Measurement of radioactive impact of the gas well accident on environment of Sylhet division in Bangladesh

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ABSTRACT

The main goal of this study is to carry out the public exposure contribution from radioactive materials occurring due to the accident of natural gas exploration well of Sylhet division in Bangladesh. This study has been performed by using a gamma spectrometry of Genie 2000 software embedded PC-based High Purity Germanium (HPGe) detector system. The measured activity concentrations of naturally occurring radionuclides ^{232}Th, ^{226}Ra, and ^{40}K have been in the public safety limit on the environment of the study areas. The specific activity of detected anthropogenic radionuclide ^{137}Cs has been found to be very little with a mean value of 1.65 ± 0.61 and 1.13 ± 0.57 Bq·kg^{-1} in the Magurchhera and Haripur gas field area respectively.

*Keywords***:** Natural gas well accident, Environmental radioactivity, Public exposure etc.

1. INTRODUCTION

Technological activities in mining and mineral processing lead to large volumes of wastes-containing naturally occurring radioactive materials (NORMS), which are dumped into the environment over prolonged periods of time. In addition, the wastes are in most cases in close proximity to densely populated areas as well as agricultural lands that spread over several kilometers. Natural radionuclide's, especially from the uranium–thorium series, may leach with time and reach dams, lakes and streams that are accessible to the public or agricultural lands. Surface water such as lakes and rivers or underground water may also be contaminated by direct discharge of large volumes of processed water that may contain radioactive materials. Farming practices can transfer contaminants through various pathways to the terrestrial environment and then to crops, vegetables and livestock. As a result, Human

beings are exposed to background radiation from radioactive contaminants of natural and man-made sources. Natural background radiation, which is equivalent to 2.4 mSv per person, accounts for ~ 80 % of the total radiation dose a person, receives annually [1]. Soil radionuclide activity concentration is one of the main determinants of the natural background radiation. Volcanic geographic structures as well as rocks that are rich in phosphate, granite and salt contain natural radionuclides like ^{238}U, ^{232}Th and ^{40}K. When rocks are disintegrated through natural processes, radionuclides are carried to the soil by rain and flows [2]. In addition to the natural sources, soil radioactivity is also affected by men. Caesium-137 is a fission by-product, has a half-life of 30.2 y with gamma radiations of 0.661 MeV [3]. Radioisotopes that are present in soil significantly affect terrestrial gamma radiation levels.

The effective gamma radiation levels are generally in the range of 18–93 nGy·h^{-1} with a mean of 59 nGy·h^{-1} [4]. Bangladesh has some natural resources; among them, natural gas contributes the largest amount. There are 26 gas fields in this country and the most of them are in Sylhet division. Three accidents have been occurred in the natural gas fields of Sylhet in the last days and amongst them, two natural gas exploration areas, Haripur and Magurchhera, have been chosen for measuring the radioactive impact on environment. The accident of these two natural gas well had been occurred in 1955 and in 1997 respectively. In Magurchhera, the exploration of this resource had been done after the necessary survey including the geophysical condition of the area. The Occidental Company of the USA had been started drilling of the gas well to elevate the natural gas in 1997. Unfortunately, the gas exploration well caught fire blew out from 840 meter depth at the time of drilling. The flame from the blazing gas rose as high as 500 feet. The sudden rise in the gas well seriously destroyed the drilling rigs and the machinery and the ^{137}Cs crystal, used in the drilling machine scattered into the environment. Sands and small pieces of stones were coming with the fire from the gas well. This might change the radiological condition of the locality. As a result, this study has been aimed for determining the environmental delayed impact of gamma radiation surrounding the gas exploration well areas and the following studies have been carried out:

(1) Determination of the radioactive concentration of natural radionuclide's ^{226}Ra, ^{232}Th and ^{40}K in soil of Magurchhara and Haripur gas exploration well area.
(2) Determination of the radioactive concentration of artificial radionuclide ^{137}Cs.
(3) Determination of the external and internal doses, radium equivalent activity and representative level index of the study area.

