

DETERMINATION OF RADIONUCLIDES IN SOIL FROM BARENDRA REGION IN RAJSHAHI AND MINING REGION IN DINAJPUR

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ABSTRACT The level of natural radioactivity in soil samples calculated from the Barendra region, Rajshahi and the Mining region, Dinajpur was measured and the radiological hazard parameters in both the regions were estimated. The activity concentrations of naturally occurring radioactive materials in the ²³⁸U and ²³²Th decay chains and in ⁴⁰K were determined by means of a gamma-ray spectrometry system using an HPGe detector in a low background configuration at Atomic Energy Centre, Dhaka. The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K found in the present study are compared with the reported values of the Northern region and other districts of Bangladesh and those of other countries of the world. Radiological hazard parameters of the two regions were estimated and compared with the world averages. The natural radioactivity in the Barendra and Mining region poses no threat to general public there. Artificial radionuclide ¹³⁷Cs was not found in the soil samples studied.

(Keywords: Radioactivity, HPGe Detector, Soil, Barendra and Mining region, Radiological hazard parameters)

INTRODUCTION

Radionuclides are the sources of radioactivity and emit nuclear radiations which have become a part of our daily lives. The majority of naturally occurring radionuclides belong to the radionuclides in the ²³⁸U and ²³²Th series, and the single decay radionuclide, ⁴⁰K [1]. According to the National Council on Radiation Protection and Measurements Report, the most significant source of radiation exposure to humans is due to natural radiation in the environment [2]. This exposure to naturally occurring radiation also accounts for up to 85% of annual exposure dose received by the world population and any dose of radiation involves a possible risk to human health [3].

In order to protect the general public health against the radiation risk originating from Naturally Occurring Radioactive Material, the measurement of radioactivity in the environment needs to be considered to assess the biological effect on the human. This has also become the focus of greater attention by the IAEA in recent years [4]. The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K and their progeny present in soil are measured in many countries of the world [5] including Bangladesh in order to monitor radiation level in the environment. Dinajpur is a district in Northern Bangladesh and located at the latitude and longitude coordinates of 25°33'N and 88°43'E (Figure 1).

There are sites of coal mine and stone mine at a distance of 40 km from the town. Coals are the major sources of radioactive materials released to the environment and coal combustion is more hazardous to health. Some regions in the Rajshahi District are called Barendra region and located at the latitude and longitude coordinates of 24°26'14N and 88°36'59E (Figure 1). It is in the Northwest part of Bangladesh. Main rivers are the Padma, the Mahananda, the Baral and the Barnai. So, it is highly possible to suffer from radioactivity due to situation of the source of the rivers in India.

Also the large number of agricultural activities, high demand for utilization of fertilizers may give rise to enhancement in the level of background radioactivity in particular areas. As these have different geological characteristics, we planned to examine the activity concentrations of the soil of the Mining region of Dinajpur and Barendra region of Rajshahi.

The present study has been carried out to evaluate the level of radioactivity due to ²³⁸U, ²³²Th and ⁴⁰K in soil samples collected from the Barendra region of Rajshahi and the Mining region of Dinajpur. The aim of this research not only the determination of natural radioactivity in soil samples but also the determination of the radiological hazard parameters such as Gamma Dose Rate (D), Radium Equivalent Activity (Ra_{eq}), External Hazard Index (H_{ex}) and Annual Effective Dose Equivalent (D_{eff}) for individuals living in both regions.

