

# Determination of Radionuclide Concentration and Estimation of Dose rate in Industrial Wastes of Selective Industrial Area of Chittagong and Sediments of Karnapuli River near the Industrial Zone, Chittagong, Bangladesh

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**Abstract:** A study has been conducted for the determination of radionuclide concentration and estimation of dose rate in industrial wastes of selective industrial area of Chittagong and sediments of Karnapuli River near the industrial zone, Chittagong, Bangladesh by using high purity germanium (HPGe) detector available at the Radioactivity Testing and Monitoring Laboratory of Bangladesh Atomic Energy Commission, Chittagong, Bangladesh. The average activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  of industrial wastes samples were found  $45.32 \pm 9.74$  ( $40.44 \pm 8.69$ - $47.85 \pm 7.09$ )  $\text{Bq.kg}^{-1}$ ,  $47.34 \pm 16.60$  ( $41.44 \pm 14.21$ - $56.93 \pm 19.30$ )  $\text{Bq.kg}^{-1}$  and  $389.57 \pm 117.14$  ( $321.7 \pm 96.15$ - $424.55 \pm 89.03$ )  $\text{Bq.kg}^{-1}$  respectively and also of sediments samples were found  $47.13 \pm 10.59$  ( $31.29 \pm 7.52$  to  $72.04 \pm 11.52$ )  $\text{Bq.kg}^{-1}$ ,  $49.54 \pm 17.01$  ( $38.90 \pm 12.57$  to  $73.17 \pm 18.06$ )  $\text{Bq.kg}^{-1}$  and  $332.85 \pm 108.83$  ( $226.43 \pm 26.81$  to  $393.55 \pm 122.24$ )  $\text{Bq.kg}^{-1}$  respectively. The radiological hazard to humans due to the radioactivity arising from radionuclides containing in industrial wastes and sediments were assessed. For the industrial wastes the mean value of Outdoor absorbed dose, Indoor absorbed dose, Indoor annual effective dose equivalent, Outdoor annual effective dose equivalent, Total annual effective dose equivalent, External radiation hazard,  $H_{\text{extm}}$ , Internal radiation hazard,  $I_{\text{int}}$ , Radium equivalent activity,  $R_{\text{eq}}$  and Representative level index,  $I_{\text{vr}}$ , Annual gonadal dose equivalent (AGDE), Activity utilization index (AUI), Excess lifetime cancer risk (ELCR) were  $87.60 \pm 20.18$   $\text{nGy.h}^{-1}$ ,  $80.91 \pm 24.21$   $\text{nGy.h}^{-1}$ ,  $0.40 \pm 0.12$   $\text{mSv.y}^{-1}$ ,  $0.08 \pm 0.02$   $\text{mSv.y}^{-1}$ ,  $0.48 \pm 0.14$   $\text{mSv.y}^{-1}$ ,  $0.39 \pm 0.11$ ,  $0.51 \pm 0.18$ ,  $143.01 \pm 42.43$   $\text{Bq.kg}^{-1}$ ,  $0.83 \pm 0.31$   $\text{Bq.kg}^{-1}$ ,  $0.51 \pm 0.18$ ,  $460.25$   $\text{mSv.y}^{-1}$ ,  $1.02$   $\text{mSv.y}^{-1}$  and  $0.29 \times 10^{-3}$  respectively which were almost same for all sediment samples. The calculated results were compared to the recommended values and corresponding values of the world. The radium equivalent activity ( $R_{\text{eq}}$ ) in all the samples were less than the upper limit ( $370 \text{ Bq.kg}^{-1}$ ) and the obtained external hazard ( $H_{\text{ex}}$ ) indices and internal ( $H_{\text{in}}$ ) indices were far below the unity ( $\leq 1$ ). The statistical methods were also applied to know the existing correlation relations between all the calculated natural radionuclides. Basic statistics and frequency distributions for all radionuclides were used to describe the statistical characteristics of the radionuclide activities. The findings of the present research could be helpful to estimate the contamination levels of radionuclides in our natural environment. Lower values of radiological indices indicated that there is no probability of immediate health effect on workers and public due to natural radioactivity present in the samples. So, it is necessary to continue research in order to monitor the situation and to include a complete ecological chain in the research.

**Keywords:** Radionuclide; Radiological parameter; Germanium Detector; Industrial waste; sediment.

## 1. INTRODUCTION

Chittagong is the Second Largest city, Prime Sea Port and the heart of all commercial and business activities in Bangladesh. Chittagong city is situated on the bank of Karnaphuly River. Kalurghat is the heavy industrial area of Chittagong city in which a number of Mills, Factories, and Industries are located. All the wastes of these mills, factories and industries drained through the Karnaphuly River, and are deposited on the riverbed. There is also some local ship breaking yards present at the city site. As a result, the probability of non-radioactive toxic as well as radioactive contamination of water, soil and sediment and hence whole environment is increasing day by day. Since the people of the country are not conscious about the health effects of radiation contamination, so from all the perspective, it is important to find the distribution of various radionuclides present in the different environmental elements.

Natural radioactivity from naturally occurring radioactive materials (NORMs) is widely spread in the earth's environment and it exists in various geological formations such as soils, rocks, water, sediment, air and in building materials. Artificial radionuclides are from nuclear weapon tests, nuclear accidents, medical and industrial applica-

tions, etc. About 87% of the radiation doses received by humans are

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from natural radiation sources, which come from the naturally occurring radioactive isotopes of  $^{238}\text{U}$  and  $^{232}\text{Th}$  and their progeny as well as  $^{40}\text{K}$  [1]. Natural environmental radioactivity and associated external exposure due to gamma radiation depend primarily on the geological conditions of soil and sediment formations of each region in the world [2]. The distribution of natural radionuclides in the seabed can be used as a tracer for both sediments and dredged soil dispersal and accumulation mechanisms [3].

Gamma radiation emitted from naturally occurring radioisotopes, such as  $^{40}\text{K}$  and the radionuclides from the  $^{232}\text{Th}$  and  $^{238}\text{U}$  series and their decay products (also called terrestrial background radiation), which exist at trace levels in all ground formations, represents the main external source of irradiation to the human body.  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{232}\text{Th}$  are the parents of the three natural decay series, called the uranium (U) series, the actinium series and the thorium (Th) series, respectively [4]. Each of these series consists of many daughter products generated through successive decay of parent radionuclides. In the three long-lived series, decay cascades produce radioactive daughter nuclides, ultimately resulting in the stable isotopes of  $^{208}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{206}\text{Pb}$ . Natural uranium is a composite of the isotopes  $^{238}\text{U}$  (99.28%),  $^{234}\text{U}$  (0.0057%), and  $^{235}\text{U}$  (0.72%) While on a mass basis there is far more  $^{238}\text{U}$  than  $^{235}\text{U}$  in a natural sample, the activity ratio is approximately 21:1 (Powell et al, 2007). The behavior and distribution of these decay series radionuclides in the environment are based on their biogeochemistry and half-life ( $t_{1/2}$ ), and the nature of their surroundings [4].

