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Natural Radioactivity of ²³⁸U, ²³²Th, ⁴⁰K in Selected Ground water of Bangladesh and Assessment of Radiological Hazards with Statistical Analyses

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Abstract: Identification and quantification of gamma emitting radionuclides such as thorium, uranium etc. with long halflife plays a key role in environmental characterization and radiation protection. In order to do, ground waters from different places of Bangladesh have been collected and their gamma radioactivity was investigated. The gamma activity of radionuclides ²³⁸U, ²³²Th and ⁴⁰K in ground water samples were determined by using HPGe gamma spectroscopy. The average gamma activity concentration of ²³⁸U is 2.59 Bq/L, ²³²Th is 2.45 Bq/L and ⁴⁰K is 32.00 Bq/L. In this present study, the radiological hazard parameters due to the radioactivity of natural occurring radionuclides (NOR), such as, Radium equivalent activity (Raeq), Representative level index (RLI), Activity utilization index (AUI), Absorbed dose (D), Annual Effective Dose equivalent (AEDE), Annual gonadal dose equivalent (AGDE), External hazard index (Hex), Internal hazard index (Hin) and Excess lifetime cancer risk (ELCR) are assessed from the ground water samples. The calculated average value of AUI and AGDE is 0.056 and 28.28 μ Sv/y, respectively. Again, the average values of RLI and ELCR are 0.063 and 0.017 mSv reapectively. The Th/U ratio was calculated to assume preferable oxidation sate of Th and U. The Pearson correlation analysis and cluster analysis are employed to analyses the data and identify the correlation between the radiological hazard parameters with the natural occurring radionuclides.

Keywords: Groundwater, Natural occurring radionuclides, Radioactivity, Gamma spectrometry system, Radiological risk assessment.

1 Introduction

Radionuclides are the sources of radioactivity and emit nuclear radiations which have become a part of our daily lives. The most common forms of ionizing radiation are alpha particles, beta particles and gamma rays [1]. There is no where on Earth that one can get away from Natural Radioactivity [2]. Extraterrestrial radiations originate in outer space as primary cosmic rays and reach the atmosphere. Terrestrial radiations are emitted from natural radionuclides present in varying amounts in all types of solids, rocks, air, water and other environmental materials around us. The radionuclides result from three decay series namely, Uranium, Thorium and Actinium. Uranium is a naturally occurring radioactive element, widely distributed in nature, consists of the isotopes ²³⁴U, ²³⁵U and ²³⁸U, with a mass ratio of 0.0054: 0.711: 99.2836% [3]. The half-life of 238 U is about 4.47 billion years and that of 235 U is 704 million years [3]. Uranium occurs in low concentrations in much natural water, sea water has an average uranium content of about 2 µg/L and most fresh waters have concentrations below 10 µg/L. Some groundwater sources have uranium concentrations as high as several milligrams per liter [4].

Water plays a big and diverse role in the world. Processed or not, water is used in industry and commercial sector, for irrigation, sanitation and primarily for supplying the population with drinking water and water for household needs. Radiological control of water is necessary due to its importance for human life and the need for minimum exposure to radiation. Therefore, maximum permissible concentration limits of radio-nuclide activity in drinking water have been prescribed by the World Health Organization [3]. Gamma spectrometry is a radiometric



technique, which means that the estimation of the isotopic activity in a sample relies on the detection of the products of the radioactive decay of that isotope. In this case gamma radiation that have to be detected. The number of gamma radiation detected per second are directly related to the isotope activity, measured in disintegration per second [5].

2 Materials and Method

2.1 Site and Sample Collection

In order to measure the natural and artificial radioactivity in groundwater, ten groundwater samples were collected randomly from different locations of Bangladesh. The water sample was collected from central ground water supplies line from different location so that it can represent the respective area. The average depth of sampling site was 1200-1500 ft. The samples were kept into previously cleaned 2 L capacity plastic bottle using manual procedure. They were appropriately coded from 1 to 10 and transferred to Environmental Radioactivity Monitoring Laboratory of

Table 1: Groundwater samples with sample code number and sample locations.

| Sample Code No. | Sample Locations |
|-----------------|------------------|
| Groundwater-1 | Dhaka University |
| Groundwater-2 | Tangail |
| Groundwater-3 | Narayanganj |
| Groundwater-4 | Narsingdi |
| Groundwater-5 | Savar |
| Groundwater-6 | Mymensingh |
| Groundwater-7 | Gazipur |
| Groundwater-8 | Kustia |
| Groundwater-9 | Dhaka Medical |
| Groundwater-10 | Kalabagan |

Health Physics Division at Atomic Energy. The lists of the collected samples are given in following Table 1.