2. MATERIALS AND METHODS

2. 1. Study area and sampling

Sylhet division is one of the charming hilly areas of Bangladesh where having various types of natural resources. Among various natural resources natural gas is the important one economically. Three gas field accidents, Haripur, Magurchhera and Tengratila (Chhatak) have been occurred in the last days. Out of which two areas have been taken into consideration of this study. The study area Magurchhera gas field is situated at Kamalgang thana and Haripur gas field is situated at Goainghat thana in Sylhet division are shown in Fig. 1A and 1B respectively. A total number of 35 samples have been collected from these two study areas among which 28 are from Magurchhera and 8 are from Haripur gas field area. All of the samples have been collected using a rotating cylindrical plastic corer with an amount about to 1 kg.
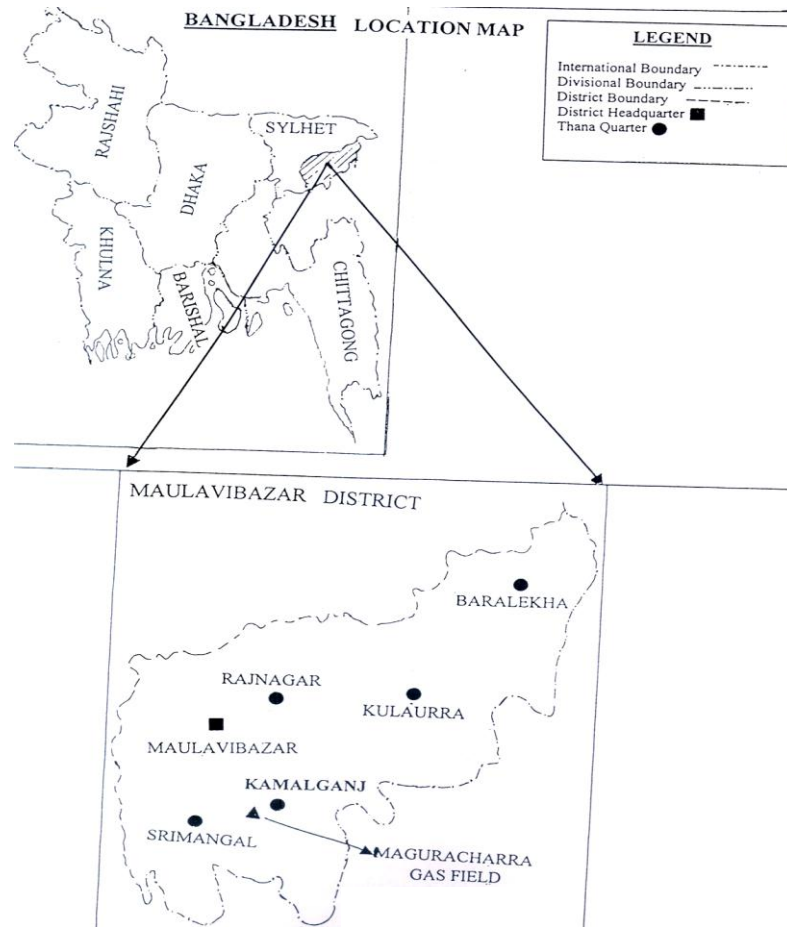
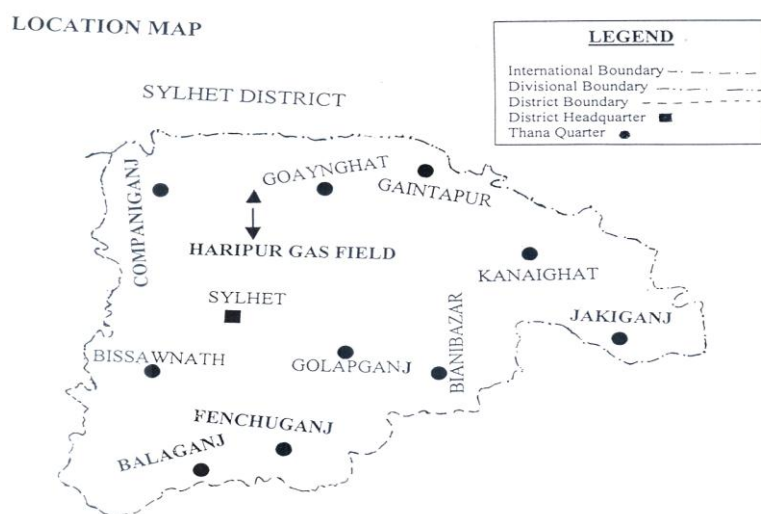**Fig. 1A.** Magurchhara gas field area in Sylhet division.

Fig. 1B. Haripur gas field area in Sylhet division.

2.2 Gamma spectrometry and analytical procedures

The preparation technique which has been used for the pre-treatment of soil samples [5] in this study is given in Fig. 2. The present research work has been performed by using a p-type coaxial lead shielded intrinsic HPGe detector of 132 cm^3 volume with relative efficiency of 35 % and resolution (FWHM) of 1.83 keV for the 1332 keV γ–ray energy of ^{60}Co. For analyzing the gamma spectrum, firstly the full-energy peak efficiency curve has been plotted by measuring the reference samples IAEA/RGU-1 (U ore), IAEA/RGTh-1 (Th ore) and IAEA/RGK-1 (K sulphate) from which the counting efficiencies of the gamma-ray peaks have been measured [6,7]. The geometry of the counting samples has been the same as that of the standard samples and the counting time for all the samples has been 20,000 seconds. All the spectrum acquisition and analysis have been performed by the computer based Canberra software of Genie 2000. For calculating the net counting of the samples, the background count due to naturally occurring radionuclide's in the environment around the detector has been subtracted from each sample counts. Corrections have also been made for self-absorption of the gamma-radiation by the samples [8,9]. The results expressed with the confidence limit or standard deviation of $\pm 1\sigma$.

In the present study, the following formulae have been used for the calculation of the activity concentrations of the samples and the corresponding radiological impact on the environment.