The most long-lived radionuclide from nuclear fallout is  $^{137}\text{Cs}$ . The concentration of  $^{137}\text{Cs}$  has a maximum concentration in the northern hemisphere between 40-50 degrees latitude and a mean surficial activity up to 2.9 kBq/m. Other causes of nuclear fallout are accidents in the nuclear power industry followed by the release of radionuclides into the atmosphere. Short-lived fallout radionuclides decay rapidly usually to negligible proportions within days or weeks. But high concentrations of  $^{137}\text{Cs}$ , accumulated in the upper 10 cm of the soil can remain in the environment for many decades, with a surficial activity in the range of hundreds and thousands of Bq/m<sup>2</sup>. This surpasses the effects of radiation due to the natural environment [5].

The main objective of the present study are to determine the concentration of naturally occurring radioisotopes ( $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ ) and anthropogenic radioisotope ( $^{137}\text{Cs}$ ) and to assess their spatial distribution and radiological parameters in the industrial wastes of Kalurghat industrial area and sediments of Karnaphuly River near the industrial zone.

The results of the present work provide background data on natural and artificial radioactive isotopes and environmental pollution by technologically enhanced naturally occurring radionuclides. This research work is important to provide assistance in national program on environmental radioactivity level in the industrial wastes of Kalurghat industrial area and sediment of Karnaphuly River near the industrial zone in order to minimize the radiation exposure to the population of Chittagong.

## 2 MATERIALS AND METHODS

### Collection and preparation of samples

In the present research work, two possible radiation elevated area were selected as the study area. They are (1) Kalurghat industrial area and (2) Karnaphuly River near the industrial zone. A total of 20 samples have been collected from these two study areas. Eight industrial wastes samples collected near the four different Industries of Kalurghat industrial area and twelve sediment samples collected from the twelve different locations of Karnaphuly River near the industrial zone. The sites are 1. Regent Textile (RT). 2. Khalurghat Cold Storage (KCS). 3. Unilever Bangladesh Limited (UBL). 4. Alfa Textile (AT). 5. Fishery Ghat (FG). 6. Chaktai Khal (CK). 7. Shah Amanat Bridge (SAB). 8. Balir Hat Mohona (BHM). 9. Jaillar Char Mohona (JCM). 10. Boa liar Char Mohona (BCM). Two samples were collected from each site for better result with small change of geographical position. The geographical positions are shown in Table 1. All the samples were collected during October, 2011.

Samples were collected scientific way. After removing grass, stones and any biological materials, the samples were packed down, sieved through 2 mm mesh, homogenized and dried in an oven at 115°C for 4 days, powdered, weighed and packed into cylindrical screw-cap plastic containers of 6.5 x 7.5 cm, sealed and stored for 4 weeks to establish secular equilibrium between  $^{238}\text{U}$  and  $^{232}\text{Th}$  series with their daughters [5, 6].

Table-1.0: Summary of the samples (with geographical position) collected from the study area.

Location No.	Sampling location	Sample Code	Geographical position	
			Latitude	Longitude
01	Regent Textile	RT	22°23'37.57"N	91°53'26.11"E
02	Khalurghat Cold Storage	KCS	22°23'22.20"N	91°53'20.36"E
03	Unilever Bangladesh Limited	UBL	22°22'52.55"N	91°52'12.31"E
04	Alfa Textile	AT	22°23'22.70"N	91°52'8.24"E
05	Fishery Ghat	FG	22°19'33.16"N	91°50'9.83"E
06	Chaktai Khal	CK	22°19'38.52"N	91°50'48.11"E
07	Shah Amanat Bridge	SAB	22°19'38.15"N	91°51'6.30"E
08	Balir Hat Mohona	BHM	22°21'54.80"N	91°51'34.08"E
09	Jaillar Char Mohona	JCM	22°21'1.71"N	91°52'4.62"E
10	Boa liar Char Mohona	BCM	22°21'54.80"N	91°52'17.13"E

### Measurement of radionuclides

Measurements were made with a  $\gamma$ -spectrometry of p-type coaxial high purity germanium (HPGe) detector having germanium cylinder crystal with 52 mm outer diameter and 49.5 mm height with relative efficiency of 20% and resolution (FWHM) of 1.80 keV for the 1332.5 keV  $\gamma$ -ray energy of  $^{60}\text{Co}$ . The detector was coupled to 8192 channel computer analyzer. To reduce  $\gamma$ -ray background the detector were shielded by cylindrical 5.08 cm thick lead shield with fixed

bottom and moving cover. The full-energy-peak-efficiency of radionuclides was measured using IAEA reference samples RGU1, RGTh1 and RGK1 [5]. The reference samples are (1) IAEA/RGU-1: Uranium ore in silica powder containing radionuclides or components U, Th K; (2) IAEA/RGTh-1: Thorium ore in silica powder containing radionuclides or components Th, U, K; (3) IAEA/RGK-1: Extra pure Potassium sulphate containing radionuclides or components K, U, Th. The standard source has known concentration of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K radionuclides supplied by Canada Center for Mineral and Energy Technology (CAMET) under a contract with IAEA. The background distribution due to naturally occurring radionuclides in the environment around the detector was determined by counting an empty plastic container in the same manner as the samples. The background was subtracted in order to get net counts for the sample. The counting time for all the samples was 20 kilo second. The results are expressed with the confidence limit of ±1σ. The Gamma ray data of the gamma counting measurement are shown in Table 2.

Table 02: Gamma ray data of the gamma counting measurement [7-9].

Radionuclide	γ-Producing nuclide	γ-ray energy (keV)	Intensity in %
<sup>232</sup> Th	<sup>212</sup> Pb	238.63	44.60
	<sup>212</sup> Bi	727.17	7.56
		510.57	21.50
		2614.53	99.79
	<sup>208</sup> Tl	583.19	85.77
		338.4	11.40
<sup>228</sup> Ac	911.07	27.70	
	969.11	16.60	
	<sup>238</sup> U	<sup>214</sup> Pb	351.92
<sup>214</sup> Bi		609.31	43.30
		1120.29	15.70
<sup>226</sup> Ra	<sup>214</sup> Pb	241.98	9.00
		295.21	19.70
	<sup>214</sup> Bi	1238.11	5.94
		1764.49	17.0
<sup>40</sup> K	<sup>40</sup> K	1460.75	10.70
<sup>137</sup> Cs	<sup>137</sup> Cs	661.66	85.21

### 3. RESULTS AND DISCUSSION

The activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs radionuclide's of all industrial wastes samples in the Kalurghat industrial area and sediment samples of Karnaphuly River near the industrial zone of Chittagong district of Bangladesh are determined by following equation [9] and is shown in Table 3.

$$\text{Activity} = \frac{\text{CPS} \times 100 \times 1000}{\epsilon_f (\%) \times I_\gamma \times w_s (gm)}$$

Where, CPS = Net counts per second (i.e., CPS for sample – CPS for background)

$\epsilon_f$  = Counting gamma energy efficiency of the detector.