2.2 Sample preparation for Gamma Counting

Ten 1.0 L capacity Maryline beakers were washed with distilled water and left to dry to avoid sample contamination. About 1.0 L of each sample was poured into a Maryline beaker. 10 mL concentrated HNO₃ was added to each water sample to avoid the collection of organic materials and changes in the oxidation state of the ions present in the samples. Subsequently, the water samples were slowly evaporated by water bath treatment at 105° C and reduced up to 250 mL approximately and each of the samples was transferred to cylindrical plastic container. The containers were then labeled properly and sealed tightly, rapped with thick vinyl tapes around their screw necks for gamma detection in HPGe detector.

2.3 Measurement of Gamma Activity

To qualitatively identify the contents of radionuclides in water sample and to quantitatively determine their activities, all prepared samples were measured by means of gammaray spectrometry system using ORTEC high purity Gedetector for 10000 sec. The volume of the detector was 83.49 cm^3 and the distance between detector and sample was 0.3cm. The equal counting time for background and sample measurement was chosen to minimize the uncertainty in the net counts. The spectrum of each sample was analyzed and the identification of unknown radionuclides was carried out by considering their peak centroid energies. The centroid energies of the peaks from the spectrum were compared with the reference gamma-ray energies obtained from the literature. The radionuclides contained in the samples were identified and the areas under the peaks were used to determine the activity concentrations of each nuclide. The number of counts under the full-energy peak areas (corrected for background peak areas), the counting time, the absolute full-energy peak efficiency for the energy of interest and the gamma-ray emission probability corresponding to the peak energy are used for the calculation of the activity concentration of a particular radionuclide in the measured samples.

The activity concentrations of ²³⁸U and ²³²Th were determined from the average concentration of nuclides [²¹⁴Pb (295.2 keV), ²¹⁴Pb (351.9 keV), ²¹⁴Bi (609.3 keV) and ²¹⁴Bi (1120.2 keV)] and [²¹²Pb(238.6 keV), ²⁰⁸Tl(583.1 keV), and ²²⁸Ac(911.2 keV), ²²⁸Ac(968.9 keV)] respectively. The activity concentration of ²³⁸U was measured by assuming secular equilibrium between ²³⁸U and ²²⁶Ra. The activity concentrations of ⁴⁰K were determined directly by measurement of the gamma-ray transitions at 1460.8 keV [6]. The specific activity, in terms of the activity concentration, is defined as the activity per unit mass of the sample. The specific activity of individual radionuclides in water samples is given by the following equation:

$$A = \frac{N \times 100 \times 100}{P_{\gamma} \times \varepsilon \times w}$$

Where, N = Net counts per second (cps) = (Sample cps) – (Background cps), P_{γ} = Transition probability of gamma ray or Branching ratio, ε = Efficiency in percent, W = Weight of the sample in litre.

In the present study, 10 transitions or gamma ray lines of the radionuclide 152 Eu mixed with 1 M HCl were used to perform the efficiency calibration.

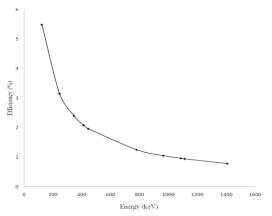


Fig. 1: Efficiency curve of the detector using 10 transitions in ¹⁵²Eu radionuclide.

Each count rate includes standard deviation and the standard deviation of the net count rate can be expressed as,

$$\sigma = \pm \sqrt{(A_s/T_s + A_b/T_b)}$$

where, σ = standard deviation, A_s = sample count rate in cps, A_b = background count rate in cps, T_s = sample count time, and T_b = background count time.