(a). The counting efficiency of the detector has been calculated by using the formula [10]

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

where, CPS = Net count per second (i.e. CPS from standard source − CPS for background)

Activity, A_C = Standard source activity for respective energy

Intensity, I_γ = Intensities of gamma energies.

(b). Specific activities of the radionuclide's presented in the soil samples have been calculated by using the formula [11]

$$\text{Activity} = \frac{CPS \times 100 \times 1000}{\varepsilon_f \ (\%) \times I_\gamma \times w_s \ (gm)} \quad \text{in Bq·kg}^{-1}$$

where, w_s are the mass of the samples.

(c). The outdoor absorbed dose rates in air at 1 m above the ground surface (in nGy·h^{-1}) have been calculated by using the following formula (conversion factors) [7,12]

$$D_{outdoor} = (0.427 \ C_{Ra} + 0.66 \ C_{Th} + 0.0432 \ C_K)$$

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

(d). The indoor contribution is assumed to be 1.2 times higher than the outdoor dose which is [7,12]

$$D_{indoor} = D_{outdoor} \times 1.2 \ (\text{nGy·h}^{-1})$$

(e). The formulae for calculating the radium equivalent activity Ra_{eq} [11] and the representative level index $I_{\gamma r}$ [7,13-15] in the present research used are

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_K$$

$$I_{\gamma r} = (C_{Ra}/150 + C_{Th}/100 + C_K/1500)$$

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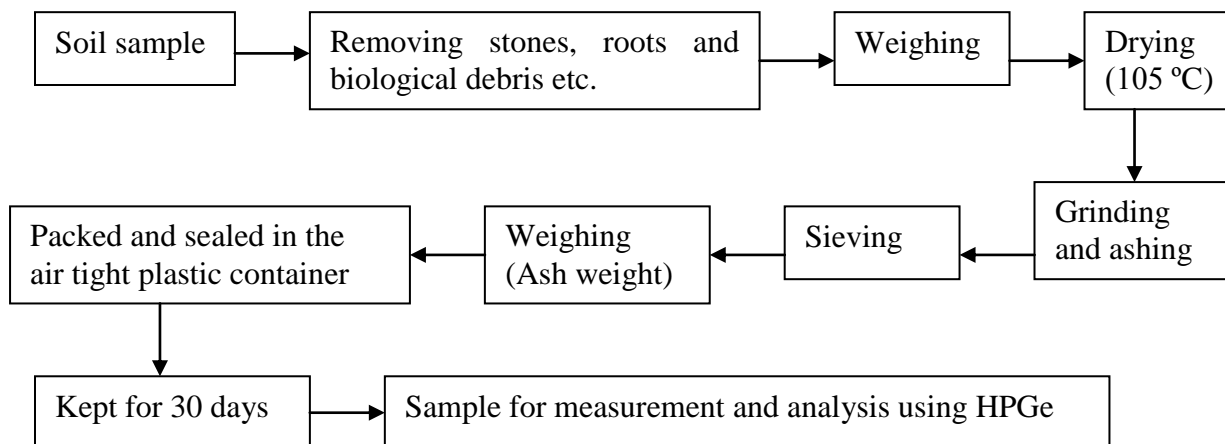

Fig. 2. Flow chart for illustrating the pre-treatment of the soil samples.

3. RESULTS

The mean specific activity concentrations of radionuclide's ^{232}Th, ^{226}Ra, and ^{40}K have been respectively found to be 52.87 ± 0.73, 23.08 ± 2.76, 426.88 ± 2.59 Bq·kg^{-1} surround the Magurchhera gas field and 57.48 ± 0.33, 22.38 ± 2.59, 369.54 ± 2.75 Bq·kg^{-1} surround the Haripur gas field explosion areas.

The activity concentrations of these radionuclide's have been respectively found to be ranged from 31.51 ± 0.66 to 88.99 ± 0.39, 12.00 ± 2.28 to 49.6 ± 2.96 and 260.04 ± 1.29 to 1094 ± 2.047 Bq·kg^{-1} for Magurchhera and 42.09 ± 0.35 to 75.12 ± 0.43, 15.25 ± 2.50 to 34.45 ± 2.66 and 213.47 ± 1.39 to 480.37 ± 2.34 Bq·kg^{-1} for Haripur gas field. The artificial radionuclide ^{137}Cs has been detected in most of the sample with an average of 1.65 ± 0.61 Bq·kg^{-1} around the Magurchhera and 1.13 ± 0.57 Bq·kg^{-1} around the Haripur gas field.

The comparative data of these radionuclides is given Table 1 and the graphical representation for Magurchhara and Haripur gas field are shown in Fig. 3A and 3B respectively. The activity concentrations of radionuclide's ^{232}Th and ^{40}K have been found to be higher and ^{226}Ra has been found to be lower than that of the world average [12] as well as worldwide value [4]. Again, the comparison of the activity concentration of radionuclide's ^{232}Th, ^{226}Ra and ^{40}K of present study with regional, other countries of the world and worldwide values [4] is given in Table 2.

