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Radioactivity Distributions in Soils from Habiganj District, Bangladesh and their Radiological Implications

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ABSTRACT

A high purity germanium detector (HPGe) which is a low background gamma-ray spectrometry system, was used for radioactivity measurement of soils from Habiganj District of Bangladesh to establish a radiation map within this area as a baseline record for future studies. The radioactivity concentration levels of ^{238}U (Ra_{eq}), ^{232}Th and ^{40}K were measured in soil samples. From the measured specific activities of the above three natural radionuclides, the radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}), the external gamma absorbed dose rate and the annual effective dose were calculated in this study. The activity concentration levels were found to be in the range of 5 to 19 Bq kg^{-1} for ^{238}U (Ra_{eq}), 7 to 38 Bq kg^{-1} for ^{232}Th, and 93 to 392 Bq kg^{-1} for ^{40}K with mean values of 11, 22 and 227 Bq kg-1, respectively. No ^{137}Cs was found in this study. Radium equivalent activity (Raeq), gamma absorbed dose rate, external hazard index (H_{ex}) and annual effective dose values were found to be 59 Bq kg^{-1}, 28 nGy h^{-1}, 0.162 and 33 µSv, respectively, which indicates that the study area is radiologically safe for human beings.

Keywords: Annual effective dose rate, HPGe detector, radioactivity, radium equivalent activity, external hazard index.

INTRODUCTION

Radioactive elements are generally classified into two categories, naturally occurring and artificially produced. Radioactivity due to natural radionuclides in rocks, soil and water generates a significant component of background radiation exposure to the population in the area. The terrestrial component of the natural background radiation is dependent on the composition of the rocks, soil and water in which the natural radionuclides are contained (Karahan and Bayulken, 2000). Among the radioactive elements in the environment, the most abundant are ^{40}K, and the radioisotopes of the natural decay series of $2^{38}U$ and ^{232}Th, which are present in the earth's crust. Therefore materials from the earth's crust such as soil, and building materials become a major source of external radiation exposure to humans in the environment (United Nations Scientific Committee on the Effect of Scientific Radiation (UNSCAER), 1993; Lopez *et al*., 2004). In general,

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the release of low levels of artificial radionuclides occurs during the normal operations of nuclear facilities (e.g., nuclear ore processing, uranium enrichment, fuel fabrication, operation of reactors, operation of particle accelerators, etc.), and the production and application of radioisotopes in the fields of nuclear medicine, research, and industrial and agricultural processes (UNSCEAR,1977; Pearson and de Fraja Frangipane, 1975). The fission product ^{137}Cs is strongly absorbed and retained by soil as natural radionuclides which are randomly distributed at different depths of soil. Humans are exposed to natural terrestrial radiation that originates predominantly from the upper 30 cm of the soil. Humans are also exposed to radiation via contamination of the food chain, which occurs as a result of direct deposition of radionuclides on plant leaves, root uptake from contaminated soil or water and from direct ingestion of contaminated water. To assess these exposures, radioactivity studies have been previously carried out on soil samples in other parts of the world. (Santos Junior *et al*., 2010; El-Arabi *et al*., 2000; Ahmed *et al*., 2006; Veiga *et al*., 2006; El-Mageed *et al*., 2010; Al-Jundi *et al*., 2003; Al-Sulaiti *et al*., 2008; El-Shershaby *et al*., 2002; Arafa *et al*., 2004; Badhan *et al*., 2009; Alaamer *et al*., 2008).

Habiganj District is a border district between Bangladesh and India. Although it is important from both geological and natural resources perspectives, information about radioactivity have not been measured This study deals with natural radioactivity for soil samples in this area to establish a baseline record in this area and to assess any radiological hazards emanating from such radioactivity. Hence, the radium equivalent activity (Ra_{eq}), gamma absorbed dose rate, the external hazard index (Hex), and annual effective dose rate were evaluated and compared to the limits proposed by United Nations (UNSCEAR, 2008).