2.4 Detection Limits

Detection limits is a term used to express the detection capability of a measurement system under certain conditions. An estimate for the lowest amount of activity of a specific gamma-emitting radionuclide that can be detected at the time of measurement can be calculated from several different expressions. A generally accepted expression for the estimate of the detection limits, which is frequently referred to as the lower limit of detection (LLD) and which contains a preselected risk of 5 % of concluding falsely that activity is present and a 95 % degree of confidence for detecting the presence of activity, is as follows.

$$LLD = \frac{4.66 \times S_{b}}{\epsilon P_{\gamma}}$$

Where, S_b is the estimated standard error of the net count rate, ϵ is the counting efficiency of the specific nuclide's energy; number <1, P_{γ} is the absolute transition probability by gamma decay through the selected energy, number <1 [7].

2.5 Multivariate Statistical Analysis

Principal component analysis (PCA) and cluster analysis reduces the dimen-sionality of data by a linear combination of original data to generate new latent variables which are orthogonal and uncorrelated to each other. It extracts the eigenvalues and eigenvectors from the covariance matrix of original variables [8]. Pearson's correlation matrix is used to identify the relationship among the pairs of parameters. The experimental groundwater data were subjected to statistical analysis using IBM SPSS software (version 20).

3 Results and Discussion

3.1 Activity Concentration for Gamma of Selected Groundwater Samples

The activity concentrations of $^{238}\text{U},~^{232}\text{Th}$ and ^{40}K in the water samples, collected from different parts of the studied areas, are presented in table 2. The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K for the water samples are shown graphically in Fig. 2, 3 & 4 respectively. In the present study the average activity concentration of ²³⁸U, ²³²Th and ⁴⁰K is 2.59 Bq/L, 2.45 Bq/L and 32.00 Bq/L respectively. The activity of ⁴⁰K is higher than the activity of ²³²Th and ²³⁸U. The higher activity ⁴⁰K found in groundwater-5 that was collected from Savar (Fig.2). The higher activity concentration of gamma emitter, ²³⁸U is found in groundwater-10 that was collected from Kalabagan and lowest value found in groundwater-6 that was collected Mymensingh (Fig.3). The from higher activity ²³²Th found in concentration of gamma emitter, groundwater-1 collected from Dhaka university area and lower value found in ground water-7 that was collected from Gazipur (Fig. 4). The water samples follows the average activity order $^{232}\rm{Th}$ < $^{238}\rm{U}$ < $^{40}\rm{K}.$ So the distribution of ²³²Th, ²³⁸U and ⁴⁰K in water samples are not uniform. Progenies of the thorium decay series are not found in significant amount in aquatic media because ²³²Th is essentially insoluble in water [9].

It was reported that average activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in groundwater of Kurigram district of Bangladesh is 8.9±3.6 Bq/L, 3.6±2.4 Bq/L, 52±22 Bq/L respectively [10] which is higher than the values of our studied samples. The activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in drinking water of different region of Bangladesh was measured in 1998 and the measured values were 0.157 Bq/L, 0.250 Bq/L and 9.00 Bq/L respectively [11]. The activity concentration found in our present study has exceeded the previous reported value. The activity concentration of ²³⁸U, ²³²Th in our samples are compared with the UNSCEAR reference mean value which are 1 mBq/L, 0.05 mBq/L in United States respectively [12]. The activity concentrations of ²³⁸U, ²³²Th in our studied samples are much more than the UNSCEAR references mean value in United States.



Uranium possesses several oxidation states. Among them tetravalent state (U^{4+}) is insoluble that is fixed under reducing environment whereas hexavalent state (U^{6+}) which is soluble and mobilized in aquatic medium. In the contrary, tetravalent thorium (Th^{4+}) is insoluble and geochemically which is associated with uranium. The value of Th/U ratio can predict the depositional environment of uranium and thorium. When the ratio was calculated to be

less than two (Th/U<2), uranium deposited into U4+ state under probable reducing environment and Th/U ratio value greater than seven (Th/U>7) indicate mobilization of uranium with U⁶⁺ state through oxidizing environment [13]. Our calculated values of Th/U ratio are less than two that indicates that presumably both thorium (Th) and uranium (U) exist in water with +4 oxidation state through reducing environment.