The graphical comparison of the mean specific activity concentration of the radionuclide's of present results with the results of several countries of the world and

worldwide value are shown in Fig. 4. The mean value of radiation impact attributes namely radium equivalent activity, representative gamma level index, outdoor dose rate, indoor dose rate have been found to be 131.45 ± 4.05 Bq·kg^{-1}, 2.30 ± 0.03 Bq·kg^{-1}, 63.30 ± 1.82 nGy·h^{-1}, 75.95 ± 2.18 nGy·h^{-1} respectively for Magurchhara and 132.92 ± 3.28 Bq·kg^{-1}, 0.97 ± 0.02 Bq·kg^{-1}, 63.57 ± 1.44 nGy·h^{-1}, 76.28 ± 1.73 nGy·h^{-1} for Haripur gas field respectively.

All these values of these parameters are higher than those of the world average values [12] and their graphical representation are shown in Figs. 5a, 5b and 5c.
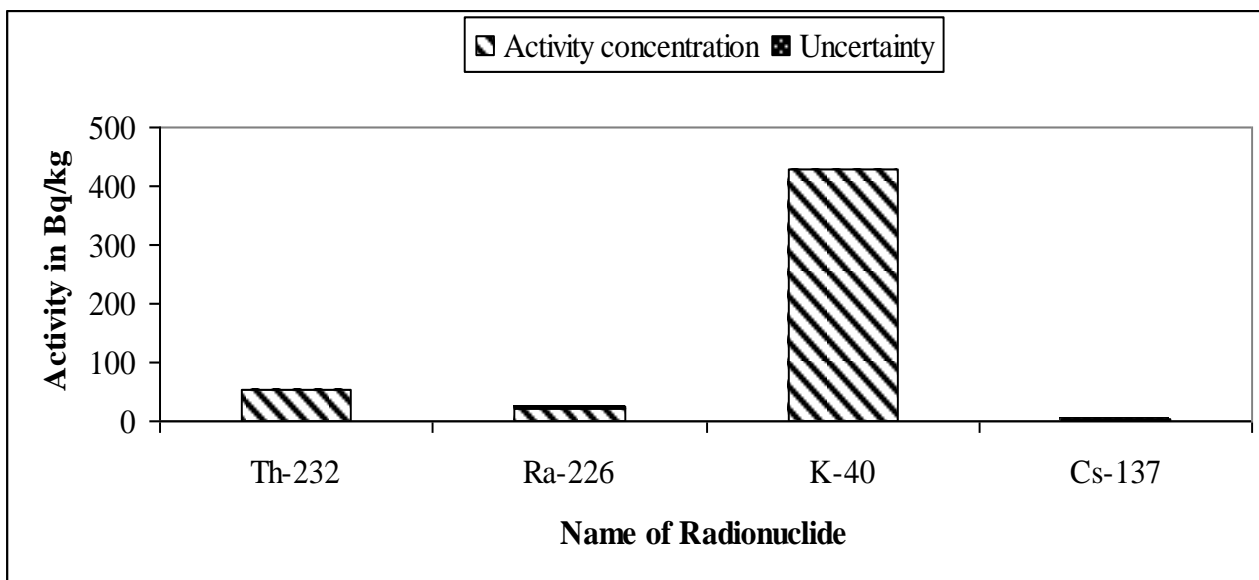

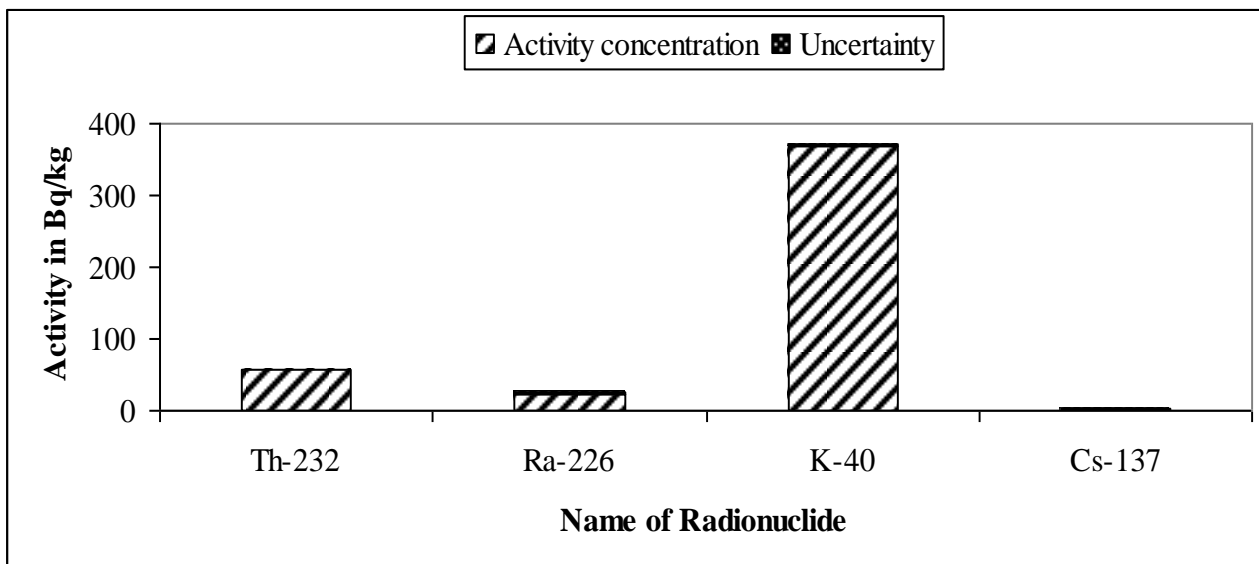**Fig. 3A.** Comparison of the activity concentration of radionuclide's in Magurchhera gas field area.