MATERIALS AND METHODS

Sample Collection and Preparation: In 2012, total 22 Soil samples were collected in the selected sites of Barendra region (Godagari, Rajshahi) and Mining region (Dinajpur) in the northern part of Bangladesh separately (11 samples from Barendra Region and other 11 samples from Mining Region). All samples were carefully collected at 5cm-10cm depth from the soil-surface in agriculture land by using shovel and each of the samples weighed approximately 1kg. At the laboratory of Health physics Division in Atomic Energy Center Dhaka, the collected samples were transferred from the polyethylene bags to the acetone-cleaned stainless steel bucket and a crowbar was used to break the big soil stone separately. Then the samples were

dried at 105°C until a constant weight was achieved and it was ensured that any significant moisture was removed from the samples. Each of the dried samples was grounded to fine powder in an agate motor separately. The powdered samples were then sieved using a fine aperture mesh screen (mesh size 2 μ m) in order to remove extraneous items like plant material, roots, pebbles etc. and to obtain a fine grained sample that would present a uniform matrix to the detector. Finally each of the grounded samples was transferred to cylindrical plastic-container (7.3 cm diameter \times 8.9 cm height). The containers were then labeled properly and sealed tightly, rapped with thick vinyl tapes around their screw necks. The samples were stored for at least four weeks before counting in order to attain secular equilibrium.

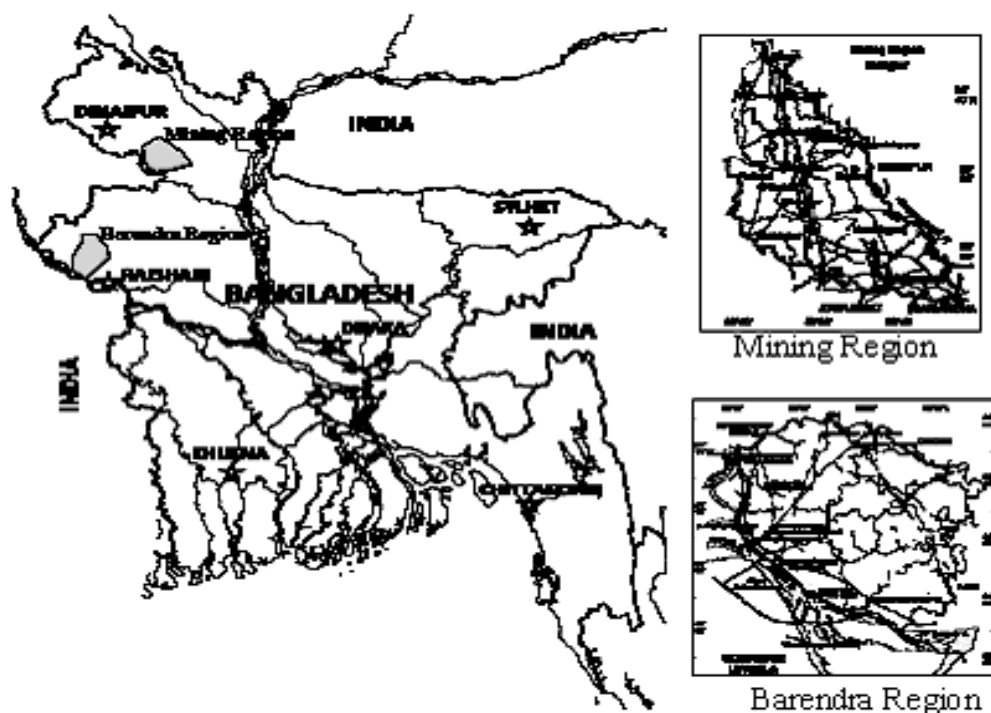


Figure 1. Map of the sample location in Bangladesh.

Measurement procedures and Data Analysis:

The activity concentration of gamma ray emitting radioisotopes in the samples were measured by using a gamma ray spectrometer with a high-resolution HPGe coaxial detector coupled with a Silena Emcaplus multichannel analyzer (MCA). The effective volume of the detector was 83.469 cm³ and energy resolution of the 1.33 MeV energy peak for ⁶⁰Co was found as 1.69 keV at full width

half maximum (FWHM) with a relative efficiency of 19.6%. The Ge-detector was cooled with a liquid nitrogen cryostat to reduce the leakage current.

