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Radiological Risk Assessment of Radionuclide Contamination in Sediment and Water of the Halda River Estuary, Bangladesh

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Abstract

To assess potential risks due to the radioactive contamination in the sediment and water of the Halda River, the HPGe gamma-ray spectrometry was utilized at Radioactivity Testing and Monitoring Laboratory (RTML) Mongla, Bangladesh Atomic Energy Commission, to measure and analyzed the activity concentrations of naturally occurring primordial radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in sediment and water samples. The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in collected sediment and water samples are found to be (14.76±2, 25.6±3.33, 299.5±29.95) Bq/Kg and (2.6±0.48, 3.85±0.92, 68±8.41) Bq/Kg respectively, which are less than the global average 35, 30 and 400 BqKg^{-1}. So far, no presence of ^{137}Cs was detected in any sample collected in our current research work. Various radiological hazard parameters were also calculated and compared with UNSCEAR (2000) report. Also, our estimated excess lifetime cancer risk is less than the world recommended level. For statistical analysis correlations, Principal Component Analysis (PCA) and Cluster Analysis (CA) were determined between both of sediment and water samples. However, analyzing the results obtained, it appears that our results do not currently pose any risk to the biodiversity of the Halda River and the lives of the people dependent on it and its water users, but in the future, due to natural and human-made causes, radiation in river sediment and water may increase, which our policymakers should be aware of.

Keywords: Halda River, HPGe-detector, Activity concentration (AC.), Radiological hazards, Excess Lifetime Cancer Risk (ELCR), Statistical Investigation.

1. Introduction

Bangladesh is a riverine country with over 410 rivers; Halda is a rocky mountain geological river one of them. The Halda River is uniquely significant as the country's only natural spawning ground for carps and the only freshwater fish breeding ground in South Asia [2-5 and 15]. Environmental pollution is a major topic of discussion in the present world, which is gradually making the human race think, and one of the most important issues is radioactive pollution. The continuous emission of alpha, beta, gamma rays from unstable elements is basically called radioactivity. Natural radioactivity (^{226}Ra, ^{232}Th, ^{40}K) is mainly caused by natural disasters such as cosmic rays, nuclear explosions, mountain erosion, etc., and on the other hand, the radioactivity that is also created by humans due to urbanization and industrialization is artificial radioactivity (^{137}Cs). Radioactivity from natural sources affects about 83% of humans. Radioactivity is found in old rocks, soil, water, air, plants, fish, food, vegetables, living things and inanimate objects, which are increasing day by day. It is also cause of dangerous diseases like cancer and heart attacks in humans [17]. Radioactivity can enter the marine environment in several ways: upwelling currents that carry radioactive material from solid particles in the water to the surface on the seabed, drifts carrying radioactive contaminants from terrestrial sources, direct deposition from atmospheric fallout and emissions, and discharges to groundwater from nuclear waste sites [10]. The direct hazards to human health posed by radioactive contamination of seafood water [5]. This poses a serious risk of contamination in the marine food chain. Similarly, it has also been discovered that radioactive isotopes such as ^{90}Sr and ^{137}Cs accumulate in the skeletal system and muscles of fish, which is dangerous to the health of both humans and marine life [18]. That's why Monitoring of environmental radioactivity is essential for establishing baseline databases and ensuring public health.

Therefore, considering the biodiversity of the Halda river and the risks to the natural fish breeding grounds and the quality of life of people on the banks of the Halda river and its water users, we felt the need to test the radiation of its sediment and water and collected samples at the Radioactivity Testing and Monitoring Laboratory (RTML) of the Bangladesh Atomic Energy Commission, Mongla. There, we determined the radioactivity and various hazard parameters of the collected samples through an HPGe detector set-up with 30% relative efficiency, such as the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were used to calculate the Radiological

Parameters, and compared with UNSCEAR (2000) report. Also the correlation coefficients between the activity of sediment and water were calculated to explain interdependence of contamination.

2. Required materials and procedures

2.1 Preparation of collected samples

By 10 samples of sediment and water were collected from the Halda River on 7 January 2025 (Shown in Fig.1). By 2 Kg per Sediment sample were sun-dried and oven-dried at 100–105°C to remove moisture, and then crushed, homogenized through a 4mm mesh, and sealed for 30 days to achieve secular equilibrium. By 1.5 L per Water sample were acidified with HCl, placed in Marinelli beakers, and similarly sealed for 30 days.