Fig. 3B. Comparison of the activity concentration of radionuclide's in Haripur gas field area.
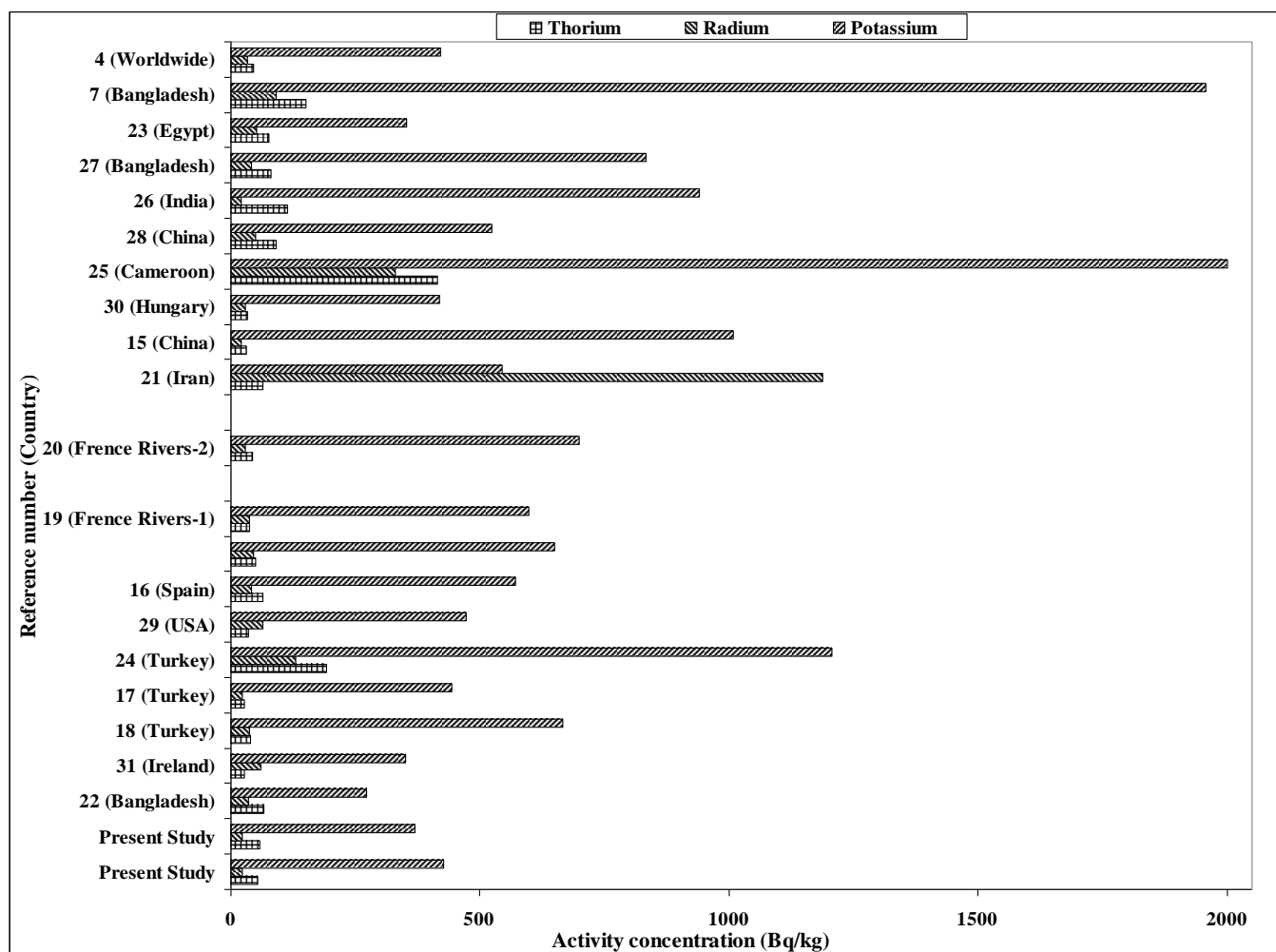
Fig. 4. Comparison of the present results with the results of various countries of the world.