$I_\gamma$  = Intensity of the gamma ray,  $w_s$  = Sample weight

The error of the measurements was expressed in terms of standard deviation (±1σ).

The mean activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K of all industrial wastes samples were found 45.32±9.74Bq.kg<sup>-1</sup>, 47.33±16.60Bq.kg<sup>-1</sup> and 389.57±117.14Bq.kg<sup>-1</sup> and the average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K of all sediment samples were found 47.13 ± 10.59Bq.kg<sup>-1</sup>, 49.54 ± 17.01Bq.kg<sup>-1</sup> and 332.85 ± 108.83Bq.kg<sup>-1</sup>. The anthropogenic radionuclide <sup>137</sup>Cs was not detected in these samples. Comparison of the mean activity concentration of <sup>232</sup>Th, <sup>238</sup>U, <sup>40</sup>K and <sup>137</sup>Cs (in Bq.kg<sup>-1</sup>) of present work with some reported value near the study area is shown in Table 4 [9].

The average activity concentrations of different radionuclides were compared with the nearby regions around the Kalurghat industrial area. <sup>238</sup>U in the Kalurghat industrial area were found to be higher than the nearby areas and almost same to the Md. Kowsar Alam *et al* and <sup>232</sup>Th in the Kalurghat industrial area was found to be lower but concentration of <sup>40</sup>K was found to be higher than other study areas except Md. Kowsar Alam *et al* as shown in Table-4. The mean activity concentration of <sup>137</sup>Cs was not found in the present study whereas the some other studies areas were found.

#### Dose rate in industrial wastes and sediment samples

The outdoor absorbed dose rate in air at 1 m above the ground surface was calculated using the conversion factors given in UNSCEAR 1988 report [9]:

$$D_{\text{outdoor}} = (0.427 C_U + 0.662 C_{\text{Th}} + 0.043 C_K) \text{ nGy.h}^{-1}$$

where  $C_U$ ,  $C_{\text{Th}}$  and  $C_K$  are the average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K respectively in the samples. The average observed outdoor dose rate was 68.9±4.78 nGy.h<sup>-1</sup> for industrial wastes samples and 67.81±11.62 for sediment samples compared with the world average value of 55 nGy.h<sup>-1</sup>. The indoor contribution is assumed to be 1.3 times higher the outdoor dose [9]:

$$D_{\text{indoor}} = D_{\text{outdoor}} \times 1.3 \text{ (nGy.h}^{-1}\text{)}$$

The annual effective dose equivalent from outdoor terrestrial gamma radiation is [9]:

$$D_{\text{eff}} = \text{Outdoor dose (nGy.h}^{-1}\text{)} \times 0.7 \text{ (Sv.Gy}^{-1}\text{)} \times 8,760 \text{ (h.y}^{-1}\text{)} \times 0.2$$

where 0.2 is the outdoor occupancy factor and 0.7 Sv.Gy<sup>-1</sup> is the quotient of effective dose equivalent rate observed dose rate in air taken from the UNSCEAR Report for environmental exposure to gamma-rays of moderate energy. This value is assumed to apply equally to males and females and to the indoor and outdoor environments. For indoor exposure, using an occupancy factor of 0.8, the annual effective dose equivalent is:

$$D_{\text{eff}} = \text{Indoor dose (nGy.h}^{-1}\text{)} \times 0.7 \text{ (Sv.Gy}^{-1}\text{)} \times 8,760 \text{ (h.y}^{-1}\text{)} \times 0.8$$

The results of outdoor, indoor and total annual effective dose equivalents are shown in Table 4. The average total (outdoor plus indoor) annual effective dose equivalent from terrestrial radiation was found to be 0.49±0.03mSv for industrial wastes samples of which 0.41±0.03mSv comes from indoor and 0.09±0.01mSv from outdoor, and 0.48±7.41 for sediment samples of which 0.40±0.07 mSv comes from indoor and 0.08±0.02mSv from outdoor, the corresponding world average value is 0.41 mSv of which 0.34 mSv from indoor and 0.07 from outdoor [9].

#### External and Internal radiation hazards

The parameters external radiation hazard,  $H_{\text{ext}}$  and internal radiation

hazard,  $H_{int}$  were calculated using the criterion formula as follows [10]:

$$H_{ext} = C_U/370 + C_{Th}/259 + C_K/4810 \text{ (Bq.kg}^{-1}\text{)}$$

$$H_{int} = C_U/185 + C_{Th}/259 + C_K/4810 \text{ (Bq.kg}^{-1}\text{)}$$

The results of external and internal radiation hazards are shown in Table 5. In soil of these locations both the hazard indices were less than unity, the recommended hazard parameters  $H_{ext}$  and  $H_{int}$  also should be less than 1.

**Radium equivalent activity,  $Ra_{eq}$  and Representative level index,  $I_{yr}$ :**

In order to compare the specific activities of materials containing different concentrations of radium, thorium and potassium an index,  $Ra_{eq}$  called the radium equivalent activity concentration. Radiation hazard parameters radium equivalent activity,  $Ra_{eq}$  values were calculated using formula [10]:

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

This equation is based on the estimate that 1 Bq.kg<sup>-1</sup> of <sup>238</sup>U, 0.7 Bq.kg<sup>-1</sup> of <sup>232</sup>Th or 13 Bq.kg<sup>-1</sup> of <sup>40</sup>K generate the same  $\gamma$ -ray dose rate.

The representative level index,  $I_{yr}$  values were calculated using formula [10]:

$$I_{yr} = (C_U/150 + C_{Th}/100 + C_K/1500) \text{ Bq.kg}^{-1}$$

$I_{yr}$  was calculated to indicate different levels of external  $\gamma$ -radiation due to different combinations of specific natural activities in specific other materials. This index can be used to estimate the level of  $\gamma$  radiation hazard associated with the natural radionuclides in the materials.