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|------------------------|--------------------|-------|-------------|----------|----------------------|
| Table 2: Activity | y concentrations (| Bq/L) | of radionuc | lides in | groundwater samples. |

| Name of Sample | Activity of ²³⁸ U | Activity of ²³² Th | Activity of ⁴⁰ K | Th/U |
|----------------|------------------------------|-------------------------------|-----------------------------|------|
| Groundwater-1 | 2.67±0.62 | 3.86±0.23 | 38.47±0.70 | 1.45 |
| Groundwater-2 | 2.70±0.56 | 2.07±0.79 | 34.75±0.60 | 0.77 |
| Groundwater-3 | 2.28±1.01 | 2.31±0.84 | 26.72±0.60 | 1.01 |
| Groundwater-4 | 2.46±1.25 | 1.79±0.30 | 35.63±0.70 | 0.73 |
| Groundwater-5 | 3.59±1.32 | 2.72±0.49 | 48.99±0.70 | 0.76 |
| Groundwater-6 | 1.67±0.59 | 3.05±0.65 | 31.18±0.69 | 1.83 |
| Groundwater-7 | 1.89±0.70 | 1.44±0.33 | 17.81±0.70 | 0.76 |
| Groundwater-8 | 2.07±1.10 | 2.82±1.10 | 37.86±0.70 | 1.36 |
| Groundwater-9 | 2.08±1.12 | 2.02±0.81 | 22.27±0.60 | 0.97 |
| Groundwater-10 | 4.48±1.58 | 2.39±0.96 | 26.28±0.70 | 0.53 |

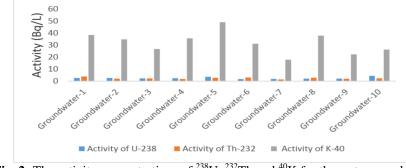
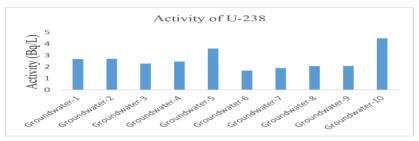
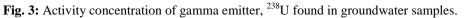


Fig. 2: The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K for the water samples.





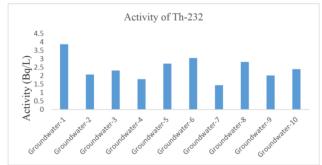


Fig. 4: Activity concentration of gamma emitter, ²³²Th found in groundwater samples.

Table 3: Average activity concentrations of 238 U, 232 Th and 40 K in groundwater for different countries with that of the present work.

| Country | ²³⁸ U (Bq/L) | ²³² Th (Bq/L) | ⁴⁰ K (Bq/L) | References |
|--------------|-------------------------|--------------------------|------------------------|---------------|
| USA | 0.077 | - | - | [12] |
| China | 0.70 | 0.012 | - | [12] |
| France | 0.93 | 0.0042 | - | [12] |
| Italy | 0.13 | - | - | [14] |
| Greece | 29.346 | - | | [15] |
| Finland | 150.0 | - | - | [12] |
| Brazil | 0.80 | 0.0003 | - | [16] |
| Iraq | 0.933 | 0.737 | 24.45 | [17] |
| Egypt | - | 0.13 | 5.29 | [18] |
| Iran | - | 2.70 | 4.23 | [19] |
| Saudi Arabia | 0.017-0.088 | 0.020-0.102 | 0.41-1.0 | [19] |
| Switzerland | 1.00 | - | - | [12] |
| Bangladesh | 2.59 | 2.45 | 32.00 | Present study |

3.2 Radium Equivalent (Ra_{eq})

Radium equivalent activity index in Bq/L is the universally accepted index for analysing the radiation exposure created by the Natural occurring radionuclides. In order to compare their combined radiological effect, a common index Radium equivalent (Ra_{eq}) was used. The radium equivalent describe the gamma output from different mixtures of ²³⁸U (²²⁶Ra), ²³²Th, ⁴⁰K in water samples from the study area and it is calculated by the formula [20],

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K$$

Where, A_U , A_{Th} and A_K are the activity concentrations of 238 U, 232 Th, and 40 K in Bq/L respectively. The calculated

radium equivalent activity (Ra_{eq}) are given in table 4 and it ranges from 5.32 Bq/L to 11.25 Bq/L with a mean value of 8.55 Bq/L. The values from all the sampling sites are very low when compared to the maximum permissible radium equivalent index of 370 Bq/L [21].