Table 1. Comparative data of the mean activity concentration (in $Bq \cdot kg^{-1}$) of ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs of two study areas.

Radionuclie	Magurchhera gas field		Haripur gas field	
	Range	**Mean**	**Range**	**Mean**
^{232}Th	31.51 ± 0.66 to 88.99 ± 0.39	52.87 ± 0.73	42.09 ± 0.35 to 75.12 ± 0.43	57.48 ± 0.33
^{226}Ra	12.00 ± 2.28 to 49.6 ± 2.96	23.08 ± 2.76	15.25 ± 2.50 to 34.45 ± 2.66	22.38 ± 2.59
^{40}K	260.04 ± 1.219 to 1094.21 ± 2.04	426.88 ± 2.59	213.47 ± 1.39 to 480.37 ± 2.34	369.54 ± 2.75
^{137}Cs	N. D.[*] to 3.06 ± 0.45	1.65 ± 0.61	N. D. to 2.61 ± 0.51	1.13 ± 0.57

[*]N. D. = Not Detected

Table 2. Comparison of the mean activity concentration of ^{232}Th, ^{226}Ra and ^{40}K (in Bq·kg^{-1}) of present results with regional, other study areas of the world and worldwide values.

| Sl No.	References	Area	^{232}Th	^{226}Ra	^{40}K
1	Present Study	Magurchhara gas field	52.87 ± 0.73	23.08 ± 2.76	426.88 ± 2.59
		Haripur gas field	57.48 ± 0.33	22.38 ± 2.59	369.54 ± 2.75
2	22	River Sediments and Coastal Soils of	65.5 ± 12.2	35.9 ± 18.9	272 ± 35
3	31	Republic of Ireland	26	60	350
4	18	Kirklareli	40 ± 18	37 ± 17	667 ± 282
5	17	South of Marmara, Turkey	26.6 ± 16	22.5 ± 13	443 ± 190
6	24	Kestanbol	192	130.93	1207
7	29	Lousiana, USA	36 ± 12	64 ± 17	472 ± 223
8	16	River Tagus	63	42	572
		All soils	49	45	650
9	19	French Rivers-1	38	38	599
10	20	French Rivers-2	44	28	700
11	21	Western Mazandaran Province, Iran.	64.92 ± 162.26	1188.5 ± 7838.4	545.10 ± 139.42
12	15	Geological Park, China.	30.1	20.4	1009.5
13	30	Hungary	32.1	29.7	418
14	25	South Cameroon	414 ± 309	329 ± 491	2001 ± 521
15	28	China	89.8 ± 74	49.7 ± 35	524 ± 162
16	26	Kanyakumari District (India).	114 ± 97	20 ± 14	940 ± 742
17	27	Southern Districts of Bangladesh	81 ± 14	42 ± 7	833 ± 358
18	23	Nile River Sediments In Upper Egypt	76.2 ± 6.2	52 ± 7.3	351.9 ± 17.6
19	17	Northern Districts of Bangladesh	151 ± 24	91 ± 10	1958 ± 400
20	4	Worldwide	45 (11–64)	33 (17–60)	420 (140–850)
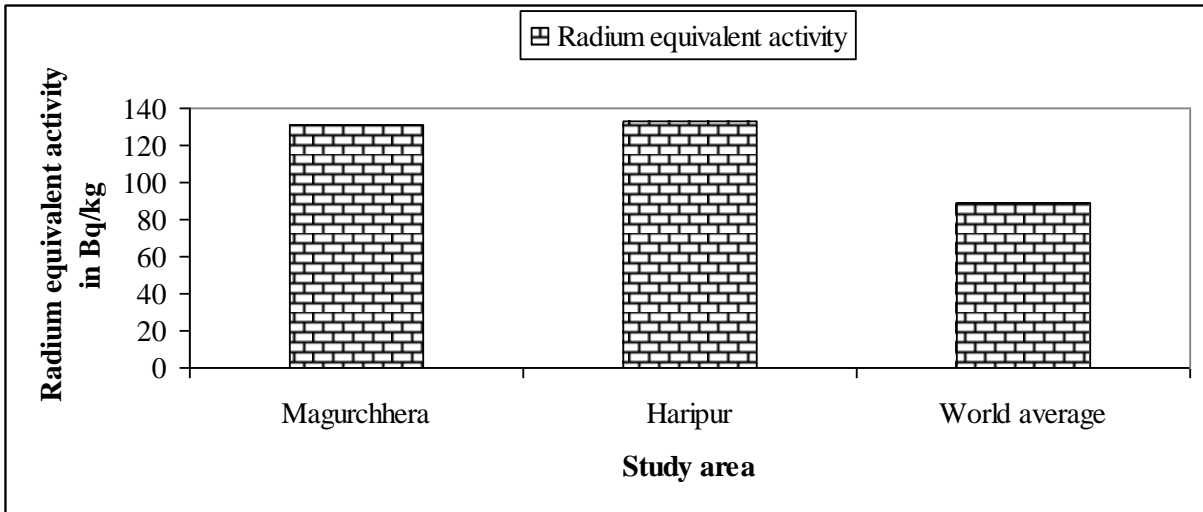
Fig. 5a. Comparison of the mean radium equivalent activity with world average value.
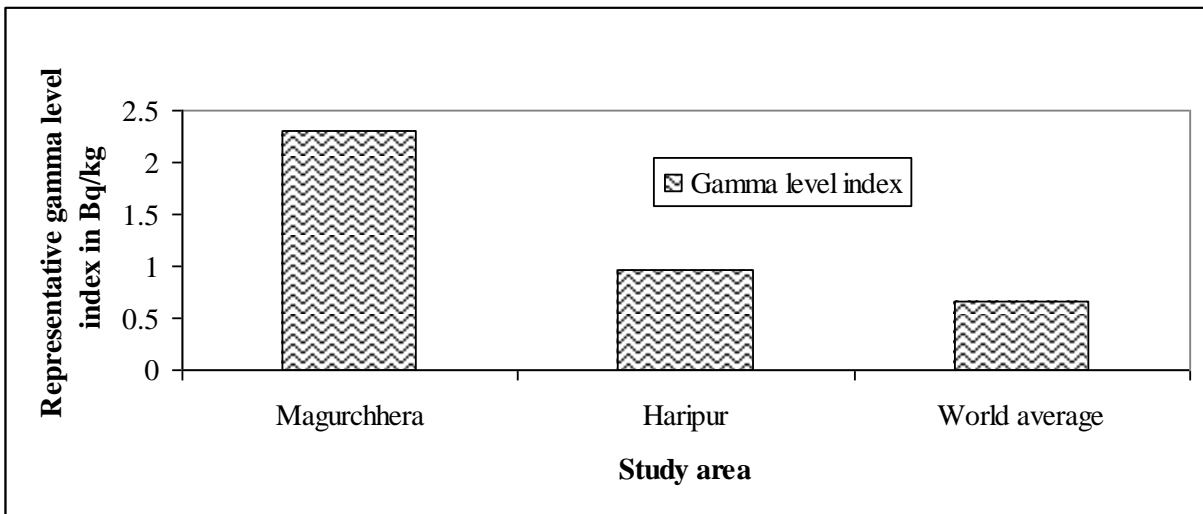