Based on the annual external dose of 1.5 mGy, the safe limits activity in terms of  $Ra_{eq}$  is 370 Bq.kg<sup>-1</sup> and  $I_{yr}$  is 1 Bq.kg<sup>-1</sup>. It is observed that  $Ra_{eq}$  activity is far below the allowable limit for all the soil samples and  $I_{yr}$  value is around unity shown in Table 5. In addition to natural radionuclides <sup>137</sup>Cs, a fallout radionuclide was not detected in any sample.

**Annual gonadal dose equivalent (AGDE):**

The annual gonadal dose equivalent is a measure of the genetic significance of the yearly dose received by the population's reproductive organs [11]. Organs with rapidly dividing cells such as gonads, the active bone marrow and bone surface cells are considered as organs of interest by the United Nations Scientific Committee on the Effects of Atomic Radiation [9]. The annual gonadal dose equivalent due to specific activities of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K were estimated using the following formula [12, 11]:

$$AGDE = 3.09C_U + 4.18C_{Th} + 0.314C_K$$

Where  $C_U$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in Bq.kg<sup>-1</sup> respectively. The values of annual gonadal dose equivalent in the industrial wastes samples ranged from 503.13 to 399.19  $\mu$ Svy<sup>-1</sup> with an average value of 460.25  $\mu$ Svy<sup>-1</sup> which is higher than the world average value of 300  $\mu$ Svy<sup>-1</sup> [9] and The values of annual gonadal dose equivalent in the sediment samples ranged from 620.70 to 385.01  $\mu$ Svy<sup>-1</sup> with an average value of 457.20  $\mu$ Svy<sup>-1</sup> which is higher than the UNSCEAR average value of 300  $\mu$ Svy<sup>-1</sup> [9] which is shown in table 7.

**Activity utilization index (AUI):**

In order to facilitate the calculation of dose rates in air from different combinations of the three radionuclides in industrial wastes and sediments by applying the appropriate conversion factors, an activity utilization index (AUI) is constructed that is given by the following equation [11]:

$$AUI = (C_U/50) f_U + (C_{Th}/50) f_{Th} + (C_K/500) f_K$$

Here,  $f_U = 0.462$ ,  $f_{Th} = 0.604$  and  $f_K = 0.041$  are the fractional contributions to the total dose rate in air due to the gamma radiation from

the <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K. The values of annual utilization index in the industrial wastes samples ranged from 1.14 to 0.90  $\mu$ Svy<sup>-1</sup> with an average value of 1.02  $\mu$ Svy<sup>-1</sup> which is lower than the world average value of 2  $\mu$ Svy<sup>-1</sup> [9] and The values of annual utilization index in the sediment samples ranged from 1.57 to 0.85  $\mu$ Svy<sup>-1</sup> with an average value of 1.06  $\mu$ Svy<sup>-1</sup> which is lower than the world average value of 2  $\mu$ Svy<sup>-1</sup> [9] which is shown in table 7. So, the samples present no radiation hazard and are not harmful for human beings. Moreover the samples can be safely used for building materials.

**Excess lifetime cancer risk (ELCR):**

Since gamma radiation provides information on the Excess of Lifetime Cancer Risks (ELCRs), it is necessary to measure this parameter. The probability or extra risk of developing lung cancer due to exposure to gaseous radionuclides indoors incurred over the lifetime of an individual is estimated using the excess lifetime cancer risk (ELCR). Based upon calculated values of annual effective dose (AEDE), ELCR was estimated using the following formula [13, 11]:

$$ELCR = AEDE + DL + RF$$

Where AEDE is the annual effective dose equivalent, DL is the average lifetime duration 60 countries including Bangladesh (70 years) and RF is the fatal risk factor per Sievert assumed to be 0.05 Sv<sup>-1</sup> in this study as per ICRP-106 [14]. The ELCR value in the industrial wastes samples ranged from  $0.25 \times 10^{-3}$  to  $0.30 \times 10^{-3}$  with an average value of  $0.29 \times 10^{-3}$  which didn't exceed the world average value of  $0.28 \times 10^{-3}$  [15]. The ELCR value in the sediment samples ranged from  $0.24 \times 10^{-3}$  to  $0.39 \times 10^{-3}$  with an average value of  $0.28 \times 10^{-3}$  which is lower than the world average value of  $0.28 \times 10^{-3}$  [15].

**Table.03: Activity concentrations (Bq.kg<sup>-1</sup>) of naturally occurring radionuclides <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs of all industrial wastes and sediment samples.**

Samples	Sample No.	Activity concentrations (Bq.kg <sup>-1</sup> ) of <sup>238</sup> U with uncertainty ( $\pm 1\sigma$ )	Activity concentrations (Bq.kg <sup>-1</sup> ) of <sup>232</sup> Th with uncertainty ( $\pm 1\sigma$ )	Activity concentrations (Bq.kg <sup>-1</sup> ) of <sup>40</sup> K with uncertainty ( $\pm 1\sigma$ )	Activity concentrations (Bq.kg <sup>-1</sup> ) of <sup>137</sup> Cs with uncertainty ( $\pm 1\sigma$ )
Industrial wastes samples	RT-01	47.85 ± 7.09	49.31 ± 22.63	417.69 ± 153.55	BDL<0.4
	RT-02	46.34±13.71	48.18±23.59	381.94±153.37	BDL<0.4
	KCS-03	45.85± 11.65	56.93 ± 19.30	393.26 ± 129.68	BDL<0.4
	KCS-04	47.57±11.68	52.62±15.61	363.11±129.52	BDL<0.4
	UBL-05	40.44 ± 8.69	41.44 ± 14.21	321.7 ± 96.15	BDL<0.4
	UBL-06	43.11±8.75	42.24±13.6	399.34±96.28	BDL<0.4
	AT-07	46.77 ± 8.18	45.25 ± 13.19	414.95± 89.53	BDL<0.4
	AT-08	44.67±8.16	42.73±10.69	424.55±89.03	BDL<0.4
<b>Mean value</b>		<b>45.32±9.74</b>	<b>47.33±16.60</b>	<b>389.57±117.14</b>	<b>BDL&lt;0.4</b>
<b>Standard Deviation</b>		<b>2.51</b>	<b>5.49</b>	<b>34.08</b>	
Sediment samples	FG-9	72.04 ± 11.52	73.17 ± 18.06	226.43 ± 26.81	BDL<0.4
	FG-10	70.59±11.49	70.37±15	345.32±123.18	BDL<0.4
	CK-11	31.29 ± 7.52	49.39 ± 12.56	348.56 ± 83.81	BDL<0.4
	CK-12	30.67±7.57	45.03±12.57	324.89±84.61	BDL<0.4
	SAB-13	45.43 ± 9.53	41.38 ± 15.44	321.43 ± 104.56	BDL<0.4
	SAB-14	42.92±9.57	43.16±15.54	373.51±105.8	BDL<0.4
	BHM-15	47.39 ± 12.68	49.45 ± 20.91	318.95 ± 141.13	BDL<0.4
	BHM-16	41.40±12.82	45.11±21.09	325.6±144.07	BDL<0.4
	JCM-17	40.25 ± 11.04	38.90 ± 12.57	335.01 ± 123.16	BDL<0.4
	JCM-18	41.48±11.17	39.10±18.24	339.05±124.65	BDL<0.4
	BCM-19	51.41± 11.07	50.88 ± 18.07	393.55 ± 122.24	BDL<0.4
	BCM-20	50.65±11.06	48.50±18.06	341.94±121.97	BDL<0.4
<b>Mean value</b>		<b>47.13 ± 10.59</b>	<b>49.54 ± 17.01</b>	<b>332.85 ± 108.83</b>	<b>BDL&lt;0.4</b>
<b>Standard Deviation</b>		<b>12.99</b>	<b>11.14</b>	<b>40.03</b>	

