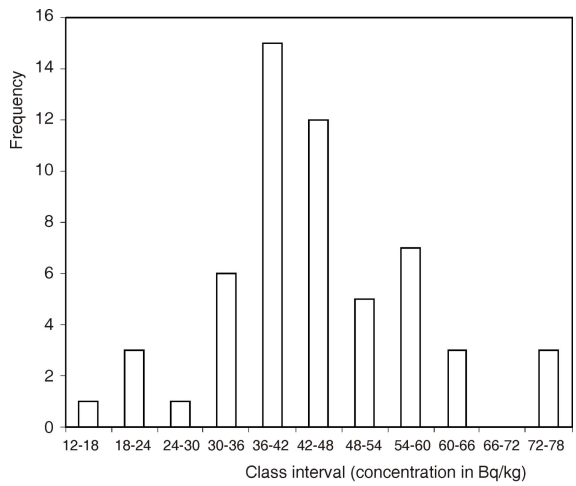

Figure 3. Frequency distribution of ^{238}U

the sample collected from Nachole [tab. 2]. The geometric mean and geometric standard deviation of ^{238}U in soil samples were determined using the cumulative frequency plot. The geometric mean was found to be 44.30 Bq/kg and the geometric standard deviation was 1.32. The close proximity of the arithmetic mean and geometric mean indicates the distribution of ^{238}U in soil samples is normal. The frequency distribution of ^{238}U concentrations in soil samples as shown in fig. 3 also indicates that the distribution is normal.

The ratio between the average concentrations of the nuclides ^{238}U and ^{232}Th was found to be 0.53 which is consistent with the UNSCEAR-2000 [18] reports.

The average level of ^{238}U in soil samples was found to be higher than those of the levels of three regional studies of Bangladesh (Dhaka, Chittagong, and Southern Districts) [19, 20, 22], five regional studies of India (Ooty, Gudalore, Kalpakkam-2, and Kanyakumari) [24-28], four local studies of Pakistan (Bahawalpur, Lahore, Punjab, and Baluchistan) [30-32, 34], Sri Lanka [17], Sichuan (China) [38], Taiwan [39], one study of Malaysia [42], Vietnam [44], Tafila (Jordan) [47], five studies of Saudi Arabia [48-52], four regional studies of Turkey (Marmara, Kirklareli, Izmir, and Bursa) [56-59], Tripoli (Libya) [60], three studies of Egypt [61, 63, 64] except one in Egyptian Sand [62], Nigeria [66], Ireland [69], Hungary [71], Cyprus [72], Louisiana (USA) [73], and the world average value [18]; lower than the corresponding values found in the studies of Northern Bangladesh [21], Jessore (Bangladesh) [23], Northern India [29], in one study of Punjab (Pakistan) [33], China [37], Songkhla (Thailand) [40], in another study of Malaysia [43], Mazandaran (Iran) [53] which was exceptionally higher, Kestanbol (Turkey) [55], South Cameroon [67], and Serbia and Montenegro [68]; and similar to those of Juban (Yemen) [45], and Caceres (Spain) [70] [tab. 4].

Potassium-40: The average concentration of radioactive potassium (^{40}K) was found to be 630.89 173.85 Bq/kg ranging between 276.78 61.47 and 923.79 69.02 Bq/kg. The highest and lowest activities of ^{40}K found in the soil samples collected from Khulna and Nabigonj, respectively, [tab. 2]. The geometric mean and geometric standard deviation of ^{40}K concentration in soil samples were found to be 632.0 Bq/kg and 1.32, respectively, from the corresponding cumulative frequency plot. The close proximity of average ^{40}K concentration in soil samples in arithmetic and geometric view indicates that the distribution is normal. The frequency distribution of the concentrations of ^{40}K found as shown in fig. 4 also manifests that the distribution is normal.