4 Assessments of Radiological Hazards

4.1 Annual Effective Dose of Uranium

The annual effective dose $(\mu Sv/y)$ due to the ingestion of uranium through drinking water was calculated as the product of activity concentration (Bq/L) of the element in water, the annual intake of water (L/y), and the dose conversion factor (Sv/Bq). In the present study, the annual intake of water was taken as 730 L/y, at the rate of 2 L/day

[22] and the dose conversion factor as 4.62×10^{-8} Sv/Bq obtained as the average of the dose coefficients for 234 U, 235 U, and 238 Uisotopes based on ICRP publications [23].

Annual effective Dose of Uranium = $A_U \times 730 \times 4.62 \times 10^{-8}$

The calculated value of annual effective dose of uranium ranges from 56.32 μ Sv/y to 151.09 μ Sv/y with an average value 87.32 μ Sv/y which is less than the annual effective dose value 100 μ Sv/y referred by WHO.

4.2 Absorbed Dose Rate (D)

The absorbed dose of radiation is the energy imparted per unit mass of the irradiated material. The measured activity concentrations of 238 U, 232 Th, and 40 K are converted into doses by applying the conversion factors 0.462, 0.604 and 0.0417 for 238 U, 232 Th, and 40 K respectively [12]. The total absorbed dose rate (D) in nGy/h is calculated using the following formula:

 $D (nGy/h) = 0.462 A_U + 0.604 A_{Th} + 0.0417 A_K$

Where, A_U , A_{Th} and A_K are the activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in Bq/L respectively. The calculated absorbed dose rates (D) are given in table 4 and it ranges from 2.51 nGy/h to 5.23 nGy/h with a mean value of 4.05 nGy/h. In our present study reveal that all the absorbed dose rate values in the study area are very lower than the maximum permissible level of 55 nGy/h [12].

4.3 Annual Effective Dose Equivalent (AEDE)

Irradiation of the human body from external sources is mainly by gamma radiation from radio-nuclides in the ²³⁸U and ²³²Th series and from ⁴⁰K present in all soils and waters [12]. Conversion coefficient from absorbed dose in air to effective dose and the indoor and outdoor occupancy factors are required to estimate annual effective doses. According to the UNSCEAR 2000 report, the conversion coefficient from absorbed dose in air to effective dose received by adults is 0.7 Sv/Gy and the occupancy factor for outdoor is 0.2 i.e. the fraction of time spent outdoors is 0.2. The annual effective dose equivalent (AEDE) in outdoor air is determined as follows [12]:

AEDE (outdoors) = 5.23 nGy/h × 8760 h × 0.2 × 0.7 Sv/Gy

The calculated outdoor AEDE values are given in table 4. The outdoor AEDE values are varying from 0.0031 to 0.0066 mSv/y . The resulting worldwide average of the annual effective dose is 0.48 mSv/y. For children and infants, the values are about 10% and 30% higher, in direct proportion to an increase in the value of the conversion coefficient from absorbed dose in air to effective dose [12].

4.4 Annual Gonadal Dose Equivalent (AGDE) The activity bone marrow and the bone surface cells are considered as the organs of interest by UNSCEAR (1988). Hence, the annual gonadal dose equivalent (AGDE) due to the specific activities of ²³⁸U, ²³²Th, and ⁴⁰K is calculated using the following

AGDE
$$(\mu Sv/y) = 3.09A_U + 4.18A_{Th} + 0.314A_K$$

The AGDE values calculated in this study are given in table 4 and it range from 17.45 to 37.85 μ Sv/y with the average value 28.28 μ Sv/y which clearly indicates that the AGDE values of all soil samples in the present study are below the world average value 300 μ Sv/y [24].