The detector was enclosed with a cylindrical lead shield to reduce the background radiation from various natural radiation sources and to isolate from other radiation sources used in nearby surroundings. All samples were counted for 5000s. The peaks identified in Figures.2 (a) and (b) are

due to Pb²¹² (238.6keV), Pb²¹⁴ (295.2keV), Pb²¹⁴ (351.9keV), Tl²⁰⁸ (583.1keV), Bi²¹⁴ (609.3keV), Ac²²⁸ (911.2keV), Ac²²⁸ (968.9keV), Bi²¹⁴ (1120.2keV) and ⁴⁰K (1460.8keV). No ¹³⁷Cs line

$$A(Bqkg^{-1}) = \frac{N}{P_{\gamma} \times \varepsilon \times W} \text{-----(1)}$$

Where A is the activity concentration of each radio-nuclide in the sample, N is the net count of each radio-nuclide which is found by subtracting the sample radio-nuclide activity counting from background activity counting, ε is the detector efficiency of the specific γ ray, P_γ is the transition probability of the specific γ ray and W is the mass of the sample (kg). Error associated with every calculation was measured by standard deviation equation.

The activity concentrations of ²³⁸U and ²³²Th were determined from the average concentration of nuclides [Pb²¹⁴ (295.2keV), Pb²¹⁴ (351.9keV), Bi²¹⁴ (609.3keV) and Bi²¹⁴ (1120.2keV)] and [Pb²¹² (238.6keV), Tl²⁰⁸ (583.1keV), and Ac²²⁸ (911.2keV), Ac²²⁸ (968.9keV)] respectively. The activity concentrations of ⁴⁰K were determined

$$Ra_{eq}(Bq.kg^{-1}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \text{----- (2)}$$

Where, C_{Ra}, C_{Th} and C_K are the activity concentrations of ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K in Bq.kg⁻¹, respectively.

In order to assess any radiological hazard, the exposure to radiation arising from radionuclides present in soil can be determined in terms of many parameters. A direct connection between radioactivity concentrations of natural

$$D(nGy.h^{-1}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \text{-----(3)}$$

To limit the radiation exposure attributable to natural radionuclides in the samples to the permissible dose equivalent limit of 1 mSv.y⁻¹, the

$$H_{ex} = \left(\frac{C_{Ra}}{370}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right) \text{-----(4)}$$

In order to keep the radiation hazard insignificant, the value of external hazard index must not exceed the limit of unity. The maximum value of H_{ex} equal to unity corresponds to the upper limit of radium equivalent activity 370 Bq.kg⁻¹ [24].

was obtained at 661.66keV. The activity concentration of individual radionuclides was calculated from the following equation [6]:

directly by measurement of the gamma-ray transitions at 1460.8keV. Error associated with every calculation was measured by standard deviation equation.

Radiological hazard parameters: The radiological hazard parameters are calculated from the measured activity concentrations of the radionuclides ²³⁸U, ²³²Th and ⁴⁰K in soil samples. Due to a non uniform distribution of natural radionuclides in the soil samples, the actual activity level of ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K in the samples can be evaluated by means of a common radiological index named the Radium Equivalent Activity (Ra_{eq}). It is the most widely used index to assess the radiation hazards and was calculated using following equation [5]:

radionuclides and their exposure is known as the absorbed dose rate in the air at 1 metre above the ground surface.

The mean activity concentrations of ²²⁶Ra (²³⁸U), ²³²Th, and ⁴⁰K (Bq.kg⁻¹) in the soil samples are used to calculate the absorbed dose rate given by the following formula [7]:

external hazard index was calculated using the following formula [5]:

The absorbed dose rate in air at 1metre above the ground surface does not directly provide the radiological risk to which an individual is exposed. The absorbed dose can be considered in terms of the annual effective dose equivalent from outdoor terrestrial gamma radiation which is converted from the absorbed dose by taking into account two

factors, namely the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor.

$$D_{eff} (\mu Sv.y^{-1}) = D(nGy.h^{-1}) \times 8760h \times 0.2 \times 0.7Sv.Gy^{-1} \times 10^{-3} \text{ -----(5)}$$

The values of those parameters used in the UNSCEAR report (2000) are 0.7 Sv.Gy⁻¹ for the conversion coefficient from absorbed dose in air to effective dose received by adults and 0.2 Sv.Gy⁻¹ for the outdoor occupancy factor.