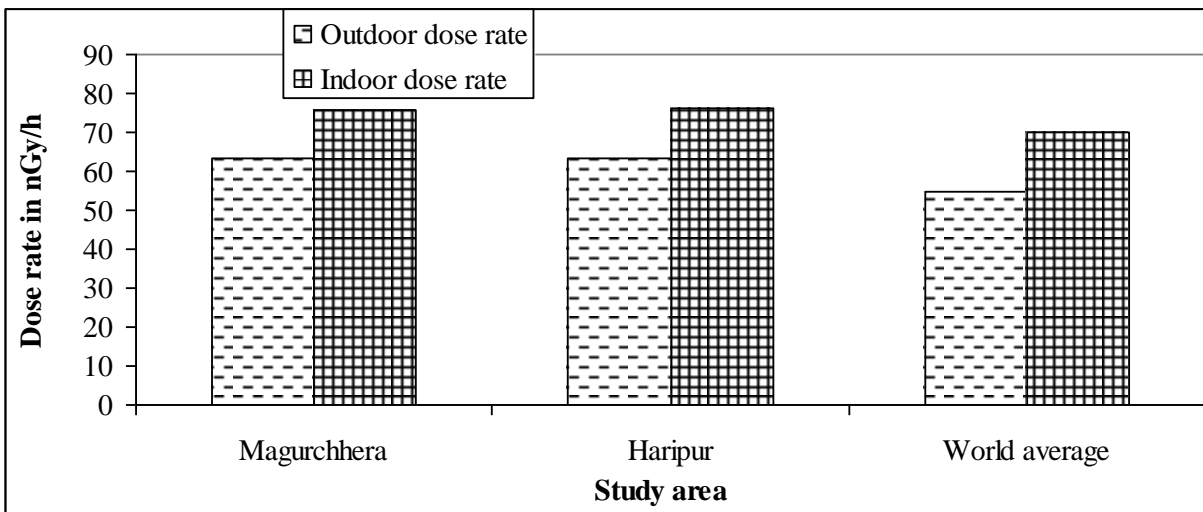**Fig. 5b.** Comparison of the mean gamma level index with world average value.

Fig. 5c. Comparison of the mean outdoor and indoor dose rate with world average value.

4. DISCUSSION

It has been found that the concentrations of Th and U-series' radionuclide's (^{232}Th, ^{226}Ra) and ^{40}K in the Magurchhera and Haripur gas field areas are relatively moderate in concentration than those of the other reported areas of the world. The mean activity concentration of ^{232}Th in the these gas well areas has been found to be higher than those of the Turkey [17], Kirklareli [18], Ireland [31], Louisiana-USA [29], French rivers-1 [19], French rivers-2 [20], China [15] and Hungary [30],. It is about to 1.6 times and 2.11 times higher for Magurchhara and 1.74 times and 2.29 times higher for Haripur area than the worldwide value [4] (Table 3) and world average [12] (Table 3) respectively. However, the mean activity concentration of ^{232}Th has been found to be lower than those of the Kestanbol [24], Iran [21], regional study [22], Egypt [23], South Cameroon [25], India [26], Southern Districts of Bangladesh [27], China [28] and Northern Districts of Bangladesh 7 (Table 2).