MATERIALS AND METHODS

Geological Outline

Habiganj District is in the north-eastern part of Bangladesh (*Figure 1*). It is located at 91°10′ E - 91° 40′ E longitude and 23°57′ N - 24°42′ N latitude. It was established as a district only 27 years ago, but is developing rapidly due to its natural resources. It has three gas fields, namely Habiganj, Bibiyana and Rashidpur. Mineral sand is also found in Habiganj. The Indian state of Tripura is to the south of Habiganj. The geographical position and natural resources of Habiganj indicate the likelihood of radioactivity but information on radioactivity is scarce as systematic measurements have not being undertaken in this area so far. This study aimed to study the concentration of natural radionuclides in soils of Habiganj District to establish baseline radioactivity data for the area.

Sample Collection and Preparation

Twenty-two soil samples were collected at a depth of 0-5 cm from twenty different locations in Habiganj District by employing a conventional method. At the laboratory, samples dried at room temperature, were crushed, cleaned, and

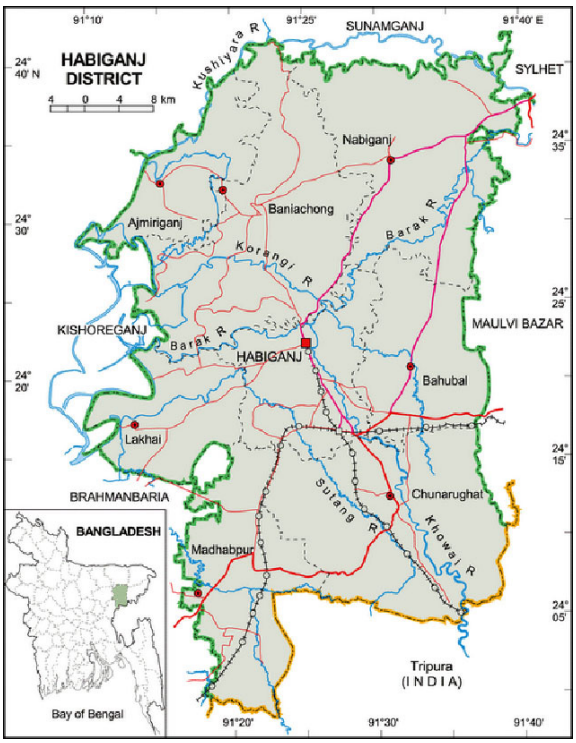

Figure 1: Map of sample locations within Habiganj District, Bangladesh

ground in a large sized mortar and their respective weights were determined. All the samples were then dried in an oven at 105ºC for 24 hours, homogeneously ground, and 250 g of each sample was then transferred into uncontaminated empty plastic containers measuring 7 cm in diameter and 8.3 cm in height and labeled. The containers were then sealed tightly with screw-on caps, with the gap between the cap and cotainer wrapped with thick vinyl tape. The filled containers were stored for a period of about 4 weeks. This was done to enable ^{226}Ra and ^{228}Ra reach their secular equilibrium with its decay products in the Uranium series and Thorium series, respectively. The sealing of the containers was also necessary to ensure that radon gas and its progeny were measured.

Experimental Procedure

Each sample was measured with a gamma-ray counting system, a high resolution EG & G Orte high purity germanium (HPGe) co-axial detector coupled with a Silena Emca Plus multichannel analyser (MCA), and associate microprocessors. The effective volume of the detector was 83.47 cm^3. The energy resolution of the detector was found to be 1.69 keV at 1332 keV energy peak of ^{60}Co (*Figure 2*) with a relative efficiency of 19.6%. The detector was shielded with copper ring (2 mm) at the side and lead (76.2 mm) at the side and the top to reduce background radiation level. To minimise the effect of scattered radiation from the shield, the detector was located at the center of the chamber. The samples were then placed

over the detector for at least 5000 seconds. The spectra were either evaluated with the computer software program Maestro (EG & G ORTEC), or manually with the use of a spread sheet (Microsoft Excel) to calculate the natural radioactivity. The γ-ray energies of ^{228}Ac (911 keV) and ^{228}Ac (969 keV) were used to determine the concentration of ^{232}Th, and the γ-transition of ^{214}Bi (609 and 1120 keV) and ^{214}Pb (295 and 351 keV) were used to determine the activity of ^{238}U. The ^{40}K and ^{137}Cs radionuclides were measured from their respective γ-ray energies of 1460 keV and 662 keV. In order to determine the background distribution in the environment around the detector, an empty sealed plastic container was measured in the same manner as the rest of the samples. The background spectra were used to correct the net peak area of gamma rays of the measured isotopes.