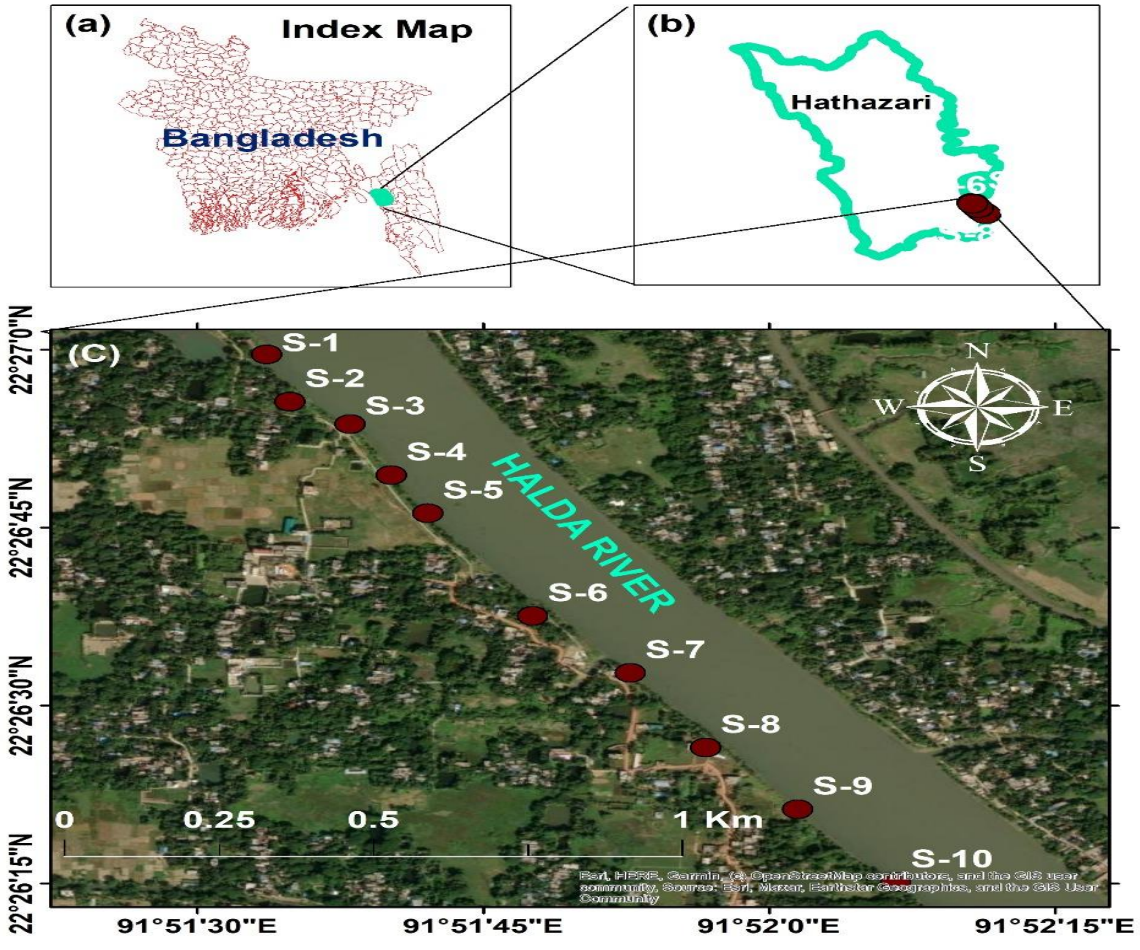


Fig.1**:** Our study area, Halda River, Hathazari, Chittagong, Bangladesh.

2.2 Instrumental Setup

Each sample was counted every 2000 seconds on an HPG (Energy Calibrated) detector with a relative efficiency of 30%. Energy scaling was then performed using known photo peaks to ensure linear accuracy of gamma energy and pulse height with the help of Multi-channel Analyzer HYBRID.

2.3 Radiological Hazard's

Radioactive parameters have been determined to assess the risks of health and biodiversity of Halda River.

