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Assessment of Natural Radioactivity and Radiation Hazard in Soil Samples of Rajbari District of Bangladesh

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Abstract
The activity of naturally occurring ^{238}U, ^{232}Th and ^{40}K in soil samples collected from different Upazila of Rajbari district of Bangladesh, have been studied and possible health effect were estimated. The experiment has been done by using gamma ray spectrometry system consisting of a HPGe detector coupled with MCA and associate electronics. The average natural radioactivities of ^{238}U, ^{232}Th and ^{40}K have been measured as 29.03±5.67 Bq/kg, 50.91±10.17 Bq/kg and 535.32±89.19 Bq/kg respectively. To estimate health effect due to the activity of those radionuclides, the radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), absorbed dose rate (D) and effective dose have been calculated and compared to the world average values. The results are also compared with the literature values reported for other regions of the world and found that the soil of the study area are not hazards by the radiation and does not pose any harmful effect to the environment. The outcome of this study may provide valuable information about radiation hazard and also may take part in the monitoring of environmental radioactivity.

Key Words: *Radionuclide, absorbed dose, external hazard index, radium equivalent activity, annual effective dose.*

Introduction
Soil provides a direct source of radioactivity in food chain due to its uptake by agriculture plants. The radionuclides also find their way into milk, meat and aquatic life. Thus soil is a major source and pathway of radio nuclides to living beings. The distribution of radionuclides in nature, their concentration and movements can seriously be affected by the activities of population. Sometimes this can result in deleterious effects such as harmful consequences on environment and health hazards to human being, either into the body through different metabolic pathways. For the assessment of effective dose equivalent to the population of Bangladesh, it is necessary to estimate the concentration of various radionuclides in the environment, entering into the body as a function of time.

The natural radioactivity in the environment is the main source of radiation exposure for human being that may come from available natural sources, nuclear weapon tests and nuclear accidents. Natural radionuclide in soil contributes a significant amount of background radiation exposure to the population through inhalation and ingestion. The main contributors of radionuclides are ^{238}U, ^{40}K and ^{232}Th and these radionuclides are not uniformly distributed in soils and vary from region to region [Miah *et al.* 2012]. Natural radioactivity is wide spread in the environment and it exists in various geological formations in soil, rocks, plants, water and air. So the Knowledge of the distribution pattern of the both anthropogenic and natural radionuclides in soil plays an important

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role in radiation protection and measurement. The radioactivity concentration of these radionuclides above permissible level is very harmful to the human body.

Therefore, measurements of natural radioactivity in soils and the radiation doses are of great interest to the researchers which have led the nationwide surveys throughout the world [Karahan *et al.* 2000, Dogerlier *et al.* 2008]. Some researchers have found higher concentrations of ^{238}U and ^{232}Th in the soils of Dhaka Export Processing (DEPZ) area, Savar of Bangladesh that might be derived from the parent materials, such as clay stones during the soil formation process [Mahfuza *et al.* 2003]. Further studies have also been carried out to determine the background levels of radionuclides in soils and thus to calculate the absorbed dose in air [Kannan *et al.* 2002].

Studies on radiation levels and radionuclides distributions in different Upazila of Rajbari district have not been carried out so far. The goal of this study is to determine natural radioactivity (^{238}U, ^{232}Th, and ^{40}K) levels in soil samples collected from different Upazila of Rajbari district. Besides these, the radium equivalent, absorbed dose rate, effective dose and external hazard indices have also been calculated and compared with the literature values to sort out the radiation hazard caused by the activity. This study will also be helpful to establish a research baseline in this area.

Materials and Methods

Sample Collection and Preparation

Soil samples (0-15 cm depth) were collected during 2009 to 2010 from twelve different locations of Rajbari district. After removing the stones and vegetation, all soil samples were oven dried at 105^0C for 24 h. Then they were grinded homogeneously and each sample of 400g was kept in the plastic container of 7.2 cm diameter and 8.3 cm in height labeled with appropriate identifications and left for four weeks to reach radioactive equilibrium. Blank plastic container of same geometry was used for background counts.