RESULTS AND DISCUSSION

The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K for different soil samples of Barendra and Mining regions are presented in Table 1. For Barendra region soil samples, it is found that the activity concentration of ²³⁸U ranges from 31.9±1.3 to 42.7±1.4 Bq.kg⁻¹ with mean 36.7±1.4 Bq.kg⁻¹, activity concentration of ²³²Th ranges from 52.9±1.6 to 68.9±1.7 Bq.kg⁻¹ with mean 59.7±1.7

The annual effective dose equivalent can be estimated using the following formula [7]:

Bq.kg⁻¹, activity concentration of ⁴⁰K ranges from 232.5±2.3 to 320.6±2.4 Bq.kg⁻¹ with mean 282.5±2.3 Bq.kg⁻¹.

For Mining region soil samples, activity concentration of ²³⁸U ranges from 31.3±1.3 to 48.3±1.5 Bq.kg⁻¹ with mean 37.1±1.4 Bq.kg⁻¹, activity concentration of ²³²Th ranges from 44.5±1.5 to 81.9±1.9 Bq.kg⁻¹ with mean 56.9±1.7 Bq/kg, activity concentration of ⁴⁰K ranges from 222.0±2.3 to 475.7±2.5 Bq.kg⁻¹ with mean 343.7±2.4 Bq.kg⁻¹. It is found that the mean values of ²³⁸U and ²³²Th concentrations are similar for both the regions. The mean of ⁴⁰K concentration in Mining region is higher than that in Barendra region.

Table 1. Activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in soil samples of Barendra region and Mining region.

Location	Sample codes	Average activity concentration in Bq.kg ⁻¹		
		²³⁸ U	²³² Th	⁴⁰ K
Barendra Region, Godagari, Rajshahi	RGMM	42.7±1.4	68.1±1.7	232.5±2.3
	RGSM	31.9±1.3	52.9±1.6	253.7±2.3
	RGCB	35.6±1.4	56.8±1.7	253.7±2.3
	RGDG	33.4±1.3	54.2±1.6	320.6±2.4
	RGKH	37.5±1.4	57.9±1.7	306.5±2.4
	RGAT	38.3±1.4	68.9±1.8	306.5±2.4
	RGBP	35.4±1.4	55.6±1.7	296.0±2.4
	RGJB	34.9±1.4	62.8±1.7	285.4±2.4
	RGBM	40.2±1.4	57.4±1.7	246.6±2.3
	RGKP	37.0±1.4	65.4±1.7	292.4±2.4
RGMD	37.3±1.4	56.9±1.7	313.6±2.4	
Mean		36.7±1.4	59.7±1.7	282.5±2.4
Mining region, Dinajpur	DFSA	37.3±1.4	59.6±1.7	243.1±2.3
	DFTP	42.4±1.5	47.6±1.6	475.7±2.5
	DFCP	38.6±1.4	55.9±1.6	352.3±2.4
	DPCM	48.3±1.5	81.9±1.9	222.0±2.3
	DPSM	31.3±1.3	51.0±1.6	250.2±2.3
	DPVP	33.8±1.3	56.6±1.7	296.0±2.4
	DHHL	29.5±1.3	53.0±1.6	377.0±2.4
	DBSA	36.5±1.4	59.7±1.7	454.5±2.5
	DBJN	32.5±1.3	44.5±1.5	429.9±2.5
	DNBP	34.9±1.4	57.4±1.7	377.0±2.4
	DNSA	43.0±1.4	58.2±1.7	303.0±2.4
Mean		37.1±1.4	56.9±1.7	343.7±2.4