The concentration level of ^{40}K in soil samples of Bangladesh was found to be higher than those of the

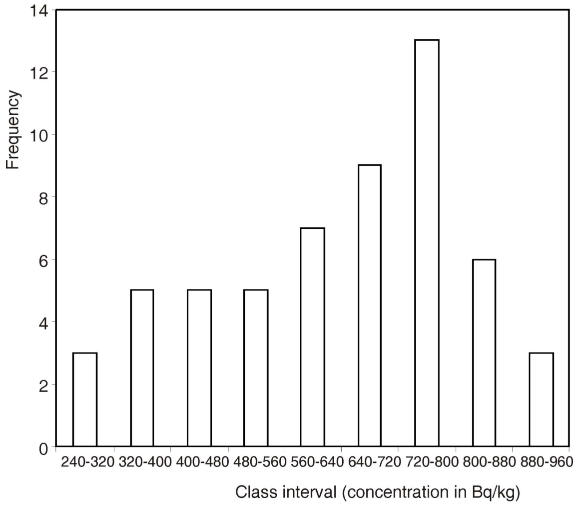

Figure 4. Frequency distribution of ^{40}K

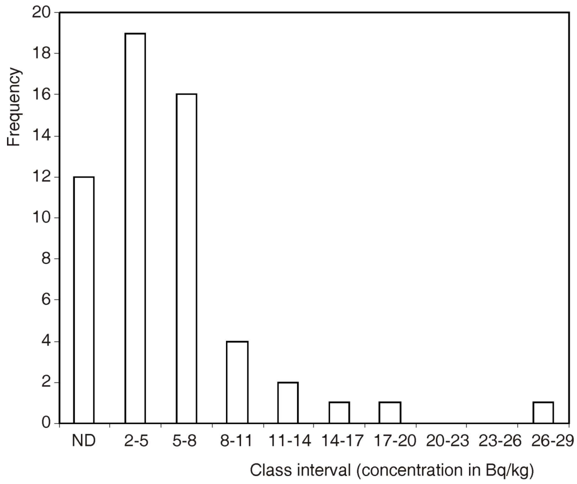

Figure 5. Frequency distribution of ^{137}Cs

levels of three other regional studies of Bangladesh (Dhaka, Chittagong, and Jessore) [19, 20, 23], five studies of India (Ooty, Gudalore, Kalpakkam-2, and Northern India) [24-27, 29], three studies of Pakistan (Lahore, Punjab and Islamabad) [31, 32, 36], Sri Lanka [17], China [37, 38], Taiwan [39], Songkhla (Thailand) [40], two studies of Malaysia [42, 43], Vietnam [44], Tafila (Jordan) [47], Saudi Arabia [48, 49, 50, 52] except that for its coastline [50], Mazandaran (Iran) [53], Marmara (Turkey) [56], Bursa (Turkey) [59], Tripoli (Libya) [60], Egyptian studies [61, 63, 64] except its sand samples [62], Nigeria [66], Serbia and Montenegro [68], Ireland [69], Hungary [71], Cyprus [72], Louisiana (USA) [73], and the world average value [18]; lower than the corresponding values of two other regional studies of Bangladesh (Northern and Southern districts) [21, 22], Kanyakumari (India) [28], two other studies of Pakistan (Bahawalpur, and Punjab) [30, 33], Juban (Yemen) [45], Kestanbol (Turkey) [55], Kirklareli (Turkey) [57], South Cameroon [67], and Caceres (Spain) [70]; and similar to the corresponding value of Izmir (Turkey) [58] (tab. 4).

Caesium-137: The average specific activity of ^{137}Cs in soil samples of Bangladesh determined was 5.37 4.87 Bq/kg with a range from 2.76 1.51 to 26.79 2.23 Bq/kg. The highest activity 26.79 2.23 Bq/kg was found in the soil sample of Jaflong while the lowest detected activity 2.76 1.51 Bq/kg was found in the soil samples of Natore (above MDC) (tab. 2). Jaflong is situated at the downstream of Indian state "Meghalaya" and the level of ^{137}Cs activity detected in the soil sample of Jaflong was a clear indication of nuclear activities in its upstream (since no nuclear activity was held at that area within Bangladesh). Out of 56 soil samples collected from 56 different locations of Bangladesh, no ^{137}Cs activity was detected in 14 samples (25%), *e. g.*, Srimangal, Chandpur, Gopalgonj, *etc*. The geometric mean and standard deviation of ^{137}Cs concentration in soil samples found were 5.30 Bq/kg and 1.96, respectively, from the corresponding cumulative frequency plot of ^{137}Cs in soil samples. The approximately equal arithmetic and geometric average concentration values of ^{137}Cs in soil samples reveal the normal distribution. The frequency distribution of the concentrations of ^{137}Cs is shown in fig. 5.