Table: 04: Comparison of the mean activity concentration of  $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  (in  $\text{Bq.kg}^{-1}$ ) of present work with some reported value near the study area.

SL No.	References	Area	$^{238}\text{U}$	$^{232}\text{Th}$	$^{40}\text{K}$	$^{137}\text{Cs}$
1	Present study	Industrial wastes samples in the Kalurghat industrial area	45.32±9.74	47.33±16.60	389.57±117.14	BDL<0.4
		Sediment samples of Karnaphuly River near the industrial zone	47.13 ± 10.59	49.54 ± 17.01	332.85 ± 108.83	BDL<0.4
2	M.N Alam <i>et al</i> [16]	Sediments of the Karnaphuly river estuary of Bangladesh	5.87±1.21-27.85±1.71	10.44±2.31-64.02±8.13	118.28±19.70-608.21±75.70	0.09±0.06 - 4.64±0.10
3	M.I Chowdhury <i>et al</i> [17]	Sediments of the Karnaphuly river of Bangladesh	37.9±20	65.5 ±12.2	272 ± 35	2.2±0.45
		Sediments of the Shango river of Bangladesh	25.4±2.9	57.5±3.0	255±18	2.1±0.5
4	Md. Kowsar Alam <i>et al</i> [18]	Karnaphuly River of Bangladesh	45.32±4.44	79.68±6.44	856.88±59.45	BDL<0.4
5	M.N Alam <i>et al</i> [19]	Sediments of the Cox's Bazar sea beach of Bangladesh	19.0±4.8	36.7±6.5	458±160	BDL<0.4

Table 05: Outdoor, Indoor absorbed dose, External, Internal radiation hazard and other radiological parameters de-

Sample code	Outdoor absorbed dose ( $\text{nGy.h}^{-1}$ )	Indoor absorbed dose ( $\text{nGy.h}^{-1}$ )	Indoor annual effective dose equivalent ( $\text{mSv.y}^{-1}$ )	Outdoor annual effective dose equivalent ( $\text{mSv.y}^{-1}$ )	Total annual effective dose equivalent ( $\text{mSv.y}$ )	External radiation hazard, $H_{\text{ext}}$	Internal radiation hazard, $I_{\text{int}}$	Radium equivalent activity, $Ra_{\text{eq}}$ ( $\text{Bq.kg}^{-1}$ )	Representative level index, $I_{\text{yr}}$ ( $\text{Bq.kg}^{-1}$ )
RT-01	68.09±28.05	85.22±29.52	0.42±0.14	0.09±0.03	0.51±0.17	0.41±0.14	0.54±0.16	383.23±51.27	1.09±0.38
RT-02	74.14±23.31	81.70±33.66	0.40±0.17	0.08±0.03	0.48±0.20	0.39±0.16	0.52±0.20	367.02±59.25	1.09±0.38
KCS-03	70.73±20.89	88.97±27.98	0.44±0.14	0.09±0.03	0.53±0.17	0.43±0.13	0.55±0.16	376.43±49.23	1.09±0.38
KCS-04	58.52±17.24	84.87±25.06	0.42±0.12	0.09±0.03	0.51±0.15	0.41±0.12	0.54±0.15	357.48±43.98	1.09±0.38
UBL-05	63.54±16.87	70.22±20.69	0.34±0.10	0.07±0.02	0.41±0.12	0.34±0.10	0.45±0.12	298.68±36.41	1.09±0.38
UBL-06	67.76±16.07	76.25±20.25	0.37±0.10	0.08±0.02	0.45±0.12	0.36±0.10	0.48±0.12	335.37±35.61	1.09±0.38
AT-07	65.62±14.39	81.31±19.28	0.40±0.09	0.08±0.02	0.48±0.11	0.39±0.09	0.51±0.11	349.39±33.94	1.09±0.38
AT-08	68.09±28.05	78.74±17.26	0.39±0.08	0.08±0.02	0.47±0.10	0.37±0.08	0.49±0.10	345.07±30.30	1.09±0.38
Mean	<b>87.60±20.18</b>	<b>80.91±24.21</b>	<b>0.40±0.12</b>	<b>0.08±0.02</b>	<b>0.48±0.14</b>	<b>0.39±0.11</b>	<b>0.51±0.18</b>	<b>143.01±42.43</b>	<b>0.83±0.31</b>