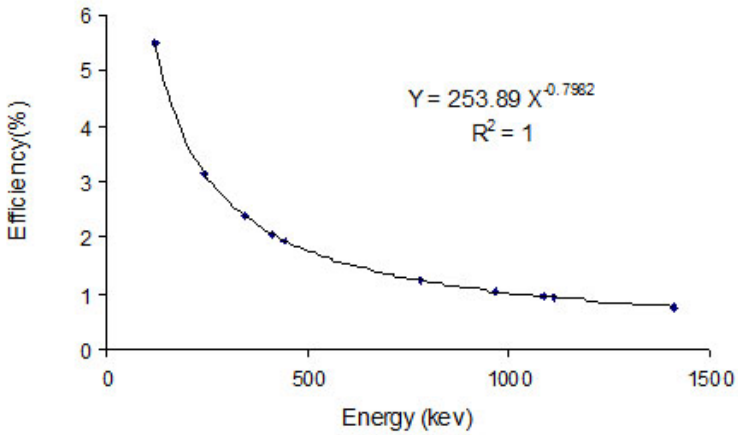

Figure 2: Efficiency calibration curve

The energy calibration of the MCA was obtained using standard point sources such as ^{22}Na, ^{57}Co, ^{60}Co, ^{133}Ba, ^{137}Cs, etc. The efficiency of the detector for different radionuclides of interest of different energies were determined by mixing standard sources of known activities and different energies such as 122, 245, 344, 411, 444, 779, 963, 1086, 1112 and 1408 keV supplied by Health Physics Division, Atomic Energy Centre, Dhaka (AECD) and following the standard method. The unknown efficiencies of different radionuclides were then calculated using Eq. (1) to draw a standard efficiency curve (*Figure 2*). The efficiency calibration curve was drawn up using different energy peaks covering arange of up to 2000 keV to obtain the efficiency of the detector for the particular gamma ray energy of interest.

$$\text{Efficiency} = \frac{net\ \ counts\ per\ \text{sec}ond}{\text{p}_\gamma \cdot A}\ \text{X}\ 100\% \qquad (1)$$

where
P_γ = fraction of number of gamma-rays emitted from a particular radionuclide
A = activity of the radionuclide present in the samples

The radioactivity concentrations of different radionuclides were based on the measured detector efficiencies (for corresponding radionuclides) as a function of the energy curve for the same counting geometry and time. Thus, using their measured efficiencies, the activities of the measured radionuclides were calculated with Eq. (2).

$$\text{Activity (A)} = \frac{net\ \ counts\ per\ \text{sec}ond}{p_\gamma \cdot \varepsilon \cdot \omega} \qquad (2)$$

where
A= activity of the radionuclide in Bq kg^{-1} present in the sample
P_γ = the fraction of number of gamma-rays emitted from a particular radionuclide
ε = absolute efficiency of the detector for a particular gamma-ray energy emitted from the specific radionuclide of interest
ω = weight of the sample

Calculation of the Radiological Effects
The most widely used radiation hazard index is called the radium equivalent activity (Ra_{eq}). The Ra_{eq} is a weighted sum of activities of the ^{238}U, ^{232}Th and ^{40}K radionuclides based on the assumption that 370 Bq kg^{-1} of ^{238}U, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same gamma ray dose rate (Kohshi, 2001). Radium equivalent activity can be calculated from the relation in Eq. (3) suggested by Beretka and Mathew (1985).

$$Ra_{eq} = (A_{Th} \times 1.43) + A_U + (A_k \times 0.077) \qquad (3)$$

where
ATh = activity concentration of ^{232}Th in Bq kg^{-1}
AU = activity concentration of ^{238}U in Bq kg^{-1}
AK = activity concentration of ^{40}K in Bq kg^{-1}

The external hazard index (H_{ex}) due to the emitted gamma-rays of the samples was calculated using Eq.(4).

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (4)$$

where
A_U, A_{Th}, and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively

The calculated average H_{ex} was found to be less than unity.

The total air absorbed dose rate (nGy h^{-1}) due to the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1} was calculated using Eq. (5) (UNSCEAR, 1993; Beck, 1972):

$$D = 0.462A_U + 0.604A_{Th} + 0.042A_k \quad (5)$$

where
A_U, A_{Th} and A_K are the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively in Bq kg^{-1}. El-Shershaby (2002) derived this equation for calculating the absorbed dose rate in air at a height of 1.0 m above the ground from the measured radionuclides concentrations in environmental materials.