The average concentration level of ^{137}Cs in soil samples of Bangladesh was found to be higher than those of other regional studies of Bangladesh (Dhaka, Chittagong, Southern districts of Bangladesh) [19, 20, 22], Pakistan (Bahawalpur, Lahore, and Punjab) [30, 31, 33], Juban (Yemen) [45], Coastline of Saudi Arabia [50], and Algeria [65]; lower than those of Jordan [46] which have exceptionally higher activity, Mazandaran (Iran) [53], Iraq [54], three regional Turkish studies (Marmara, Kirklareli, and Bursa) [56, 57, 59], Serbia and Montenegro [68], and Louisiana (USA) [73]; and similar to that of Sichuan (China) [38]. The details of the ^{137}Cs concentration in soil samples along with the concentration of natural radionuclides in the soil of some of the countries of the world are given in tab. 4.

Correlation between the activities of radionuclides

The correlation coefficients between the concentrations of radionuclides ^{232}Th, ^{238}U, ^{40}K, and ^{137}Cs found in soil samples were calculated and are shown in tab. 3. In soil samples, the highest correlation coefficient (r = 0.88) was found for ^{232}Th and ^{238}U concentration levels; and the lowest (r = 0.13) was found for ^{40}K and ^{137}Cs. A good correlation between ^{232}Th and ^{238}U in soil samples and a very poor correlation between ^{40}K and ^{137}Cs were observed. The highly significant correlation between ^{232}Th and ^{238}U is consistent with the geochemical behaviour of their complexes, namely, the tendency of uranium and thorium to concentrate in the fluid phase during magmatic differentiation [14].

Table 3. The correlation coefficients between the concentrations of radionuclides

Serial number	Names of radionuclides between which correlation coefficient is calculated	Correlation coefficient (r)
1	^{232}Th and ^{238}U	0.8817
2	^{232}Th and ^{40}K	0.3562
3	^{232}Th and ^{137}Cs	0.2929
4	^{238}U and ^{40}K	0.3411
5	^{238}U and ^{137}Cs	0.1744
6	^{40}K and ^{137}Cs	0.1254

Radiological hazard indices

Radium equivalent activity: The highest Ra_{eq} 308.55 Bq/kg was found in the soil sample of Rangpur, while the lowest Ra_{eq} 95.23 Bq/kg was found in the soil of Nabigonj with an average 212.26 43.93 Bq/kg. The highest Ra_{eq} value found in the present study was consistent with the study done by Hamid *et al*. [21].

The average radium equivalent activity Ra_{eq} of the soil samples of Bangladesh was found to be higher than those of three other regional studies of Bangladesh (Dhaka, Chittagong, and Jessore) [19, 20, 23], Gudalore (India) [25], Kalpakkam (India) [27], Northern India [29], Pakistan [30-33, 36], Sri Lanka [17], Sichuan (China) [38], Taiwan [39], Songkhla (Thailand) [40], one study of Malaysia [42], Vietnam [44], Juban (Yemen) [45], Tafila (Jordan) [47], Saudi Arabia [48-52], four studies of Turkey (Marmara, Kirklareli, Izmir, and Bursa) [56-59], Tripoli (Libya) [60], three studies of Egypt [61, 63, 64] but its sand samples [62], Nigeria [66], Serbia and Montenegro [68], Ireland [69], Cacers (Spain) [70], Hungary [71], Louisiana (USA) [73], and the world average value [18]; lower than those of two other studies of Bangladesh (Northern and Southern Districts of Bangladesh) [21, 22], Ooty (India) [24], Kanyakumari (India) [28], China [37], another study of Malaysia [43], Mazandaran (Iran) [53] which was remarkably higher, Kestanbol (Turkey) [55], and south Cameroon [67]; and similar to the corresponding value of another Kalpakkam study [26]. The details of the values are shown in tab. 4.

Representative level index: The I_r was found to be ranged from 2.25 Bq/kg to 0.70 Bq/kg with an average 1.55 0.32 Bq/kg. The highest and lowest level indices were respectively found in the samples of Rangpur and Nabigonj.

External hazard index: The average values of the H_{ex} was found to be 0.29 0.06 which is below the permissible value 1. The H_{ex} values were found to be ranged between 0.42 and 0.13 for the soil samples of Rangpur and Nabiganj, respectively.

Internal hazard index: The mean value of the H_{in} was found to be 0.69 0.15 which is below the permissible value 1. The highest and the lowest values of H_{in} were also found in soil samples of Rangpur and Nabiganj with the values 1.01 and 0.31, respectively. The highest H_{in} value was slightly above (1%) the permissible value. This finding is consistent with the findings of Hamid *et al.*, [21]. Since Bangladesh has no country-wide formal cancer registry, therefore no comment could be drawn on the radiation hormesis or cancer incidence at the north-west part of Bangladesh especially at the Rangpur region.