Activity concentration (AC.): By using a calibrated HPGe detector the amounts of radionuclides and their activity concentration (A) in $Bqkg^{-1}$ of collected and prepared samples were determined.

$$AC. = \frac{CPS}{E \times I \times W} \quad (1)$$

Here,

'CPS' is the sample net counts per second for the background are represented. 'E' is the detector's photo peak efficiency at the relevant gamma-ray energy. 'I' is the probability of the sample's emission at that energy, and 'W' is the sample's net weight in kilograms respectively. [16].

$$\sigma^2 = \{C_{SC}/(t_{SC})\text{^}2 + C_{BC}/(t_{BC})\text{^}2\} \quad (2)$$

Where, C_{BC} is the background counts recorded in time t_{BC}, and C_{SC} is the sample counts recorded in time t_{SC}. By using equation (1) and (2) the standard deviation (±2σ) was calculated.

Radium Equivalent (Ra_{eq}): To determine the risk index of radium, thorium, and potassium, it is necessary to know the radium equivalent, so our research determined it using the formula below.

$$Ra_{eq} = AC._{Ra} + 1.43\, AC._{Th} + 0.077\, AC._{K} \quad (3)$$

Outdoor and Indoor Absorbed Doses (D_{out} and D_{in}): To determine the annual effective dose, we are determined the external and internal doses the following equation:

$$\mathrm{D(out)} = 0.46\mathrm{AC._{Ra}} + 0.59\mathrm{AC._{Th}} + 0.043\mathrm{AC._{K}} \quad (4)$$

$$\mathrm{D\ (in)} = 1.4 \times Dout(From\ Equ.4) \quad (5)$$

Annual Effective Dose Equivalent (AEDE) ($\mathrm{mSvyr^{-1}}$) **:** The annual effective dose equivalent was determined by assuming a person's 8760 hours of indoor and outdoor exposure to be 0.8 and 0.2 respectively, where 0.7 is the radiological health risk level.

Outdoor Annual Effective Equivalent, (EAED)

$$\mathrm{E_{(Out)}} = \mathrm{D(out)} \times 24 \times 365 \times 0.2 \times 0.7 \times 10^{-6} \quad (6)$$

Indoor Annual Effective Equivalent, (EAED)

$$\mathrm{E_{(in)}} = \mathrm{D(in)} \times 24 \times 365 \times 0.8 \times 0.7 \times 10^{-6} \quad (7)$$

Hazards of Outdoor and Indoor (H_{out} and H_{in}): To know the risk level of external and respiratory damage to humans due to gamma radiation and radon, it is necessary to know the value of external and internal hazard, which is safe if the value is less than 1 according to world standards. We determined it through the following equations

$$H(out) = \frac{AC.(Ra)}{370} + \frac{AC.(Th)}{259} + \frac{AC.(K)}{4810} \quad (8)$$

$$H(in) = \frac{AC.(Ra)}{185} + \frac{AC.(Th)}{259} + \frac{AC.(K)}{4810} \quad (9)$$

External (H_{out}) and Internal (H_{in}) hazard indices both safe limit < 1.

External and Internal Excess Lifetime Cancer Risk ($ELCR_{out}$ and $ELCR_{in}$): We used the following internationally accepted equations to quantify the cancer risk associated with radiation exposure, which we discuss extensively in our Results and Conclusions section.

$$\mathrm{ELCR}_{Out} = E_{AED(Out)} \times D_L \times R_F \quad (10)$$

$$\mathrm{ELCR}_{in} = E_{AED(in)} \times D_L \times R_F \quad (11)$$

Here, Annual effective dosage equivalent (E_{AED}), the average lifetime of people in Bangladesh is D_L=72.3 years. Also R_F=0.05 according to the International Commission on Radiological Protection (ICRP) recommended fatal risk factor per Sievert [1, 6, 7, 13, 14, 16].

3. Results and Clarification

3.1 Activity Concentration (AC.): Table-1: Activity Concentration (AC.) for each ten collected sample of Sediment and Water.