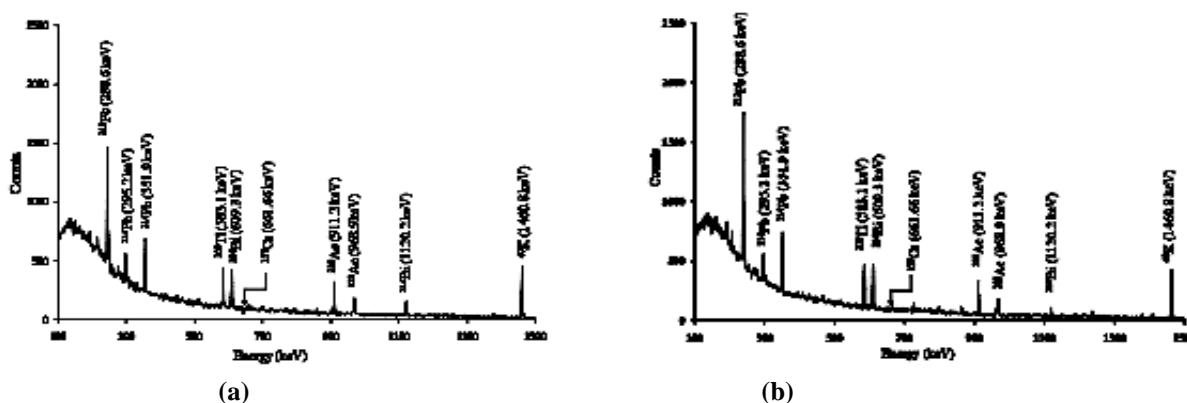


Figure 2. Spectrum of the gamma-rays from (a) one soil sample (sample code RGMM) of Barendra region and (b) one soil sample (sample code DPCM) of Mining region.

The radionuclide concentrations in the soil samples of the two regions are presented along with those of other places in Bangladesh in Table 2. The results of the present study are compared with those of the northern region at first. The ^{238}U and ^{232}Th concentrations of Rajshahi Barendra region and Dinajpur Mining region are about 2.5 times less than the high radioactivity zones of Rangpur, Lalmonirhat and Kurigram [8]. The ^{40}K activity concentration of the former is about 6 times less than the latter.

Thus, the high radioactivity zone seems to be localized. ^{238}U content in the two regions is comparable with the values of other districts in Bangladesh except that in Sylhet where the value is about 50% higher. ^{232}Th content in most of the districts are almost similar to these in the two regions except that in Sylhet and Kuakata Sea Beach where the values are \sim 110% and 50% higher respectively. The concentrations of ^{40}K in these two regions are much lower than the values in all other districts. No peak of man-made fission fragment ^{137}Cs (661.66 keV) was observed in the gamma-ray spectrum. It may be concluded that there is no ^{137}Cs radionuclide in the soil samples under study. However, it may be stated that the ^{137}Cs activity was beyond the detection limit of 1.54 Bq in our measurement system.

It seems that there is fresh nuclear fallout in the region studied. The average activity concentrations of ^{238}U , ^{232}Th and ^{40}K of the two regions are shown along with the values of other countries in Table 3. The ^{238}U concentrations in both regions are comparable with these of most of other countries of the world. In fact, the averages of these two regions are roughly equal to the world average. The ^{232}Th

concentrations of the two regions, on the other hand, are higher than those of most other countries and are almost double the world average. The ^{40}K concentrations of the two regions are smaller than the values of most countries and about 25% lower than the world average.