Absorbed dose rate: The average absorbed dose rate at gonad level (1 m above the ground) due to the natural radionuclides in the soil samples was found to be 97.27 20.03 nGy/h ranging from 43.50 to 140.97 nGy/h. The lowest and the highest dose rates were, respectively, found due to the radioactivity in the soil samples of Nabiganj (in Habiganj District) and Rangpur.

Annual effective dose rate: The average annual effective dose rate was found to be 119.37 24.58 μSv/y due to the activity of naturally occurring radionuclides in surface soil having a range from 53.39 to 173 μSv/y obtained for the mentioned areas, respectively, by considering the outdoor occupancy factor of 20%. The similarity of arithmetic and geometric mean values of the terrestrial radiation dose levels indicated the normal distribution of terrestrial radiation throughout Bangladesh.

Outdoor dose level

The average of the outdoor dose rates at the points of sample collection was found to be 0.20 0.07 μSv/h, the minimum being 0.16 0.02 μSv/h in Sylhet, Srimangal, and Sitakundo; while the maximum being 0.28 0.04 μSv/h in Nachole. The average annual outdoor dose rate was estimated to be 1.87 0.27 mSv/y. By considering the average worldwide indoor-outdoor dose ratio of 1.27 [74] and the time occupancy factors of 66.67% indoors and 33.33% outdoors for Bangladeshi people, the average total environmental radiation dose rate was found to be 2.21 mSv/y which is close to the corresponding world average value [4]. The annual outdoor dose rates of different sampling points are given in the last column of tab. 1. All of the outdoor dose levels were further investigated by cumulative frequency plot (probability plot) to find out the geometric means and geometric standard deviations. It was found that there was no variation in the arithmetic and geometric mean values for all types of measurement, which indicates that the data were normally distributed. A poor correlation coefficient 0.34 was found between the effective annual outdoor dose rate due to radioactivity in surface soil and the outdoor dose rate measured by the portable radiation dose ratemeter. This indicated a non-uniform variation of cosmic and other non-terrestrial radiation levels at the measuring points during the study period.

CONCLUSIONS

The activity concentration levels of radionuclides in soil samples of different regions are somewhat higher than that of most reported values of the other countries

Table 4. Comparison of data on average radioactivity [Bqkg^{-1}] in surface soil in different countries of the world