Activity Table	Activity Concentration (AC.) **Sediment** (Bq/kg)			Activity Concentration (AC.) **Water** (Bq/kg)			GPS Location of Sample collection Point	
Sample ID	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K	Latitude	Longitude
S_1	11.9±1.666	25±3.25	290±29	3.3±0.561	4.3±0.989	70±9.1	22°26'59.613"N	91°51'33.627"E
S_2	15.7±2.041	25±3.25	290±29	2.8±0.504	04±0.96	68±8.84	22°26'55.267"N	91°51'33.499"E
S_3	12.4±1.86	26±3.38	294±29.4	03±0.51	4.2±0.924	78±9.36	22°26'53.723"N	91°51'38.018"E
S_4	16.8±2.184	25±3.25	310±31	2.5±0.5	3.1±0.868	63±8.19	22°26'49.42"N	91°51'40.17"E
S_5	14.2±1.988	26±3.38	310±31	2.6±0.494	3.5±0.875	64±8.32	22°26'46.195"N	91°51'42.07"E
S_6	16.1±2.093	26±3.38	300±30	2.2±0.44	3.5±0.805	66±8.58	22°26'36.889"N	91°51'46.037"E
S_7	17.2±2.236	25±3.25	291±29.1	02±0.38	04±0.92	58±7.54	22°26'32.76"N	91°51'52.704"E
S_8	14±1.96	24±3.12	300±30	2.3±0.437	04±0.96	65±8.45	22°26'26.458"N	91°51'56.657"E
S_9	12.6±1.89	27±3.51	310±31	2.6±0.468	3.8±0.95	60±7.8	22°26'21.265"N	91°52'1.496"E
S_{10}	16.7±2.171	27±3.51	300±30	2.7±0.513	4.1±0.984	61±7.93	22°26'15.097"N	91°52'6.647"E
Average	14.76±2.009	25.6±3.328	299.5±29.95	2.6±0.481	3.85±0.921	68±8.411		
World Average [14,16]	35	30	400					

182 **Table-2**: Data for various radiological hazard parameters.

Sample ID	Radium Equivalent (Raeq) Bq/kg	Outdoor absorbed Dose (Dout) nGy/h	Indoor Absorbe Dose (Din) nGy/h	Hazard of Outdoor (Hout)	Hazard of Indoor (Hin)	Outdoor annual Effective Dose (Eout) mSvy/r	Indoor annual Effective Dose (Ein) mSvy/r	External Excess lifetime cancer risk (ELCR out) $\times 10^{-3}$	Internal Excess lifetime cancer risk (ELCRin) $\times 10^{-3}$
	Sediment								
S_1(S)	69.981	32.694	45.772	0.189	0.222	0.040	0.161	0.145	0.582
S_2(S)	73.781	34.442	48.219	0.199	0.242	0.042	0.169	0.152	0.611
S_3(S)	72.218	33.686	47.160	0.195	0.229	0.041	0.166	0.148	0.600
S_4(S)	76.421	35.808	50.131	0.206	0.252	0.044	0.176	0.159	0.636
S_5(S)	75.250	35.202	49.283	0.203	0.242	0.043	0.173	0.156	0.625
S_6(S)	76.380	35.646	49.904	0.206	0.258	0.044	0.175	0.159	0.633
S_7(S)	72.974	35.175	49.245	0.204	0.257	0.043	0.173	0.156	0.626
S_8(S)	71.420	33.50	46.90	0.193	0.231	0.041	0.164	0.148	0.593
S_9(S)	75.080	35.056	49.078	0.203	0.237	0.043	0.172	0.156	0.622
S_{10}(S)	78.410	36.512	51.117	0.212	0.257	0.045	0.179	0.163	0.647
Average	**74.192**	**34.772**	**48.681**	**0.201**	**0.243**	**0.043**	**0.171**	**0.154**	**0.618**
	Water								
S_1(W)	14.839	6.840	9.576	0.040	0.049	0.009	0.034	0.033	0.123

S_2(W)	13.757	6.572	9.201	0.037	0.045	0.008	0.033	0.029	0.119
S_3(W)	15.012	7.212	10.097	0.041	0.049	0.009	0.036	0.033	0.130
S_4(W)	11.784	5.689	7.965	0.031	0.039	0.007	0.028	0.025	0.101
S_5(W)	12.533	6.013	8.412	0.034	0.041	0.008	0.039	0.029	0.140
S_6(W)	12.287	5.916	8.282	0.033	0.039	0.008	0.029	0.029	0.105
S_7(W)	12.190	5.774	8.084	0.033	0.038	0.007	0.029	0.025	0.105
S_8(W)	13.030	6.214	8.699	0.035	0.041	0.008	0.030	0.029	0.108
S_9(W)	12.654	6.018	8.425	0.034	0.042	0.008	0.035	0.029	0.127
S_{10}(W)	13.262	6.284	8.798	0.036	0.043	0.008	0.031	0.029	0.112
Average	**13.145**	**6.253**	**8.754**	**0.036**	**0.043**	**0.008**	**0.033**	**0.029**	**0.117**
World Average [14, 16].	**370**	**59**	**84**	**1**	**1**	**0.07**	**0.34**	**0.29×10^{-3}**	**1.16×10^{-3}**

190 **Table-3**: Average value of activity and radiological hazards with Correlation Coefficient of
191 radionuclides in the by ten sediment and water sample of Halda River at Chittagong in
192 Bangladesh.