Method of Analysis

Radioactivity of soil samples were measured using a low level gamma counting system, a HPGe detector with a relative efficiency of 19.6% and energy resolution of the 1.33 MeV for the 1332 keV ^{60}Co peak. The corresponding background counts for each sample have been measured for 5000s and it was subtracted from the sample activity counting. The radioactivities of ^{238}U and ^{232}Th were estimated on the assumption that they are in secular equilibrium with their respective daughter products within four weeks [Chowdhury *et al.* 1999]. The measurement was based on the recording natural radioactivity quantities of three natural long-lived elements: ^{238}U, ^{232}Th and ^{40}K which are considered the photopeaks at 1760, 2610 and 1460 keV respectively. The concentration of ^{212}Pb, ^{208}Tl, ^{214}Pb and ^{214}Bi occur in the samples due to disintegration of ^{238}U and ^{232}Th respectively. Therefore, the activities of ^{212}Pb, ^{208}Tl, ^{214}Pb and ^{214}Bi in the samples indicate the presence of ^{238}U and ^{232}Th respectively. In Table 1 the γ-ray energy and emission rate for those of nuclides are tabulated [Akkurt *et al.* 2009]. The γ- activity was calculated from the equation (1)

$$Activity(A) = \frac{C}{\xi(E) \times I_\gamma \times W} \quad \text{..... (1)}$$

Here,
C =peak area counts per sec.
ξ(E) =efficiency of the detector at energy E (keV)
$I\gamma$ = γ- ray intensity of the source of interest
A = Activity in Bq/kg
W = weight of the soil sample (kg).

Table 1: γ-ray energy and emission rate for ^{238}U, ^{232}Th and ^{40}K radionuclide

Element	Nuclide	Half life	γ- energy (keV)	Emission rate
^{238}U	^{214}Pb	26.8 min	295.2, 351.2	19, 38.9
	^{214}Bi	19.9 min	609.3, 1764.7	43.3, 17
^{232}Th	^{208}Tl	3.05 min	583.2, 2614.7	185.77, 99.79
	^{228}Ac	6.13 h	911.1	27.7
^{40}K		$1.3\text{x}10^9$ yr	1460.8	10.7

The distribution of ^{238}U, ^{232}Th and ^{40}K in soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/kg to compare the specific activity of materials containing different amounts of ^{238}U, ^{232}Th and ^{40}K. It is calculated using the following relation [UNSCEAR, 1988]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.07C_K \quad \text{……………………………………………………(2)}$$

Where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively. It has been assumed that 370 Bq/kg of ^{226}Ra or 259 Bq/kg of ^{232}Th or 4810 Bq/kg of ^{40}K produces the same gamma dose rate [Prasong *et al.* 2008].
The soils and sediments are used for making earthen huts, bricks and pottery materials and, hence, the external hazard index, H_{ex} is estimated using formula as [UNSCEAR, 1988]:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \quad \text{………………………………………… (3)}$$

Where C_{Ra}, C_{Th}, and C_K have the same meaning in equ. (2). The value of this index must be less than unity in order to keep the radiation hazard insignificant. The maximum value of H_{ex} equal to unity corresponds to the upper limit of radium equivalent activity (370 Bq/kg).
The external gamma absorbed dose rate in air at 1m above ground level was calculated according to the following relation [UNSCEAR, 1988]:

$$D = 0.462\ C_{Ra} + 0.604\ C_{Th} + 0.042\ C_K \quad \text{……………………………………(4)}$$

Where D is the dose rate in nGy/h and C_{Ra}, C_{Th} and C_K have the same meaning in equ. (2).

In order to judge the health effects of the absorbed dose, annual effective dose should be obtained. The conversion coefficient from the absorbed dose in air to the effective dose (0.7Sv/Gy) and the outdoor occupancy factor (0.2) proposed in ref. [UNSCEAR, 1988], the annual effective doses are calculated:

Annual effective dose ($mSvy^{-1}$) = D ($nGyh^{-1}$) x 24x 365(hy^{-1}) x 0.2 x 0.7($SvGy^{-1}$) x10^{-6}…. …(5)

Results and Discussion

The results of activity concentrations of radionucldes obtained from gamma spectrometry measurements for 12 soil samples collected from Rajbari district are presented in Table 2. Besides the resulting radium equivalent activity, gamma dose rate, annual effective dose and external hazard indices due to activity concentration of the natural radionuclides of soil samples were measured and the values are presented in Table 3. Distribution of the radionuclides ^{40}K, ^{232}Th and ^{238}U was found as 87%, 8% and 5% respectively in the studied soil samples.