The values of the radiological hazard parameters are shown in Table 4. The Radium Equivalent Activity R_{eq} ranges from 125.2 to 158.3 Bq.kg^{-1} with mean 141.9 Bq.kg^{-1} for the Barendra region soil samples and ranges from 121.8 to 180.9 Bq.kg^{-1} with mean 142.5 Bq.kg^{-1} for the Mining region soil samples. It is found that the R_{eq} values for all soil samples in the present work are lower than the accepted safety limit value of 370 Bq.kg^{-1} as recommended by the Organization for Economic Cooperation and Development (OECD) [9]. The values are graphically shown in Fig 3(a).

The estimated Absorbed Dose Rate D based on soil radioactivity ranges from 61.4 to 72.2 nGy.h^{-1} with a mean value 64.9 nGy.h^{-1} for the Barendra region soil samples and ranges from 55.8 to 81.1 nGy.h^{-1} with mean 65.9 nGy.h^{-1} for the Mining region soil samples. These values are slightly greater than the worldwide mean value 60 nGy.h^{-1} [9]. The comparison is shown in Fig 3(b).

As listed in Table 4, the calculated values of the External Hazard Index H_{ex} vary from 0.34 to 0.43 and the average value is found to be 0.39 for the Barendra region and vary from 0.33 to 0.49 and the average value is found to be 0.39 for the Mining region.

Table 2. Comparison of average activity concentration of ^{238}U , ^{232}Th and ^{40}K in soil samples of different regions within Bangladesh.

Locations	Average activity concentration in Bq.kg^{-1}		
	^{238}U	^{232}Th	^{40}K
Chittagong [Chowdhury <i>et al.</i> , 1999]	35	60	438
Pabna [Roy <i>et al.</i> , 2001]	33	47	449
Dhaka [Miah <i>et al.</i> , 1998]	33	55	574
Nine southern districts [Chowdhury <i>et al.</i> , 2006]	42	81	833
Jessore [Kabir <i>et al.</i> , 2009]	48	53	481
Sitakunda [Rahman <i>et al.</i> , 2012]	31	62	467
Kuakata Sea Beach [Islam <i>et al.</i> , 2012]	29	91	875
Cox's Bazar [Alam <i>et al.</i> , 1999]	19	37	458
Sylhet [Miah <i>et al.</i> , 2012]	55	125	497
Rangpur [Hamid <i>et al.</i> , 2002]	87	140	1844
Lalmonirhat [Hamid <i>et al.</i> , 2002]	93	152	1951
Kurigram [Hamid <i>et al.</i> , 2002]	98	167	2168
Barendra region (present study)	36.7	59.7	283
Mining region (present study)	37.1	56.9	344

The results show that the H_{ex} values from all soil samples are below the limit of unity, meaning that the radiation dose is below the permissible limit of 1 mSv.y^{-1} recommended by ICRP 60 [10]. This is displayed in Fig 3 (c). The Annual Effective Dose Equivalent D_{eff} from outdoor terrestrial gamma radiation was estimated by taking into account the conversion coefficients from absorbed dose in air to effective dose and the outdoor occupancy factor.

The Annual Effective Dose Equivalent D_{eff} for the Barendra region in this study vary from 70.3 to $88.5 \mu\text{Sv.y}^{-1}$, with mean value of $79.6 \mu\text{Sv.y}^{-1}$ and for the Mining region it varies from 68.4 to $99.4 \mu\text{Sv.y}^{-1}$, with mean value of $80.8 \mu\text{Sv.y}^{-1}$, which is similar with the worldwide effective dose of $80 \mu\text{Sv.y}^{-1}$ [9]. The values are graphically depicted in Fig 3(d).

Table 3. Comparison of Average activity concentration of ^{238}U , ^{232}Th and ^{40}K in soil samples of different countries with the present study.