The mean specific radioactivity level of ^{226}Ra in the study areas has been found to be higher than those of the Turkey [17], India [26], China [15], although it has been found to be lower than those of the Kestanbol [24], Kirklareli [18], Spain [16], French rivers-1 [19], French rivers-2 [20], Ireland [31], Iran [21], regional study [22], Egypt [23], Southern Districts of Bangladesh [27], South Cameroon [25], Louisiana-USA [29] and Northern Districts of Bangladesh [7] (Table 2). It has been found to be about 0.51 times and 0.92 times for Magurchhera and 0.50 times and 0.89 times for Haripur than the worldwide value [4] (Table 3) and world average [12] (Table 3) values respectively.

Table 3. Comparison of overall results of the present study with corresponding world average values, worldwide value and regional study.

Study Area	Finding Attributes	Results of Present Study (average ± 1σ)	Regional Study [22]	World Average [12]	World Wide [4]	Ratio: present average/ world average (Wide)
Magurchhara gas field	^{232}Th (Bq·kg^{-1})	52.87 ± 0.73	35.9 ± 18.9	25	33	2.11 (1.6)
	^{226}Ra (Bq·kg^{-1})	23.08 ± 2.76	65.5 ± 12.2	25	45	0.92 (0.51)
	^{40}K (Bq·kg^{-1})	426.88 ± 2.59	272 ± 35	370	420	1.15 (1.02)
	I_{yr} (Bq·kg^{-1})	2.30 ± 0.03	1.08 ± 0.27	0.66	-	3.48
	Ra_{eq} (Bq·kg^{-1})	131.45 ± 4.05	151 ± 39	89	-	1.48
	Outdoor Dose (nGy·h^{-1})	63.30 ± 1.82	70.78 ± 18.26	55	-	1.15
	Indoor Dose (nGy·h^{-1})	75.95 ± 2.18	-	70	-	1.09
Haripur gas field	^{232}Th (Bq·kg^{-1})	57.48 ± 0.33	35.9 ± 18.9	25	33	2.29 (1.74)
	^{226}Ra (Bq·kg^{-1})	22.38 ± 2.59	65.5 ± 12.2	25	45	0.89 (0.50)
	^{40}K (Bq·kg^{-1})	369.54 ± 2.75	272 ± 35	370	420	1.0 (0.88)
	I_{yr} (Bq·kg^{-1})	0.94 ± 0.02	1.08 ± 0.27	0.66	-	1.42
	Ra_{eq} (Bq·kg^{-1})	132.92 ± 3.28	151 ± 39	89	-	1.49
	Outdoor Dose (nGy·h^{-1})	63.57 ± 1.44	70.78 ± 18.26	55	-	1.16
	Indoor Dose (nGy·h^{-1})	76.28 ± 1.73	-	70	-	1.09

The mean specific radioactivity level of ^{40}K in the soil samples has been found to be higher than those of the regional study [22], Ireland [31], Hungary [30], Egypt [23], but the activity concentration of ^{40}K has been found to be lower than those of the Turkey [17], Kirklareli [18], Spain [16], French rivers-1 [19], French rivers-2 [20], Iran [21], Southern Districts of Bangladesh [27], Kestanbol [24], China [15], South Cameroon [25], Northern Districts of Bangladesh [7], and India [26] (Table 2). The specific activity of ^{40}K has been found to be about 1.02 times and 1.15 times higher for Magurchhera area than the worldwide value [4] (Table 3) and world average [12] (Table 3) value respectively; similar results have also been found for Haripur area. Although, the artificially produced anthropogenic radionuclide ^{137}Cs has been detected in most of the samples in low level, further study is necessary for confirming that's the anthropogenic radionuclide remained in these gas well areas. The mean values of the representative level index I_{yr}, radium equivalent activity Ra_{eq}, outdoor dose rate and indoor dose rate due to the radioactivity in the Magurchhera and Haripur gas well areas have been found to be higher than those of the world average [12] (Table 3) values.

5. CONCLUSION

The average activity of ^{226}Ra in the study areas is comparable with the average value, but it is higher than the world average value in case of ^{232}Th. In Magurchhera area, the mean activity of ^{40}K is higher than the world average value [12], but for Haripur area it coincides with the world average value. The mean activity of ^{137}Cs has been found very little in the study areas. So it may be concluded that the Magurchhera gas field as well as Haripur gas field area has not been contaminated by ^{137}Cs due to the accident occurred there. The values of the dose rates, radium equivalent activity and gamma level index recommend that the environment of the study areas is safe for the public health in respect of radioactivity.

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