Objectives	Average of the values obtained from ten samples of each for same location		World Average
	Activity Concentration (AC.) (Bq/Kg)		
Radioactive Material	Sediment	Water	
Radium, (^{226}Ra)	14.76±2	2.6±0.48	35
Thorium, (^{232}Th)	25.6±3.33	3.85±0.92	30
Potassium, (^{40}K)	299.5±29.95	68±8.41	400
Radiation Indices	Sediment	Water	World Recommended
Equivalent of Radium, Raeq. (Bq/Kg)	74.192	13.145	370
Outdoor Absorbed Dose, D(out) (nGy/h)	34.772	6.253	59
Indoor Absorbed Dose, D(In) (nGy/h)	84.681	8.754	84
Hazard of Outdoor, H(out)	0.201	0.036	1
Hazard of Indoor, H(In)	0.243	0.043	1
Outdoor Annual Effective Dose , E(Out) (mSv/y)	0.044	0.008	0.07
Indoor Annual Effective Dose of , E(In) (mSv/y)	0.172	0.033	0.34
External Excess Lifetime Cancer Risk, ELCR(out) ×10-3	0.155	0.029	0.29
Internal Excess Lifetime Cancer Risk, ELCR(in) ×10-3	0.619	0.117	1.16

Our Average Findings (From Table-1 & Table-2, Shown in Table-3) shows that natural radioactivity in the Halda River is well within safe limits according to world recommended value which are shown by figures (Shown in Table-3, Fig.2, Fig.3, And Fig.4).