To estimate the annual effective dose, the following was taken into account: (a) the conversion coefficient from absorbed dose in air to effective dose; and (b) the indoor occupancy factor. Applying the conversion factor of 0.7 Sv Gy^{-1} to convert absorbed dose in air to human effective dose, and an outdoor occupancy factor of 0.2 as recommended by UNSCEAR (2000), the average annual effective dose due to gamma radiation from terrestrial sources of all soil samples in this study was assessed using Eq. (6) (UNSCAER, 2000).

$$D_{eff}\,(Sv) = D(nGy\ h^{-1}) \times (24 \times 365)(h) \times 0.7 \times 0.2 \quad (6)$$

where
D_{eff} = effective dose equivalent in Sv
D = total absorbed dose rate in air in (nGy h^{-1})

RESULTS AND DISCUSSION

Activity Concentrations in Soil Samples

The highest activity concentration for ^{238}U and ^{232}Th was observed in Nobigonj and Azmirigonj soils (18.83 ± 0.016 and 38.14 ± 0.034 Bq kg^{-1}), respectively, whilst the lowest activity concentrations were observed in Chunarughat (5.28 ± 0.015 and 7.13 ± 0.025 Bq kg^{-1}, respectively) (Table 1). The average concentration of ^{238}U and ^{232}Th was measured as 11.09 ± 0.01 Bq kg^{-1} and 21.98 ± 0.029 Bq kg^{-1}, respectively. The highest activity concentration for ^{40}K was observed in Lakhai (392 ± 0.74 Bq kg^{-1}) whilst the lowest activity concentration was observed in Baniachong soil (93.14 ± 0.62 Bq kg^{-1}). The average concentration of ^{40}K was measured as 227 ± 0.68 Bq kg^{-1}. It was also observed that the measured activity concentration of ^{40}K exceeded markedly the values of both uranium and thorium as it was the most abundant radioactive element under consideration (Table 1).

TABLE 1
Radioactivities of ^{238}U, ^{40}K and ^{232}Th in soil samples at different locations of Habigonj district.

Sample location	Sample no	^{238}U (Bq kg^{-1})	^{232}Th(Bq kg^{-1})	^{40}K(Bq kg^{-1})
Habiganj Sadar	1	9.83 ± 0.016	22.51 ± 0.029	169.64±0.66
	2	8.91 ± 0.012	24.62 ± 0.029	263.52±0.69
	3	6.96 ± 0.015	17.93 ± 0.028	263.35±0.69
Chunarughat	4	14.16 ± 0.015	16.99 ± 0.031	321.57±0.72
	5	7.54 ± 0.015	**7.13 ± 0.025**	212.22±0.67
	6	**5.28 ± 0.015**	13.30 ± 0.027	151.44±0.65
Bahubal	7	6.45 ± 0.027	13.76 ± 0.026	241.56±0.67
	8	8.70 ± 0.015	17.22 ± 0.028	165.52±0.65
	9	11.74± 0.015	14.19 ± 0.027	161.25±0.66
Lakhai	10	14.71 ± 0.012	34.82 ± 0.033	231.85±0.68
	11	14.69 ± 0.011	34.33 ± 0.032	350.29±0.72
	12	11.08 ± 0.012	21.81 ± 0.029	**392.63±0.74**
Azmirigonj	13	12.65 ± 0.012	**38.14 ± 0.034**	233.19±0.68
	14	11.24 ± 0.012	27.88 ± 0.031	191.65±0.66
Baniachong	15	12.36 ± 0.012	32.86 ± 0.033	218.61±0.68
	16	12.61 ± 0.012	30.37 ± 0.032	**93.14±0.62**
Nobigonj	17	**18.83 ± 0.015**	19.25 ± 0.028	284.78±0.69
	18	11.25 ± 0.016	21.28 ± 0.029	356.54±0.73
	19	11.55 ± 0.012	16.47 ± 0.028	176.91±0.66
Madhabpur	20	14.71 ± 0.016	17.97 ± 0.029	250.71±0.69
	21	10.52 ± 0.012	20.23 ± 0.028	154.43±0.64
Average		**11.09 ± 0.01**	**21.98 ± 0.03**	**227.9± 0.67**