rived from activity of Industrial Wastes samples

Sample code	Outdoor absorbed dose (nGy.h <sup>-1</sup> )	Indoor absorbed dose (nGy.h <sup>-1</sup> )	Indoor annual effective dose equivalent (mSv.y <sup>-1</sup> )	Outdoor annual effective dose equivalent (mSv.y <sup>-1</sup> )	Total annual effective dose equivalent (mSv.y)	External radiation hazard, H <sub>ext</sub>	Internal radiation hazard, I <sub>int</sub>	Radium equivalent activity, Ra <sub>eq</sub> (Bq.kg <sup>-1</sup> )	Representative level index, I <sub>yr</sub> (Bq.kg <sup>-1</sup> )
FG-9	88.84±18	106.60±21.60	0.52±0.11	0.11±0.02	0.63±0.13	0.52±0.11	0.72±0.14	194.11±39.41	1.36±0.28
FG-10	91.50±22.15	109.80±26.58	0.54±0.13	0.11±0.03	0.65±0.16	0.53±0.13	0.73±0.16	197.81±46.81	1.40±0.34
CK-11	61.02±18.87	73.22±22.64	0.36±0.11	0.07±0.02	0.43±0.13	0.35±0.11	0.43±0.13	128.76±40.06	0.93±0.29
CK-12	56.85±18.85	68.22±22.62	0.33±0.11	0.07±0.02	0.40±0.13	0.32±0.11	0.41±0.13	120.08±40	0.87±0.29
SAB-13	60.60±22.51	72.71±27.01	0.36±0.13	0.07±0.03	0.43±0.16	0.35±0.13	0.47±0.15	129.35±47.74	0.93±0.34
SAB-14	62.95±22.46	75.54±26.95	0.37±0.13	0.08±0.03	0.45±0.16	0.36±0.13	0.48±0.15	133.40±47.62	0.97±0.34
BHM-15	66.65±21.77	79.98±26.12	0.39±0.13	0.08±0.03	0.47±0.16	0.39±0.12	0.51±0.16	142.66±45.77	1.02±0.33
BHM-16	61.52±21.89	73.27±26.27	0.36±0.13	0.08±0.03	0.44±0.16	0.35±0.12	0.47±0.16	130.98±45.99	0.94±0.34
JCM-17	57.33±18.33	68.80±22	0.34±0.11	0.07±0.02	0.41±0.13	0.33±0.10	0.44±0.13	121.67±38.50	0.88±0.28
JCM-18	58.16±18.44	69.80±22.13	0.34±0.11	0.07±0.02	0.41±0.13	0.33±0.10	0.45±0.13	123.50±38.73	0.89±0.28
BCM-19	70.30±19.89	84.37±23.87	0.41±0.12	0.09±0.02	0.50±0.14	0.41±0.11	0.55±0.14	150.50±41.90	1.08±0.31
BCM-20	70.64±22.24	84.77±26.68	0.42±0.13	0.09±0.03	0.51±0.16	0.41±0.13	0.54±0.16	150.31±46.98	1.09±0.34
Mean	87.65±20.45	80.64±24.54	0.40±0.12	0.8±0.03	0.48±0.15	0.39±0.12	0.52±0.15	143.59±43.29	1.03±0.0.31

Table 06: Outdoor, Indoor absorbed dose, External, Internal radiation hazard and other radiological parameters derived from activity of Sediment samples.

Table 07: AUI, AGDE, ELCR derived from activity of Industrial Wastes Samples and Sediment samples.

Industrial Wastes Samples				Sediment Samples.			
Sample code	AUI	AGDE (mSv.y <sup>-1</sup> )	ELCR	Sample code	AUI	AGDE (mSv.y <sup>-1</sup> )	ELCR
RT-01	1.07	485.12	0.30×10 <sup>-3</sup>	FG-9	1.56	599.55	0.38×10 <sup>-3</sup>
RT-02	1.04	464.51	0.29×10 <sup>-3</sup>	FG-10	1.53	620.70	0.39×10 <sup>-3</sup>
KCS-03	1.14	503.12	0.31×10 <sup>-3</sup>	CK-11	0.91	412.58	0.26×10 <sup>-3</sup>
KCS-04	1.10	480.95	0.30×10 <sup>-3</sup>	CK-12	0.85	385.01	0.24×10 <sup>-3</sup>
UBL-05	0.90	399.19	0.25×10 <sup>-3</sup>	SAB-13	0.94	414.27	0.26×10 <sup>-3</sup>
UBL-06	0.94	435.16	0.27×10 <sup>-3</sup>	SAB-14	0.94	430.31	0.27×10 <sup>-3</sup>
AT-07	1.01	463.95	0.29×10 <sup>-3</sup>	BHM-15	1.06	453.28	0.28×10 <sup>-3</sup>
AT-08	0.96	449.95	0.28×10 <sup>-3</sup>	BHM-16	0.95	418.72	0.26×10 <sup>-3</sup>
Range	1.14-0.90	503.12-399.19	0.25×10 <sup>-3</sup> -0.30×10 <sup>-3</sup>	JCM-17	0.87	392.16	0.24×10 <sup>-3</sup>
Mean	1.02	460.25	0.29×10 <sup>-3</sup>	JCM-18	0.88	398.07	0.24×10 <sup>-3</sup>
World Average [22]	2	300	0.29×10 <sup>-3</sup>	BCM-19	1.12	478.90	0.30×10 <sup>-3</sup>
				BCM-20	1.09	482.81	0.30×10 <sup>-3</sup>
				Range	1.57-0.85	620.70-385.01	0.39×10 <sup>-3</sup> -0.24×10 <sup>-3</sup>
				Mean	1.06	457.20	0.28×10 <sup>-3</sup>

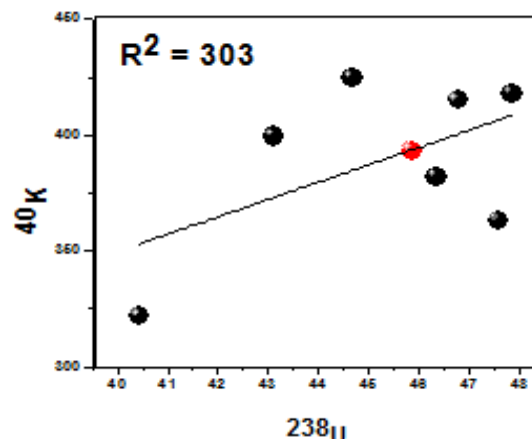
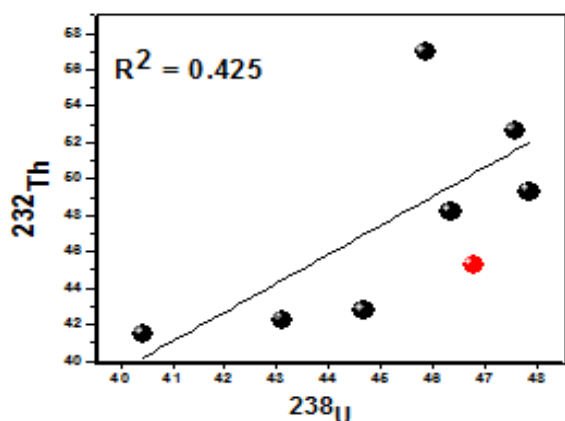
	World Average [22]	2	300	$0.29 \times 10^{-3}$
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**Basic Statistical analysis:**

Activity concentrations of naturally occurring radionuclide's ( $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) were measured by using Gamma Spectrometry analysis. Statistical analysis is established using the activity concentrations of naturally occurring radionuclide's. The basic statistical analyses were used to imply the statistical characteristics of radionuclide's and its spatial distribution.