4.5 Hazard indices (Hex and Hin)

The hazardous effects of gamma radiation due to the specified radioactive elements in environmental samples are assessed by calculating the following two hazard indices using the below given relations [25]:

$$\begin{split} H_{ex} &= (A_{U}/370 \text{ Bq/L}) + (A_{Th}/259 \text{ Bq/L}) + (A_{K}/4810 \text{ Bq/L}) \\ H_{in} &= (A_{U}/185 \text{ Bq/L}) + (A_{Th}/259 \text{ Bq/L}) + (A_{K}/4810 \text{ Bq/L}) \end{split}$$

Where, A_U , A_{Th} and A_K are the activity concentrations of 238 U, 232 Th, and 40 K in Bq/L respectively. The internal hazard index (H_{in}) is used to control the internal exposure to radon and its short-lived products which are also dangerous to the respiratory organs [26]. The calculated hazard indices are listed in table 4. The H_{ex} and H_{In} values vary from 0.014 to 0.030 and 0.019 to 0.040 respectively. The reference value by UNSCEAR (2000) report for the hazard indices is less than unity. It is clear from Table 4 that the hazard indices calculated in this study are well below the reference value.

4.6 Excess Lifetime Cancer Risk (ELCR)

Low doses of ionising radiation with long exposure time can increase the risk of cancer this risk becomes clear at doses above 100 mSv. The risk of cancer increases as the dose of radiation increases. Exposure to one Sievert of radiation spread out over time is estimated to increase the lifetime risk of fatal cancer. Excess lifetime cancer risk (ELCR) is calculated using the below given formula [27] and listed in Table 4.

$$ELCR = AEDE \times DL \times RF$$

Where, AEDE, DL, and RF are annual effective dose equivalent, duration of life(70 years) and risk factor (0.05 Sv^{-1}), respectively. For stochastic effects, ICRP 60 uses values of 0.05 for the public [28]. The calculated value of ELCR ranges from 0.011 mSv to 0.023 mSv. The world average value for ELCR is 0.29 mSv [12].The ELCR values from water samples in the present study are lower to the world average value.

4.7 Representative Level Index (RLI)

Representative level index is the level of gamma

radioactivity associated with different concentrations of some specific radioactive elements which can be measured using [29] the following formula:

$$RLI = (A_U / 150) + (A_{Th} / 100) + (A_K / 1500)$$

Where, A_U , A_{Th} and A_K are the activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in Bq/L, respectively. The RLI values measured for all the water samples are given in table 4 and it varies from 0.039 to 0.084 with the average of 0.063. The maximum limit for RLI is 1 [29] and when it is compared with the present study, RLI of most of the samples are lower to the maximum limit.

4.8 Activity Utilization Index (AUI)

The dose rates in air from different combinations of 238 U, 232 Th, and 40 K (Bq/L) in water samples and by applying the suitable conversion factors, activity utilization index (AUI) is calculated from the following relation [30]:

$$AUI = (A_U/50)f_U + (A_{Th}/50)f_{Th} + (A_K/500)f_K$$

Where, A_U , A_{Th} and A_K are the activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in Bq/L in water samples, respectively, and f_U (0.462), f_{Th} (0.604) and f_K (0.042) are the respective fractional contributions from the actual activities of ²³⁸U, ²³²Th, and ⁴⁰K to the total dose rate in air [26]. Calculated

values of AUI are provided in table 4 and its range from 0.036 to 0.075. This value exhibits that AUI <2, which means that annual effective dose <0.3 mSv/y [30].

5 Statistical Analyses of Data

5.1 Descriptive statistics

Descriptive statistics of the radionuclides of the ground waters are given following Table 5. The standard deviation was largest for ⁴⁰K and smallest for ²³²Th. In statistical analyses skewness is a measure of symmetry whereas kurtosis is a measure of whether the data are heavy-tailed or light-tailed relative to a normal distribution. The activity concentrations of all of the radionuclides in this study have positive kurtosis values, which indicate that the distributions are peaked in nature. In the present study, the distributions associated with ²³²Th and ⁴⁰K radionuclides have positive skewness values, indicating asymmetric distribution with extended tail that are more positive, and ²³⁸U radionuclide have negative skewness values, indicating flat distributions asymmetric distribution with extended tail that are more negative. From histograms it's clear that U & Th were distributed in normal distribution. On the contrary K shows few degree of multi-modality.