Sl No.	Reference	Area	^{238}U	^{232}Th	^{40}K	^{137}Cs	Ra_{eq}
01	Present study	Bangladesh	44.35 12.65	83.56 17.96	630.89 173.85	5.37 4.87	212.26 43.93
02	Miah *et al*. [19]	Dhaka, Bangladesh	33 7[b]	55 14[c]	574 111	7 2	155.73
03	Chowdhury *et al*. [20]	Chittagong, Bangladesh	37.2 21.0	60.0 29.2	438 142	1.08 1.09	154 74
04	Hamid *et al*. [21]	Northern Bangladesh	91 10[b]	151 24	1958 400	NA[a]	426 60
05	Chowdhury *et al*. [22]	Southern Bangladesh	42 7[b]	81 14	833 358	2.08 3.35	221 40
06	Kabir *et al*. [23]	Jessore, Bangladesh	48 9[b]	53 9	481 78	NA[a]	161 20
07	Selvasekarapandian *et al*. [24]	Ooty, India	43.2 23.2	114.6 52.5	274.6 86.7	NA[a]	228.04
08	Selvasekarapandian *et al*. [25]	Gudalore, India	37.7 10.1	75.3 44.1	195.2 85.1	NA[a]	160.29
09	Kannan *et al*. [26]	Kalpakkam, India	16	119	406	(1.0-2.8)[f]	217.23
10	Sowmya *et al*. [27]	Kalpakkam, South India	22.6 12.6	92.8 44.3	434.1 131.1	NA[a]	188.56
11	Shanthi *et al*. [28]	Kanyakumari, India	20 14[b]	114 97	940 742	NA[a]	255.16
12	Mehra *et al*. [29]	Northern India	50.50	83.04	337.53	NA[a]	198.86
13	Matiullah *et al*. [30]	Bahawalpur, Pakistan	32.9 0.9[b]	53.6 1.4	647.4 14.1	1.5 0.2	158.5 4.1
14	Akhtar et al. [31]	Lahore, Pakistan	25.8[b]	49.2	561.6	Below LLD	139.29
15	Tahir *et al*. [32]	Punjab, Pakistan	35 7[b]	41 8	615 143	NA[a]	141 27
16	Jabbar *et al*. [33]	Punjab, Pakistan	50.6 1.7	62.3 3.2	662.2 32.1	3.1 0.3	190.8 8.7
17	Mujahid *et al*. [34]	Baluchistan, Pakistan	(15-27)[b,f]	(20-37)[f]	(328-648)[f]	NA[a]	NA[a]
18	Mujahid *et al*. [35]	Sind, Pakistan	(18-47)[b,f]	(24-69)[f]	(254-769)[f]	NA[a]	NA[a]
19	Hewamanna *et al*. [17]	Srilanka	34.84	72.17	584.60	NA[a]	182.91
20	Ziqiang *et al*. [37]	China	61.5 37	89.8 74	524 162	NA[a]	230.09
21	Wang *et al*. [38]	Sichuan, China	26[b]	49	440	6	130
22	Tsai *et al*. [39]	Taiwan	22.53	33.43	406.62	NA[a]	101.72
23	Kessaratikoon *et al*. [40]	Songkhla, Thailand	67.66 4.96[b]	45.00 3.19	213.05 23.03	NA[a]	146.92
24	Abdul Rahman *et al*. [41]	Ulu Tiram, Malaysia	44.41 4.77[e]	215.45 11.57[e]	NA[a]	NA[a]	NA[a]
25	Alias *et al*. [42]	Malaysia	19.45[b]	28.55[d]	103.08	NA[a]	60.68
26	Saat *et al*. [43]	Malaysia	99.13[b]	139.98[d]	598.24	NA[a]	334.49
27	Huy *et al*. [44]	Vietnam	42.77 18.15[b]	59.84 19.81	411.93 230.69	NA[a]	160.06 54.48
28	Abd El-mageed *et al*. [45]	Juban, Yemen	44.4 4.5[b]	58.2 5.1	822.7 31	4.779 0.4	190.83
29	Al Hamarneh *et al*. [46]	Jordan	NA[a]	NA[a]	NA[a]	88 31	NA[a]
30	Abu-Haija, [47]	Tafila, Jordan	22.3	27.91	285.02	NA[a]	84.10
31	El-Aydarous, [48]	Taif, Saudi Arabia	23.8 2.4[b]	18.6 1.7	162.8 7.6	NA[a]	62.85
32	Alaamer, [49]	Riyadh, Saudi Arabia	14.5 3.9[b]	11.2 3.9	225 63	NA[a]	47.8
33	Al-Trabulsy *et al*. [50]	Coastline, Saudi Arabia	11.4 1.5[b]	22.5 3.7	641.1 61.3	3.5 0.7	92.9
34	Al-Zahrani, [51]	Albaha, Saudi Arabia	37[b]	32	343	NA[a]	116
35	El-Taher *et al*. [52]	Al-Qassim, Saudi Arabia	9.5 2.8[b]	12.6 3.3	546 23	NA[a]	68.1
36	Abbaspour *et al*. [53]	Mazandaran, Iran	1189 7838	65 162	545 139	10.41 7.86	1323.78
37	Ali *et al*. [54]	Iraq	NA[a]	NA[a]	NA[a]	21.9	NA[a]
38	Merdanoglu *et al*. [55]	Kestanbol, Turkey	105.12	192	1207	(0.37 0.22 to 36.03 0.54)[f]	498
39	Kilic *et al*. [56]	Marmara, Turkey	21.77 12.08	26.63 15.90	442.51 189.85	27.46 21.84	93.85
40	Taskin *et al*. [57]	Kirklareli, Turkey	28 13	40 18	667 281	8 5	136.45
41	Fusun Cam *et al*. [58]	Izmir, Turkey	38	63	633	NA[a]	176.69
42	Akkaya *et al*. [59]	Bursa, Turkey	25 3[b]	26 3	435 44	8.52 2.59	96
43	Shenber, [60]	Tripoli, Libya	10.5	9.5	270	NA[a]	44.84
44	Saleh *et al*. [61]	Alexandria, Egypt	16.43 2.89[b]	18.31 5.25	268.18 81.65	NA[a]	63.22
45	Abel-Ghany *et al*. [62]	Egypt (sand samples)	94.94	80.22	700.79	NA[a]	263.60
46	Harb *et al*. [63]	Egypt	15.6	11.98	430.63	NA[a]	65.84
47	El-Taher *et al*. [64]	Safaga Dunes, Egypt	28.82[b]	14.03	558.39	NA[a]	91.57
48	Baggoura *et al*. [65]	Algeria	(5-176)[b,f]	(3-144)[c,f]	(36-1405)[f]	(0.3-0.41)[f]	NA[a]
49	Avwiri *et al*. [66]	Nigeria	19.16 1.23	21.26 1.41	224.26 10.1	NA[a]	60.75
50	Beyala Ateba *et al*. [67]	South Cameroon	134 64[b]	177 102	1482 280	NA[a]	500.86
51	Bikit *et al*. [68]	Serbia and Montenegro	51 9	53 8	554 92	19.54 9.38	169.33
52	McAulay *et al*. [69]	Ireland	37	26	350	NA[a]	101.07
53	Baeza *et al*. [70]	Caceres, Spain	45[b]	49	650	NA[a]	165
54	Papp, [71]	Hungary	33.3	32.1	418	NA[a]	111.31
55	Tzortzis *et al*. [72]	Cyprus	(0.01-39.3)[f]	(0.01-39.8)[f]	(0.04-565.8)[f]	NA[a]	NA[a]
56	DeLaune *et al*. [73]	Louisiana, USA	34 29	36 12	472 223	23 1	121.74
57	UNSCEAR-2000. [18]	Worldwide	32	45	420	NA[a]	128.59