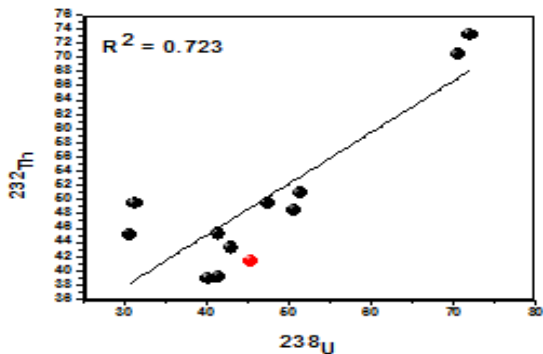
- **Pearson's correlation coefficient:**

Pearson's correlation coefficient is used to determine the mutual relationships and strength of association between pairs of variables through the calculation of the linear Pearson's correlation coefficient. It gives the idea of how well data fits a line or curve. Pearson's correlation coefficient returns a value of -1 (a perfect negative fit) and +1 (a perfect positive fit). The values of  $R^2$  can vary based upon the "type" of data being examined. The correlation between  $^{238}\text{U}$  and  $^{232}\text{Th}$  in the industrial wastes samples is found to be moderate positive correlative ( $R^2 = 0.425$ ) indicates that their origin and behavior in the Kalurghat industrial area are the same and exist together in nature while the correlation between  $^{238}\text{U}$  and  $^{40}\text{K}$  in the industrial wastes samples is found to be weak positive correlative ( $R^2 = 0.303$ ) indicates that their origin may be same but behavior environment differ and the emission of gamma radiation is principally due to  $^{238}\text{U}$  and  $^{232}\text{Th}$ . The activity concentrations of  $^{232}\text{Th}$  and  $^{40}\text{K}$  (y- coordinate) increase with the increase of activity concentration of  $^{238}\text{U}$  (x-coordinate). Hence, all the three radionuclides have positive correlation coefficient; they have good distribution and contribute significantly to gamma-ray emission at the sampling point.

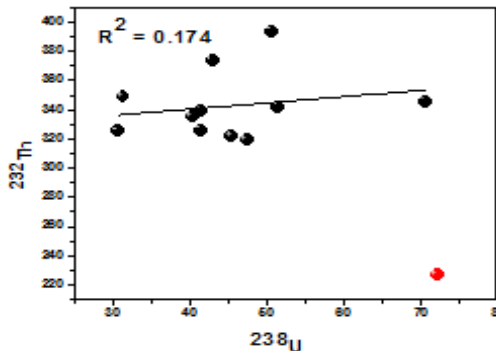




**Figure-3:** Correlation between  $^{238}\text{U}$  and  $^{232}\text{Th}$  in industrial wastes samples.



**Figure-4:** Correlation between  $^{238}\text{U}$  and  $^{40}\text{K}$  in industrial wastes samples.

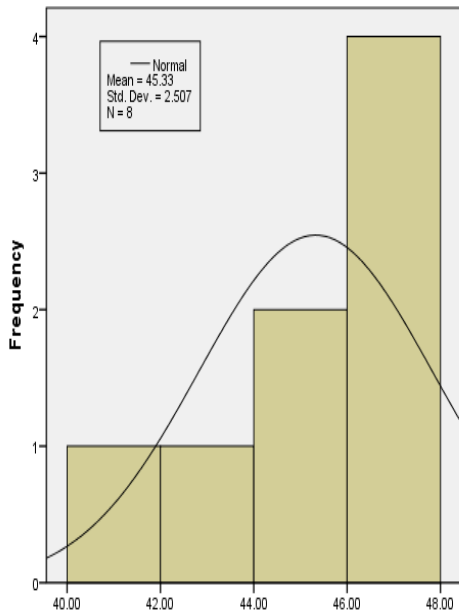


**Figure-5:** Correlation between  $^{238}\text{U}$  and  $^{232}\text{Th}$  in Sediment samples.

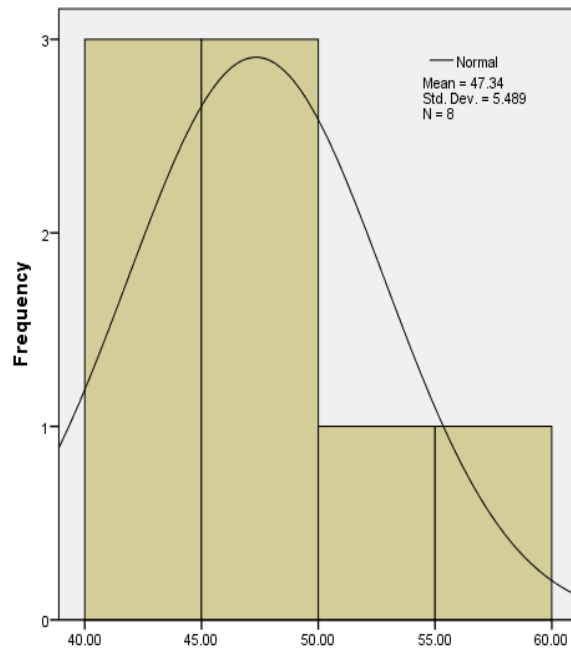
**Figure-6:** Correlation between  $^{238}\text{U}$  and  $^{40}\text{K}$  in Sediment samples.

- **Histogram analysis:**

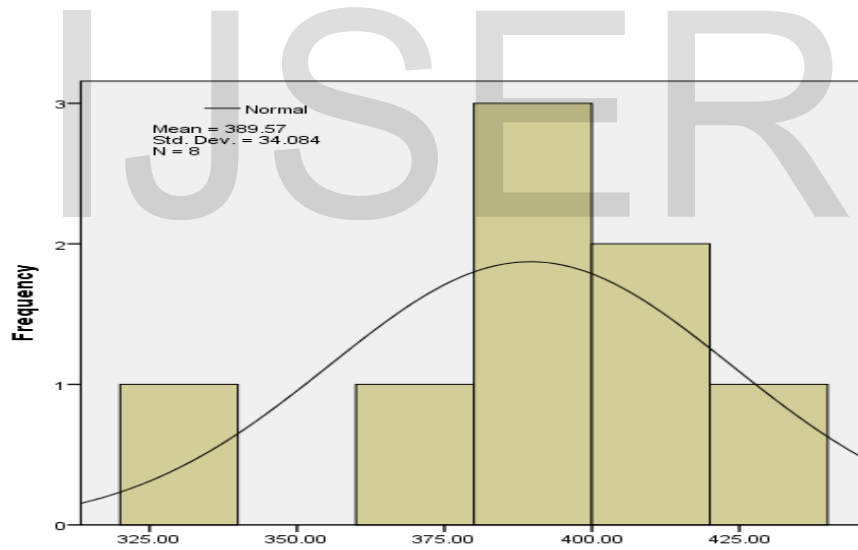
A histogram is the common form of graphical representation of a frequency distribution. It is an estimate of the probability distribution of continuous variables. It represents the tabulated frequencies, shown as adjacent rectangles, erected over discrete intervals (bins), with an equal area to the frequency of the observations in the intervals. The height of a rectangle is also equal to the frequency density of the interval. The total area of the histogram is equal the number of data. The rectangles touch each other to indicate that the original variable is continuous (radioactivity is also continuous process). The frequency distributions of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the industrial wastes samples and sediment samples were analyzed and the histograms are given below (**Figure: 5-6**). Theses radionuclides demonstrate a normal distribution (bell-shaped) and there is no complexity of minerals in the investigated samples due to the absent of degree of multi-modal feature. All the figures are asymmetric due to positive and negative Skewness of the dataset (Skewness is a measure of the asymmetry of the probability distribution of a real valued random variable). Histogram studies (graphs) were performed using the commercial statistics software package **IBM SPSS statistics** (version 23) for Windows