| Samples No. | Ra _{eq} | RLI | D | AUI | AGDE | AEDE | Hex | Hin | ELCF |
|----------------|------------------|-------|---------|-------|--------------|---------|-------|-------|-------|
| | (Bq/L) | | (nGy/h) | | $(\mu Sv/y)$ | (mSv/y) | | | (mSv |
| Groundwater-1 | 11.15 | 0.082 | 5.23 | 0.075 | 36.46 | 0.0064 | 0.030 | 0.037 | 0.023 |
| Groundwater-2 | 8.34 | 0.062 | 3.98 | 0.053 | 27.91 | 0.0049 | 0.023 | 0.030 | 0.01′ |
| Groundwater-3 | 7.64 | 0.056 | 3.60 | 0.051 | 25.09 | 0.0044 | 0.021 | 0.027 | 0.010 |
| Groundwater-4 | 7.77 | 0.058 | 3.74 | 0.047 | 26.29 | 0.0046 | 0.021 | 0.028 | 0.01 |
| Groundwater-5 | 11.25 | 0.084 | 5.39 | 0.070 | 37.85 | 0.0066 | 0.030 | 0.040 | 0.023 |
| Groundwater-6 | 8.43 | 0.062 | 3.97 | 0.055 | 27.70 | 0.0049 | 0.023 | 0.027 | 0.017 |
| Groundwater-7 | 5.32 | 0.039 | 2.51 | 0.036 | 17.45 | 0.0031 | 0.014 | 0.019 | 0.01 |
| Groundwater-8 | 9.02 | 0.067 | 4.29 | 0.056 | 30.07 | 0.0053 | 0.024 | 0.030 | 0.013 |
| Groundwater-9 | 6.68 | 0.049 | 3.14 | 0.045 | 21.86 | 0.0039 | 0.018 | 0.024 | 0.014 |
| Groundwater-10 | 9.92 | 0.071 | 4.65 | 0.072 | 32.09 | 0.0057 | 0.027 | 0.039 | 0.020 |
| Average | 7.71 | 0.063 | 4.05 | 0.056 | 28.28 | .0050 | 0.023 | 0.030 | 0.01 |

 Table 4: Radiological hazard parameters in different groundwater samples.



| Variables | ²³⁸ U | ²³² Th | ⁴⁰ K |
|----------------|------------------|-------------------|-----------------|
| Mean | 2.59 | 2.45 | 32.00 |
| Std. Deviation | 0.93 | 0.69 | 9.08 |
| Variance | 0.88 | 0.48 | 82.58 |
| Skewness | -0.424 | 0.688 | 0.246 |
| Kurtosis | 0.308 | 0.680 | 0.072 |
| Range | 3.24 | 2.42 | 31.18 |
| Maximum | 4.48 | 3.86 | 48.99 |
| Minimum | 1.67 | 1.44 | 17.81 |

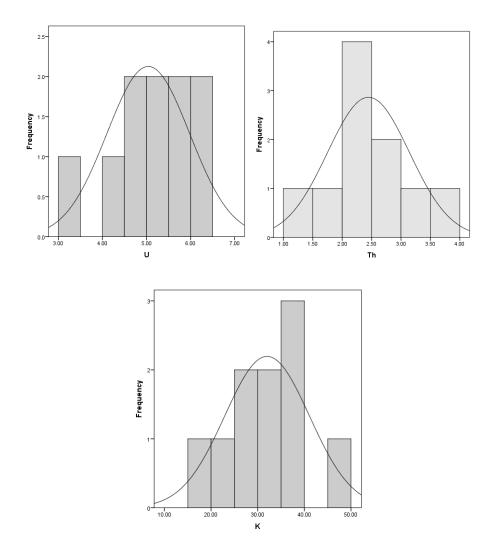


Fig. 5: Frequency distribution of 238 U, 232 Th & 40 K.



5.2 Pearson's Correlation Matrix Analysis

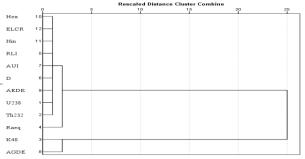
The Pearson's correlation matrix (Table 6) reveal that the association between the natural occurring radionuclides and the radiological hazards parameters. The positive correlation is found between 232 Th and the radiological parameters and significant positive correlation is observed between radiological parameters and Ra_{eq}, 40 K respectively. It express that the gamma radiation in collected ground water samples mostly comes from 232 Th and 40K radionuclides. The absorbed dose date (D) is strongly correlated with 40 K & 232 Th. So, we can assume that activity of 40 K & 232 Th dominate to the dose absorbed by the human beings. It also observed that 238 U is weakly correlated with 232Th and 40K. It reveals that the natural source of origin of 238 U, 232 Th and 40K are different.