[a]NA – not available; [b]measurement had been doe on ^{226}Ra in the corresponding decay chain; [c]measurement had been done on ^{228}Ac in the corresponding decay chain; [d]measurement had been done on ^{228}Ra in the corresponding decay chain; [e]elemental concentration ppm given in the paper had been converted into radiological concentration [Bqkg^{-1}]; and [f]range

(tab. 4). This is perhaps due to the geological characteristics of the earth. No significant amount of ^{137}Cs in the soil samples was detected. No significant difference was found in average concentration levels of the measured radionuclides in soil of different regions of Bangladesh. So there is no obvious influence of nuclear explosions and accidents on the environment of Bangladesh and have no radiation impact of Chernobyl accident on the environment of Bangladesh. However, slightly elevated level of internal hazard index (H_{in}) was found in Rangpur region. The present study will serve as a baseline radiological data for Bangladesh.

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РАДИОЛОШКА ПРОЦЕНА ПОВРШИНСКОГ ЗЕМЉИШТА БАНГЛАДЕША

Употребом HPGe детектора високе резолуције измерене су специфичне активности радионуклида ^{232}Th, ^{238}U, ^{40}K и ^{137}Cs на 56 локација нетакнутог површинског земљишта, које прекривају читав простор Бангладеша. Средње вредности специфичних активности поменутих радионуклида износила су по редоследу: 83.56 17.96 Bq/kg, 44.35 12.65 Bq/kg, 630.89 173.85 Bq/kg и 5.37 4.87 Bq/kg. Уочена је добра корелација између активности ^{232}Th и ^{238}U. Израчунати су следећи радиолошки параметри: еквивалентна активност радијума (Ra_{eq}), индекс репрезентативног нивоа ($I_{\gamma r}$), индекс спољашњег хазарда (H_{ex}), индекс унутрашњег хазарда (H_{in}), јачина апсорбоване дозе (D) и спољашња јачина дозе услед природних радионуклида. По датом редоследу, средње вредности ових параметара су: 212.26 43.93 Bq/kg, 1.55 0.32 Bq/kg, 0.29 0.06, 0.69 0.15, 97.27 20.03 nGy/h, и 119.37 24.58 μSv/y. Нивои зрачења на местима сакупљања узорака мерени су преносивим инструментима. Средња вредност спољашње јачине дозе била је 0.20 0.07 μSv/h, у опсегу од 0.16 0.02 μSv/h до 0.28 0.04 μSv/h. Утврђено је да је расподела природних радионуклида нормална. Нивои концентрације различитих радионуклида упоредиви су са одговарајућим регистрованим вредностима у земљиштима других држава.

Кључне речи: површинско земљиште, Бангладеш, гама спектрометрија, индекс радијационог хазарда, јачина дозе