Moreover, the average values obtained fell within the range of corresponding world values and other published results mentioned in Table 2. The world average activity concentration and ranges of ^{238}U, ^{232}Th , and ^{40}K are 35 Bq kg^{-1} with a range of 17 - 60 Bq kg^{-1}, 30 Bq kg^{-1} with a range of 11 - 64 Bq kg^{-1} and 400 Bq kg^{-1} with a range of 140 - 850 Bq kg^{-1}, respectively (UNSCEAR, 2000). The average activity concentrations for ^{238}U, ^{232}Th and ^{40}K in this study were lower than world averages for these radionuclides in the soils. The anthropogenic radionuclide ^{137}Cs was also analysed in this study that has been assumed to be deposited in soil of Bangladesh as a result of atmospheric fallout following the Chernobyl disaster on 26 April 1986 and other previous atmospheric tests of nuclear devices around the world (Mollah *et al*., 1986; Miah *et al*., 1985). No ^{137}Cs was detected in any of the samples. The errors quoted were the standard deviations from the means and represent the spread in the concentrations of the natural radionuclides in the soil. It can be seen that the activity concentration of ^{238}U, ^{232}Th and ^{40}K in this study were comparable with other published results in Bangladesh (Kabir *et al*., 2009).

TABLE 2
Comparison of radioactivity levels of the soil samples of different countries with that of this study

Countries	Average Specific radioactivity of ^{238}U (Bq kg^{-1})	Average Specific radioactivity of ^{232}Th (Bq kg^{-1})	Average Specific radioactivity of ^{40}K(Bq kg^{-1})	References
Egypt	17	18	320	(UNSCEAR, 2000)
USA	40	35	370	(UNSCEAR, 2000)
China	32	41	440	(UNSCEAR, 2000)
Japan	33	28	310	(UNSCEAR, 2000)
Malaysia	67	82	310	(UNSCEAR, 2000)
India	29	64	400	(UNSCEAR, 2000)
Iran	28	22	640	(UNSCEAR, 2000)
Denmark	17	19	460	(UNSCEAR, 2000)
Poland	26	21	410	(UNSCEAR, 2000)
Greece	25	21	360	(UNSCEAR, 2000)
Romania	32	38	490	(UNSCEAR, 2000)
Spain	32	33	470	(UNSCEAR, 2000)
Luxembourg	35	50	620	(UNSCEAR, 2000)
Bangladesh	48	53	481	(Kabir, K. A., 2009)
Saudi Arabia	15	11	225	(Alaamer, A. S., (2008)
Nigeria	14	19	896	(Okeyode, I., Oluseye, A. 2010)
Turkey	21	25	298	(Bozkurt A., 2007)
Pakistan	30	56	642	(Akhtar, N., 2004)
West Bank-Palestine	69	48	630	(Dabayneh K M., 2008)
Worldwide average	35	30	400	(UNSCEAR, 2000)
Habigonj (Bangladesh Present study)	11.09	21.98	227	

Radiation Hazard Indices

The average Ra_{eq} was 59 Bq kg^{-1} (Table 4). This was below the recommended value 370 Bq kg^{-1} which the OECD deems safe (1979) (OECD,1979). The average absorbed gamma dose measured in air was 28 nGy h^{-1} (Table 4). According to the recent UNSCEAR reports, the corresponding world average value is 58 nGy h^{-1}. This indicates that the average absorbed dose rate in air outdoors from the soil was lower in Habiganj District than the world average.

The calculated value of H_{ex} obtained was 0.16 (Table 4). The calculated values of H_{ex} obtained in this study ranged from 0.135 – 0.346, which were found

TABLE 3
Average activity concentration of ^{238}U, ^{232}Th & ^{40}K in soil samples for different regions within Bangladesh.