5.3 Cluster Analysis

Hierarchical Cluster Analysis (HCA) is a multivariate statistical analysis which is used to classify the objects of the system into groups based on their similarities and to find an optimal grouping for which the observations or objects within each group are similar, but the groups are dissimilar from each other. In this study, HCA with average linkage method is further employed to explore the associations between radio-activity and radiological parameters. Three clusters are distinguished (Fig. 6): the first cluster is primarily composed of ²³⁸U, ²³²Th, D, RLI, AUI, AEDE (both outdoor and indoor), ELCR, Hex and Hin, the second cluster consisted of Ra_{eq} and main radiological parameters; and the third cluster of $^{40}{\rm K}$ and AGDE.This cluster analysis reveals that all the radiological parameters except Raeq and AGDE in the study area are due to the activity of ²³⁸U and ²³²Th. The activities of 40K mostly contribute to AGDE in the study area than any other radiological parameters.

| Paramete rs | ²³⁸ U | ²³² Th | ⁴⁰ K | Ra _{eq} | RLI | D | AUI | AGDE | AEDE | Hex | Hin | ELCR |
|-------------------|------------------|-------------------|-----------------|------------------|------------|--------|--------|--------|--------|--------|--------|------|
| ²³⁸ U | 1 | | | | | | | | | | | |
| ²³² Th | .101 | 1 | | | | | | | | | | |
| ⁴⁰ K | .297 | .553 | 1 | | | | | | | | | |
| Ra _{eq} | .621 | .784 | .804** | 1 | | | | | | | | |
| RLI | .592 | .780 | .838** | .998** | 1 | | | | | | | |
| D | .619 | .766 | .828** | .999** | .999* * | 1 | | | | | | |
| AUI | .704 | .776 | .622 | .966** | .948* * | .954** | 1 | | | | | |
| AGDE | .607 | .763 | .843** | .997** | 1.000 | 1.000 | .946** | 1 | | | | |
| AEDE | .619 | .766 | .827** | .999** | .999* * | 1.000 | .955** | 1.000 | 1 | | | |
| Hex | .627 | .782 | .792 | .998** | .995* * | .996** | .969** | .994** | .996** | 1 | | |
| Hin | .816** | .607 | .706 | .959** | .948* * | .958** | .960** | .953** | .958** | .961** | 1 | |
| ELCR | .618 | .784 | .800** | .998** | .996* * | .996** | .964** | .995** | .997** | .996** | .956** | 1 |

Table 6: Pearson correlation matrix of radiological parameters & radionuclides.







6 Conclusion

The gamma activity concentrations of radionuclides like ²³⁸U, ²³²Th and ⁴⁰K were determined in groundwater [4] samples by using HPGe gamma spectrometry. In the present study the average gamma activity concentration of 238 U is 2.59 Bq/L, 238 Th is 2.45 Bq/L and the average activity concentration of 40 K is 32.00 Bq/L. The above result has exceeded the guideline values of UNSCEAR in United States for groundwater. Not only that our studied gamma activity concentration of ground water is much more than some Middle East countries like Iran, Iraq, Egypt [5] & Saudi Arabia and European country like Italy, France, Greece. The calculated ratios of Th/U predict the preferable oxidation states of thorium and uranium in ground water according to Adams and Weaver (1958). The calculated value of radiological hazards parameters are consistent with guide line value given by UNSCEAR. Statistical analysis indicates that the probable sources of origin of studied natural occurring radionuclides are different from each other. Statistical analysis also indicates that ⁴⁰K, ²³²Th mostly contributes to the gamma radiation in all of our studied area. From the present work, it seems that the dwellers are not supposed to acquire any serious radiological complication from groundwater system. The data gathered in this study will provide baseline radiometric values of groundwater as well as drinking water in this region that can be used to evaluate the possible changes in future.

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Conflicts of interest

The authors declare no conflict of interest.

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