Countries	Average activity concentration in Bq.kg^{-1}		
	^{238}U	^{232}Th	^{40}K
Denmark [UNSCEAR, 2000]	17	19	460
Poland [UNSCEAR, 2000]	26	21	410
Greece [UNSCEAR, 2000]	25	21	360
Romania [UNSCEAR, 2000]	32	38	490
Spain [UNSCEAR, 2000]	32	33	470
Luxembourg [UNSCEAR, 2000]	35	50	620
Saudi Arabia [Alaamer, 2008]	15	11	225
Nigeria [Okeyode <i>et al.</i> , 2010]	14	19	896
Turkey [Bozkurt <i>et al.</i> , 2007]	21	25	298
Pakistan [Akhtar <i>et al.</i> , 2004]	30	56	642
West Bank-Palestine [Dabayneth <i>et al.</i> ,	69	48	630
Worldwide average [UNSCEAR, 2000]	35	30	400
Barendra region (present study)	37	60	283
Mining region (present study)	37	57	344

Table 4. Radium Equivalent Activity, Dose Rate, External Hazard Index and Annual Effective Dose Equivalent for samples in both regions.

Locations	Sample codes	Radium Equivalent Activity, Ra_{eq} (Bq/kg)	Absorbed Dose Rate, D (nGy/h)	External Hazard Index, H_{ex}	Annual Effective Dose, D_{eff} (10^{-6} Sv)
Barendra Region, Godagari, Rajshahi	RGMM	156.3	70.6	0.43	86.6
	RGSM	125.2	57.3	0.34	70.3
	RGCB	134.5	61.4	0.37	75.3
	RGDG	133.3	61.6	0.37	75.6
	RGKH	141.8	65.2	0.39	79.9
	RGAT	158.3	72.2	0.43	88.5
	RGBP	135.6	62.4	0.37	76.5
	RGJB	144.7	66.1	0.40	81.0
	RGBM	139.5	63.6	0.38	78.0
	RGKP	151.0	68.9	0.41	84.5
	RGMD	140.6	64.8	0.39	79.4
Mean		141.9	65.0	0.39	79.6
Mining Region, Dinajpur	DFSA	139.6	63.5	0.38	77.8
	DFTP	143.8	68.3	0.40	83.80
	DFCP	143.1	66.4	0.39	81.4
	DPCM	180.9	81.1	0.49	99.4
	DPSM	121.8	55.8	0.33	68.4
	DPVP	135.5	62.3	0.37	76.4
	DHHL	131.7	61.5	0.36	75.4
	DBSA	153.7	72.1	0.42	88.3
	DBJN	126.3	60.0	0.35	73.5
	DNSP	143.5	66.7	0.39	81.8
	DNSA	147.5	67.8	0.40	83.1
Mean		142.5	65.9	0.39	80.8

CONCLUSIONS

The activity concentrations of ^{238}U and ^{232}Th in the samples of the two regions are found to be similar; the ^{40}K concentration of Mining region is slightly higher. The ^{238}U and ^{232}Th concentrations are, in general, similar with those in most districts in Bangladesh except in Sylhet and in the high radiation background of Rangpur, Lalmonirhat and Kurigram districts. The ^{40}K concentrations in the samples of the present study are lower than those in the samples of all other districts in Bangladesh. The average ^{238}U concentration in the samples of the present study is similar with the world average. On the other hand, the average ^{232}Th concentration is double the world average.

The average ^{40}K concentration is lower than the world average. No ^{137}Cs is found in the samples of the present study. It seems that there is no fresh nuclear fallout in places under study. The Radium Equivalent Activity Ra_{eq} due to the radionuclides in the samples has been estimated. This value is found to be about 2.5 times less than the permissible

maximum value of 370 Bq.kg^{-1} . The Absorbed dose rate D estimated from the soil activity of the two regions are similar, but slightly greater than the worldwide mean value 60 nGy.h^{-1} . The External Hazard Index H_{ex} of the two regions is found to be 0.39 which is much below the maximum limit of unity. The Effective Dose Equivalents D_{eff} of the two regions under study is found to be $80\mu\text{Sv y}^{-1}$ which is similar with the worldwide effective dose of $80\mu\text{Sv y}^{-1}$. The natural radioactivity in the two regions poses no health-hazards to the population there.

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