Sample location	Average activity concentration in Bq kg^{-1}			Reference
	^{238}U	^{232}Th	^{40}K	
Chittagong	35	60	438	(Chowdhury *et al.,* 1999)
Pabna	33	47	449	(Roy*et al.,* 2001)
Dhaka	33	55	574	(Miah *et al.,* 1998)
Nine southern districts	42	81	833	(Chowdhury *et al.,* 2006)
Jessore	48	53	481	(Miah *et al.,* 1985)
Sitakunda	31	62	467	(Rahman *et al.,* 2012)
Kuakata Sea Beach	29	91	875	(Islam *et al.,* 2012)
Sylhet	55	125	491	(Chowdhury *et al.,* 1999)
Bangladesh (average)	48	53	481	(Miah *et al.,* 1985)
Habiganj (Bangladesh, Present study)	11.09	21.98	227	

to be lower than the recommended value of 1 implying that the radiation hazard is insignificant for the population living in the investigated areas of Habiganj District (UNSCEAR, 2008). The annual effective dose was calculated as 9.07 μSv, which was below annual maximum permissible dose level (1mSv or 1000 μSv) to the general public (ICRP, 1990).

CONCLUSION

The measurements made showed that the levels of radioactivity from the decay chain of ^{238}U and ^{232}Th, as well as the primordial radionuclide ^{40}K were present in all soil samples. The measured activity concentrations of ^{238}U, ^{232}Th and ^{40}K across the soil samples varied from 5.28 ± 0.02 Bq kg^{-1} to 18.83 ± 0.12 Bq kg^{-1}, 7.13 ± 0.04 Bq kg^{-1} to 38.14 ± 0.13 Bq kg^{-1}, and 93.14 ±0.06 to 392 ±0.74 Bq kg^{-1},respectively and with mean values of 11.09 ± 0.01 Bqkg^{-1}; 21.98 ± 0.03 Bq kg^{-1}, and 227 ± 0.70 Bq kg^{-1},respectively. The average activity concentrations for ^{238}U, ^{232}Th and ^{40}K in this study were lower than the worldwide average for these radionuclides in soils. The artificial radionuclide ^{137}Cs was not observed in statistically significant amounts above background level in this study. This radioactivity monitoring study may be continued to establish a complete database of environmental radioactivity in Bangladesh.

Ra_{eq}, gamma absorbed dose rate, the H_{ex}, and annual effective dose rate of the soil samples collected were 59 Bq kg^{-1}, 28 nGy h^{-1}, 0.162, 33 μSv, respectively. The results show that the H_{ex} values for all soil samples were below the limit of unity, meaning that the radiation dose is below the permissible limit of 1 mSv y^{-1} recommended by ICRP 60 for general public (ICRP, 1990).

The values of the radiation hazard parameters from this study were not extremely high compared to either the world averages or the recommended limits,

TABLE 4
Radium equivalent activity, dose rate, annual effective dose and External Hazard Index for soil samples at different locations in Habiganj

Sample location	Sample no	Radium equivalent activity, Ra_{eq} ($Bqkg^{-1}$)	Dose Rate, D (nGy/h)	External hazard index H_{ex}	Annual effective dose, D_{eff} (10^{-6} Sv)
Habiganj Sadar	1	53.90	25.26	0.155	30.98
	2	62.57	30.06	0.174	36.86
	3	51.04	25.11	0.143	30.80
Chunarughat	4	60.97	30.31	0.171	37.17
	5	32.60	16.71	0.092	20.49
	6	34.90	16.83	0.097	20.64
Bahubal	7	43.04	21.44	0.121	26.29
	8	44.91	21.37	0.124	26.21
	9	43.33	20.77	0.120	25.48
Lakhai	10	80.74	37.57	0.222	46.07
	11	88.31	42.24	0.245	51.80
	12	69.76	34.79	0.196	42.66
Azmirigonj	13	83.52	38.68	0.230	47.43
	14	64.53	30.08	0.178	37.17
Baniachong	15	74.67	34.75	0.206	20.49
	16	62.56	28.08	0.171	30.98
Nobigonj	17	66.28	32.28	0.184	39.59
	18	66.65	33.03	0.187	40.51
	19	47.48	22.71	0.132	27.86
Madhabpur	20	57.96	28.18	0.161	34.56
	21	50.26	23.56	0.139	28.90
	22	47.33	21.96	0.130	26.94
Average		**58.51**	**27.99**	**0.160**	**33.18**

and therefore unlikely to cause additional radiological health risks to the people living in the areas studied. The average concentrations of the radionuclides in soil samples collected from Habiganj District were at the normal environmental levels and were similar to the concentrations obtained in the surrounding countries.

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