Table 2: Concentrations of ^{238}U, ^{232}Th and ^{40}K in soil samples

Sample Id. No.	Concentration of Radioactivity in Bq/kg		
	^{232}Th	^{238}U	^{40}K
S-1	46.25±2.33	28.01±2.10	595.55±54.11
S-2	54.10±2.97	20.61±1.92	446.69±50.12
S-3	36.97±1.72	23.50±2.53	603.15±53.01
S-4	44.12±2.08	35.13±2.61	448.29±51.03
S-5	60.80±3.10	36.75±3.65	510.31±52.61
S-6	49.23±2.72	30.25±2.04	738.60±54.35
S-7	51.10±2.50	29.01±2.41	576.20±54.21
S-8	75.01±2.85	33.85±1.94	460.11±48.55
S-9	50.98±2.72	24.51±1.25	576.32±55.01
S-10	45.31±2.32	36.14±3.50	480.98±62.50
S-11	54.05±2.53	25.43±1.89	458.52±45.25
S-12	43.01±1.95	25.15±1.82	529.15±42.51

(± Values represent counting error).

The activity of ^{238}U for soil samples in the investigated area ranged from 20.61±1.92 to 36.75±3.65 Bq/kg with an average of 29.03±5.67 Bq/kg. For ^{232}Th, the activity concentration was found in the range of 36.97±1.72 to 75.01±2.85 Bq/kg, with an average of 50.91±10.17 Bq/kg. The activity concentration of ^{40}K was obtained from its photo-peak of 1460.80 keV and the obtained results for soil ranged from 446.69±50.12 to 738.60±54.35 Bq/kg with an average of 535.32±89.19 Bq/kg respectively. The resulting radium equivalent activity Ra_{eq} values for the soil samples in this study was calculated using the eq.(2) and the values are shown in Table 3. Table 2

shows that the activity concentration of thorium is higher than uranium, which is evident from the fact that thorium is 1.5 times higher than that of Uranium in earth's crust [Kabir *et al.* 2009]. It is also observed that the activity of ^{40}K exceeds significantly the values of both ^{238}U and ^{232}Th, as it is the most abundant radioactive element under consideration. Moreover, the excessive use of the Potassium containing fertilizer in the area adjacent to the sampling sites may contribute to the higher values of K activity. In the present study, activity concentrations of ^{238}U, ^{232}Th and ^{40}K are comparable (except northern district ref. [Hamid *et al.* 2002]) with the reported values presented in Table 4, especially with the different regions of Bangladesh [Chowdhury *et al.* 1999, Roy *et al.* 2001, Miah *et al.* 1998, Chowdhury *et al.* 2006]. Moreover, our obtained values fall within the range of corresponding world average values and other published results mentioned in Table 4. The comparison of absorbed dose rate obtained in our study with that of others of the world is shown in Table 5.

Table 3: Radium equivalent activity, dose rate, annual effective dose and external hazard index of soil samples

Samples Id. No.	Ra_{eq} (Bq/kg)	Dose rate (nGy/h)	Annual effective dose (mSv/y)	External hazard index
S-1	117.63	58.47	0.072	117.63
S-2	138.42	68.79	0.084	138.42
S-3	152.43	77.28	0.095	152.43
S-4	137.55	69.13	0.085	137.55
S-5	157.01	78.00	0.096	157.01
S-6	138.55	69.01	0.085	138.55
S-7	142.80	71.62	0.088	142.80
S-8	160.68	78.92	0.097	160.68
S-9	129.78	64.14	0.079	129.78
S-10	108.1	54.21	0.066	108.10
S-11	130.21	64.63	0.079	130.21
S-12	132.22	66.19	0.081	132.22