$^{238}\text{U}$  (Positive Skewness)

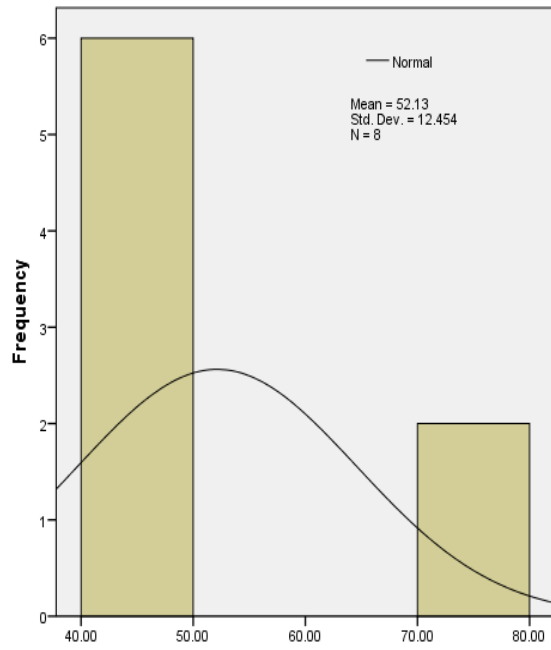
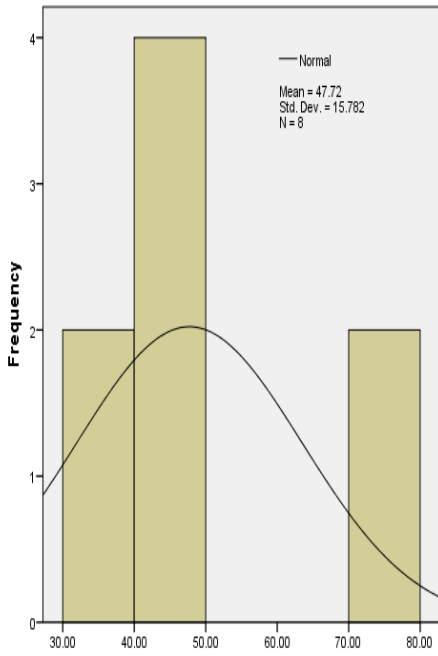


$^{232}\text{Th}$  (Negative Skewness)



$^{40}\text{K}$  (Positive Skewness)

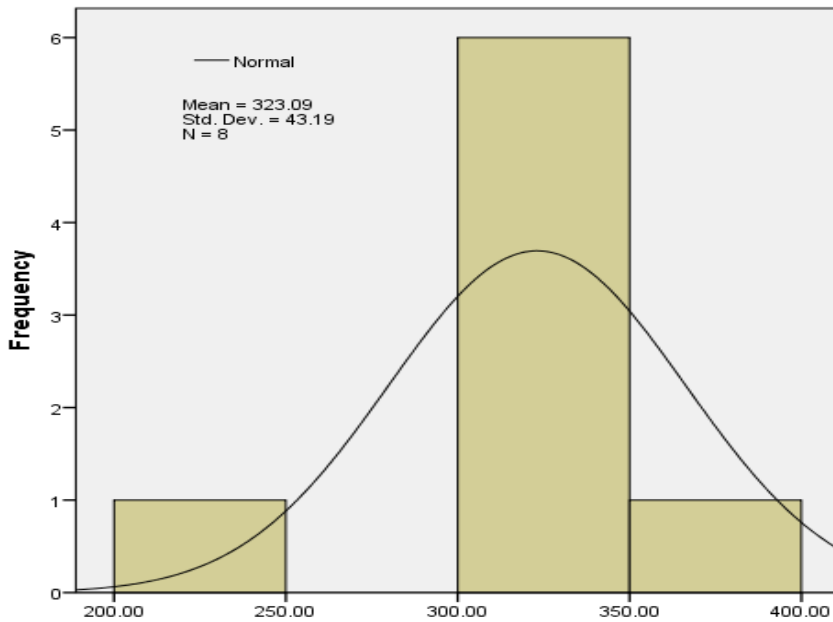
Figure-5: The frequency distribution of the activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in industrial wastes sample.



$^{238}\text{U}$  (Negative Skewness)

$^{232}\text{Th}$  (Negative Skewness)

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$^{40}\text{K}$  (Positive Skewness)

Figure-6: The frequency distribution of the activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in sediment sample.

## Conclusion

The activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in industrial wastes and sediment samples were determined. Using the activity concentrations of these radionuclides, radiological hazard indices such as Outdoor absorbed dose, Indoor absorbed dose, total Annual Effective Dose Equivalent (AEDE), radium equivalent ( $Ra_{eq}$ ), Representative level index ( $I_{yr}$ ), external hazard index ( $H_{ex}$ ), internal hazard index ( $H_{in}$ ), Annual gonadal dose equivalent (AGDE), Activity utilization index (AUI) and Excess lifetime cancer risk (ELCR) were estimated in order to assess the radiation effects to humans resulting from the industrial wastes samples and sediment samples. The External radiation hazard index and internal hazard index value of the each sample below the world average value (1) reported in UNSCEAR-1988. It seems therefore that no potential radiological health hazard may directly be associated with the industrial wastes and sediment samples. Average value of ELCR indicates no cancer risk in the Kalurghat industrial area and Karnaphuly River near the industrial zone. The processed statistical methods (Pearson correlation coefficient and Histogram) also confirm that these study areas do not pose significant gamma radiation effects. The future research needs to study more extensively on the other locations. The estimated values in this research can be used as a baseline for future research and the data obtained in this study may be useful for radiological mapping of the study area.

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