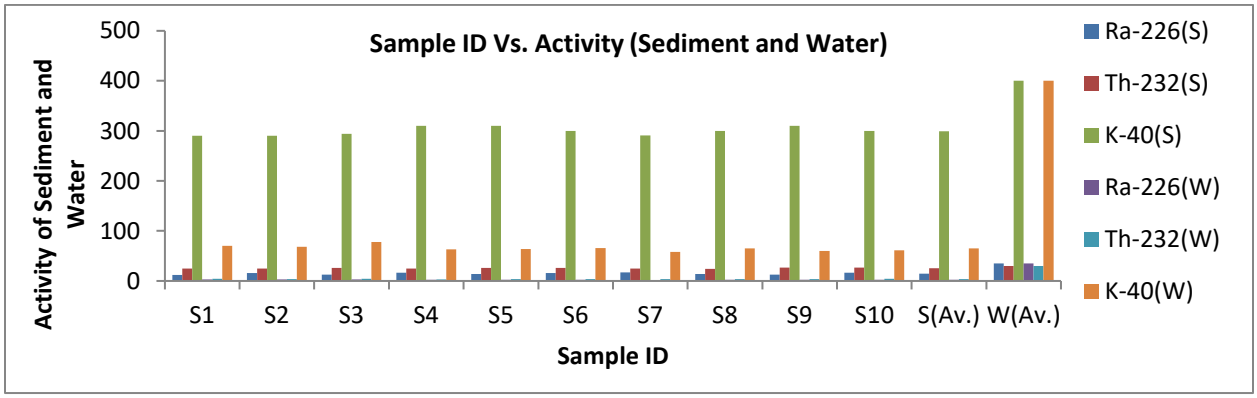


Fig.2: Sample ID vs. Activity of Sediment and Water

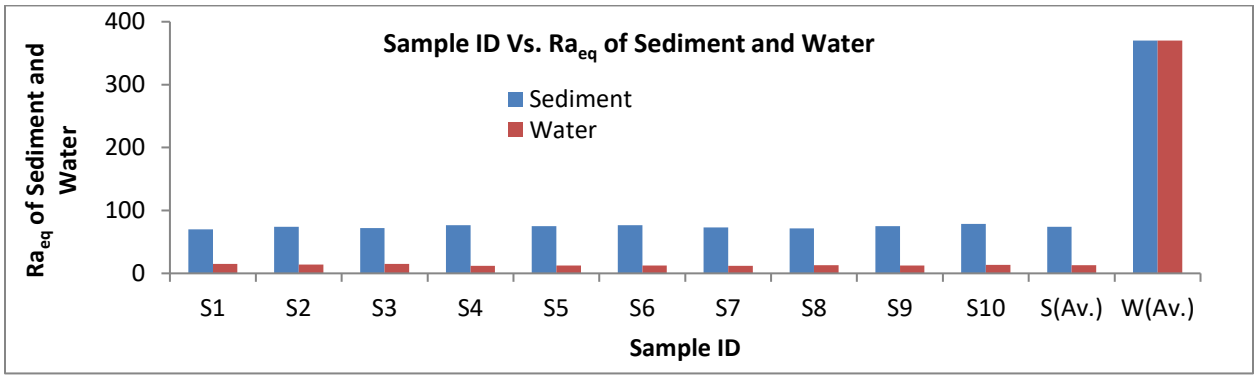


Fig.3: Sample ID vs. Ra_{eq} of Sediment and Water

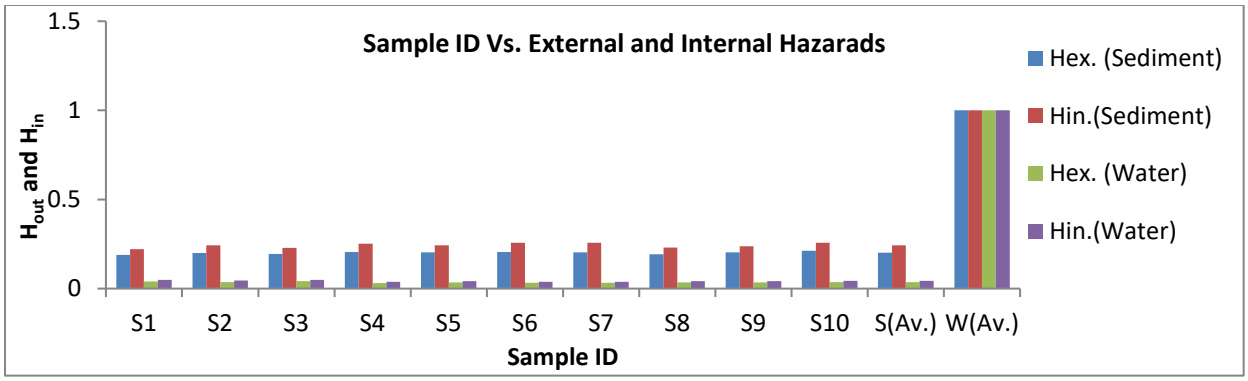
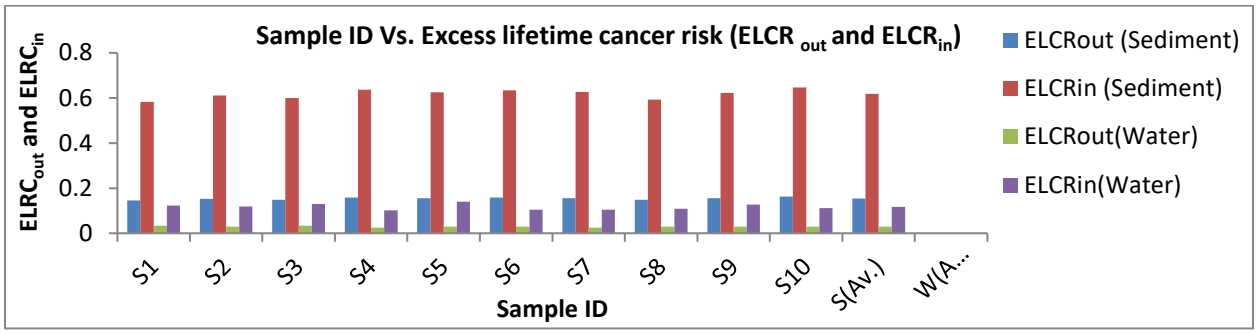


Fig.4: Sample ID vs. Outdoor and Indoor Hazards

205 **3.2 Assessment of Risk**

206 Our calculated average Raeq values in sediment (74.192 Bq/kg) and water (13.145 Bq/kg) are
207 below the world average 370 Bq/kg. Calculated external hazard index (H_{ex}) is below the
208 threshold of 1.0, indicating negligible radiation risk for health and biodiversity of Halda.
209 Furthermore, the absence of ^{137}Cs in our tests confirms that there has been no recent
210 anthropogenic radioactive contamination in the area, as shown in the figure below, compared to
211 global standards (Shown in Table-3, Fig.5).