The Ra_{eq} values for soil in the study area varied from 118.59 Bq/kg to 173.32 Bq/kg. These values are less than 370 Bq/kg, which are acceptable for safe use [UNSCEAR, 1988]. The mean absorbed gamma dose rate due to the activity of ^{238}U, ^{232}Th and ^{40}K in soil samples was measured as 66.64±6.81 nGy/ h (range 58.52 nGy/h to 80.27 nGy/h) and found to be comparable with the world average value of 55 nGy/h [UNSCEAR, 1993]. The calculated values of external hazard index obtained in this study due to natural gamma radiation were 0.33 to 0.47 with the average of 0.39. Since these values are lower than unity, we can say that the radiation hazard due to soil is low for this region. The estimated values of annual effective dose range from 0.072 mSv to 0.098 mSv (average 0.082 mSv), is lower than the world average of 0.480 mSv.

Table 4: Comparison of radioactivity level of the of soil samples of different countries with that of the present work

Country	Activity in Bq/kg		
	^{238}U	^{232}Th	^{40}K
Chittagong (Chowdhury, M.I. *et al*., 1999) (Bangladesh)	34.6	60	438
Pabna (Roy, S. *et al*., 2001) (Bangladesh)	33	47	449
Dhaka (Miah, F. K. *et al*., 1998) (Bangladesh)	33	55	574
Nine southern districts (Bangladesh) (Chowdhury, M. I. *et al*., 2006)	42	81	833
Jessore (Kabir, K. A. *et al*., 2009) (Bangladesh)	48.32 (28-67)	53.34 (33-70)	481.35 (345-674)
Northern districts (Bangladesh) (Hamid, B. N. *et al*., 2006)	91	151	1958
Sylhet (Miah, A. *et al*., 2012) (Bangladesh)	55.25 (23.91-143.70)	125.27 (53.25-369.60)	497.91 (369.35-593.02)
All India average (Prasad, N. G. S. *et al*., 2008)	31	63	394
Peshwar (Ali, S. *et al*., 1996) (Pakistan)	65	84	646
Louisiana (USA) (Delaune, R. D. *et al*., 1986)	43-95	50-190	43-729
Zacatecas (Mexico) (Noordin, I. (1999)	23	19	530
Rajbari (Bangladesh) [Present study]	29.03±10.17	50.91±5.67	535.32±89.19
Worldwide average (UNSCEAR, 1993)	40(15-50)	40(7-50)	580(100-700)

Table 5: Comparison of absorbed dose rate obtained in our study with that of others of the world

Country	Year	Absorbed dose rate in $nGyh^{-1}$	Range $nGyh^{-1}$	References
Bangladesh (Sylhet)	2012	124.12±7.59	70.52-315.60	Miah, A. *et al*., 2012
Bangladesh (Jessore)	2009	77	62-100	Kabir, K. A. *et al*., 2009
Bangladesh (North. dist.)	2002	222	160-274	Hamid, B. N. *et al*., 2006
China	1972	69	--	UNSCEAR, 1993
Romania	1979	81	32-210	UNSCEAR, 1993
Nigeria	2000	128	5-186	Ajayi, O.S., 2000
Rajbari (Bangladesh)	2012	66.64	58.52 - 80.27	Present study

Conclusion

The study showed that the radioactivity levels of ^{238}U, ^{232}Th and ^{40}K were 29.03±10.17 Bq/kg, 50.91±10.17 Bq/kg and 535.32±89.19 Bq/kg respectively and were comparable with the other regions. However, slight variation in the radioactivity content in soil observed with different locations worldwide mainly due to soil type, formation and transport process involved. This may be the reason for the variation observed results. The mean absorbed dose rate obtained in the present study (66.64±6.81 nGy/ h) is comparable to the world average (55 nGy/h). The calculated annual effective dose with average value of 0.082 mSv is lower than the world average value (0.480 mSv). All values obtained for radium equivalent activity are less than 370 Bq/kg, which are acceptable for safe use as recommended by the IAEA [UNSCEAR, 1988]. The measured values of external hazard index found in this study range from 0.33 to 0.47 (all are lower than unity), which means that it does not cause any health hazard to the population living in the investigated area.

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