212

213 Fig.5: Sample ID vs. Excess lifetime risk ($ELCR_{out}$ and $ELCR_{in}$)

214

215 **3.3 Statistical Investigation**

216

217 **Pearson correlation matrix (Sediment and Water Samples)**

Sediment	^{226}Ra	^{232}Th	^{40}K	Ra_{eqa}	D_{out}	D_{in}	H_{ex}	H_{in}	E_{out}	E_{in}	$ELCR_{out}$	$ELCR_{in}$
^{226}Ra	1.00											
^{232}Th	**-0.07**	1.00										
^{40}K	0.03	0.38	1.00									
Ra_{eqa}	0.61	0.63	0.57	1.00								
D_{out}	0.75	0.54	0.51	0.96	1.00							

D_{in}	0.75	0.54	0.51	0.96	1.00	1.00						
H_{out}	0.74	0.58	0.46	0.95	1.00	1.00	1.00					
H_{in}	0.92	0.26	0.20	0.79	0.91	0.91	0.91	1.00				
E_{out}	0.75	0.54	0.51	0.96	1.00	1.00	0.99	0.91	1.00			
E_{in}	0.75	0.55	0.50	0.95	1.00	1.00	1.00	0.91	0.99	1.00		
$ELCR_{out}$	0.74	0.55	0.51	0.96	1.00	1.00	0.99	0.90	1.00	0.99	1.00	
$ELCR_{in}$	0.75	0.55	0.49	0.95	1.00	1.00	1.00	0.91	0.99	1.00	0.99	1.00

218

219

Water	^{226}Ra	^{232}Th	^{40}K	Ra_{eqa}	D_{out}	D_{in}	H_{ex}	H_{in}	E_{out}	E_{in}	$ELCR_{out}$	$ELCR_{in}$
^{226}Ra	1.00											
^{232}Th	0.46	1.00										
^{40}K	0.65	0.36	1.00									
Ra_{eqa}	0.83	0.79	0.80	1.00								
D_{out}	0.80	0.74	0.86	0.99	1.00							
D_{in}	0.80	0.74	0.86	0.99	1.00	1.00						
H_{ex}	0.81	0.81	0.79	1.00	0.99	0.99	1.00					

H_{in}	0.93	0.69	0.79	0.98	0.96	0.96	0.96	1.00				
E_{out}	0.78	0.62	0.78	0.89	0.88	0.88	0.89	0.89	1.00			
E_{in}	0.57	0.24	0.39	0.47	0.49	0.49	0.50	0.54	0.60	1.00		
$ELCR_{out}$	0.78	0.62	0.78	0.89	0.88	0.88	0.89	0.89	1.00	0.60	1.00	
$ELCR_{in}$	0.58	0.25	0.38	0.48	0.49	0.49	0.51	0.55	0.61	1.00	0.61	1.00

Pearson correlation matrix analysis [08] (by using Table-1 and Table-2) shows a negative **(-0.07**) relationship between ^{226}Ra and ^{232}Th, indicating their distinct mineral sources and geographical distribution. The positive relationship between ^{226}Ra and ^{40}K in water indicates their similar solubility and mobility in aqueous solutions.

In view of the above discussion, we can say that both the sediment and water of the Halda River are potential radiation sources whose source may be old rocks because Halda is a rocky mountain geological river. But the values obtained in our current study do not pose any risk for the time being because they are much lower than the international tolerable standards.

Analysis of Principal Component (PC) and Cluster (CA)

According to Table-1 and Table-2, the activity concentration and radiological hazards obtained in our study were analyzed by PCA (Multivariate Statistical Technique) and cluster analysis (Ward's method), and the results are presented below along with their analysis. This helped us to find the source of radiation along with the reasons for the similarities and differences between the ten sediment and ten water samples we collected.

Principal Component Analysis (PCA) for Sediment and Water Samples by using Multivariate Statistical Technique [08]

Sample Name	Radionuclide and Hazards indices	PC1	PC2	Eigenvalue For		Variance % For		Probability of Radiation Source
				PC1	PC2	PC1	PC2	
Sediment	^{226}Ra	0.615	0.745	4.94	1.25	82%	13%	Looking at the values of PCA1, we can say Due to rock erosion, since the Halda River is a mountain river, there may be mineral sands and other natural causes.
	^{232}Th	0.842	-0.212					
	^{40}K	0.785	-0.324					
	Ra_{aqe}	0.965	0.184					
	D_{out}	0.982	0.082					
	H_{ex}	0.979	0.092					
	H_{in}	0.795	0.585					
Water	^{226}Ra	0.815	0.456	5.44	1.07	70%	17%	
	^{232}Th	0.745	-0.512					
	^{40}K	0.835	-0.214					
	Ra_{aqe}	0.985	0.054					
	D_{out}	0.992	-0.012					
	H_{ex}	0.988	-0.045					
	H_{in}	0,954	0.231					

PCA1 (about 82%) which implies that there is a strong effect between radionuclide and hazard Ra_{aeq}, D_{out} , H_{ex}, and H_{in}. Also, ^{232}Th and ^{40}K exhibit different behavior in PCA2 (about 13%). Therefore, according to PCA1, we can say that the **sediment** hazard parameters have a highly correlated with each other.

On the other hand, we got PCA1 for **water** is 70%, that means the radiation dose originates from radionuclides dissolved in the water, and PCA2 is 17%, indicating that discrete change in ^{232}Th and ^{40}K.

Analysis of Cluster (CA)

Using Ward's method, we can analyze the data from our 10 samples and divide them into three main clusters.

In the case of **sediment**, (S_1, S_3, S_8) are **cluster-1** which indicates very low radioactivity, **cluster-2** (S_2, S_7) are medium and (S_4, S_5, S_6, S_9, S_{10}) are samples with high radioactivity of **cluster-3**. However, it is noted that S_{10} shows the highest level of radioactivity in the sample. In the case of **water**, (S_1, S_3) **cluster-1** in which the concentration of radionuclides is slightly

higher, **cluster-2** (S_4, S_6, S_7) have low levels and **cluster-3** sample elements (S_2, S_5, S_8, S_9, and S_{10}) have medium levels (Shown in Fig.2).

In the overall analysis, we can say that from the correlation matrix and PCA, there is a strong positive relationship between ^{226}Ra and almost all hazard parameters.

Again, in the cluster analysis, we see that S_{10} and its nearby S_4, S_5, S_9 have much higher radioactivity, which is much lower than the global standard.

Therefore, in the above discussion, we can say that differences in radioactivity are observed in sediment and water due to geography and water flow.

4. Conclusions

In our current research, we collected ten quantitative sediment and water samples from the same location, approximately one kilometer apart, within a total area of fifteen kilometers, along the banks of the Halda River, the only freshwater fish breeding ground in North Chittagong, South Asia, and the collected samples were measured using the HPGe detector at the Bangladesh Atomic Energy Commission's Radioactivity Testing and Monitoring Laboratory (RTML) in Mongla to see radiation level (activity concentrations of naturally occurring primordial radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in sediment and water samples), if there is a threat to the biodiversity of the Halda River and any health risks to the people depend on it. But the great joy is that in our current research work, we have found much lower levels of radiation than the internationally permitted levels. Also, we calculated the radium equivalent average activity (Ra_{eq}), the average absorbed dose rate (internal and external), annual effective dose (internal and outdoor), the external and internal hazard indices and the values of excess lifetime cancer risk (ELCR). Notably, parameters values were less than the word average value. So, the study area's natural radioactivity does not appear to pose an immediate health risk to the biodiversity of the Halda River and dependent people. So, we can say that people living in the surrounding areas through its regional protein source in our study area are not at any risk of cancer for now. Also, there is no any risk to human life as a result of drinking processed water supplied by Chittagong WASA from the Halda River. Another important point is that we did not find the existence of

^{137}Cs in our research work. Also, calculated the correlation, Principal Component Analysis (PCA) and Cluster Analysis (CA) between exposure to radiation and associated risks is significant for both Halda River sediment and water, indicating that the geochemical and geological patterns of radionuclides influence risk assessment methods in both sediment and water which is proven by the results obtained in our statistical analysis. However, we would like to draw the attention of our responsible people to the fact that due to natural and artificial region day by day, radiation may increase in the future and affect the biodiversity of Halda and the people lives depend on it. Therefore, we need to keep a vigilant eye on all these issues.

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