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Determination of naturel radioactivity and assessment of radiation hazard indices in different sediment and soil samples from Oued Mouilah (Hammam Boughrara) in Tlemcen using a gamma spectrometry

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ABSTRACT — The aim of this study is to determine natural radioactivity (238U, 232Th, 40K) levels in soils and sediments collected from different areas of Oued Mouilah. Also other radiological hazard like the radium equivalent activity (Raeq), the absorbed dose rate, excess lifetime cancer risk and the external and internal hazard indices, which resulted from the natural radionuclides in soil were calculated using a gamma spectrometer with a scintillation detector with an efficiency (NI(Tl)) crystal 2×2 well shaped detector (Canberra Inc)

The concentration in activity of 238U, 232Th and the 40K, in all the measured samples varied from 0.09 to 1.627 Bq/Kg, 0.168 to 1.75 Bq/Kg and 0.067 to 0.504 Bq/Kg respectively in samples of soil. For samples of sediment the values varied from 0.066 to 9.19 Bq/Kg, 0.09 to 17.7 Bq/Kg, and 0.055 to 0.874 Bq/Kg for 238U, 232Th and 40K respectively.

The average activities of natural radionuclides and the level of radium equivalent activity and other hazard indices in this area were found to be lower than the world average. Our results compare fairly well with other published results.

***Keywords**: Oued Mouilah, natural radioactivity, gamma spectrometer, scintillation detector NaI(Tl),Hazard indices.*

I. Introduction

Naturally occurring primordial have been present in the environment since the formation of the earth. Predominant part of the radioactivity of soil and sediment derives from the decay of the primordial radionuclides **^{238}U, ^{235}U, ^{232}Th, ^{87}Rb** and **^{40}K** and the numerous decay products of the first three of them. Significant amount of manmade radionuclides **^{137}Cs** and **^{90}Sr** may also present in the soil and sediment as a result of testing of nuclear weapons in the atmosphere accidents such as the Chernobyl accident and the routine discharge of radionuclides from nuclear installations. The contribution of other nuclides to the total activity is negligible [1].

Radioactivity of various building materials was measured by many authors, including river sediment, soil, water, ceramics and so on in different parts of the world (Avwiri and Ononugbo (2012), Ramassamy, et al (2006) and (2009), Vesterbacka,(2007) Ononugbo et al, (2013), Agbalagba et al,(2013).

Natural waters contain small and variable quantities of alpha and beta emitters from decay of **uranium**, **thorium** and their daughters together with **^{40}K** (UNSCEAR, 1993). River sediments are also known to contain natural radionuclide and concentration of which if beyond certain limits can cause adverse health effects (Okeyode, 2012).

The concentration of naturally occurring radionuclide in river sediments is measured in an effort to better understand the spatial distribution of the radionuclide and their associated health hazards. The growth of human population and rapid industrialization has led to increased use of urban waters as sewers, compromising their uses. The discharge of industrial effluents has led inevitably to

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alterations in the quality and ecology of receiving waters (Wahid, et al., 1999) [2].

Researches on effects of radiation on humans, has shown that exposure to radiation could lead to lung, pancreas, hepatic, bone, skin and kidney cancers, cataracts, sterility, atrophy of the kidney and leukaemia (Taskin, et al., 2009). Knowledge of natural radioactivity present in river sediment, water and surface soil enables one to assess any possible radiological hazard to humans by the use of such materials [3] so the determination of natural radioactivity and the environmental hazards is very important.

The concentration activity in the sol and sediment of Oued Mouilah (Hammam Boughrara) were investigated, the radium equivalent dose and the radiological hazard indices like excess lifetime cancer risk ***(ELCR),*** absorbed dose rate ***(D),*** annual effective dose equivalent ***(AEDE),*** hazard indices **(*Hin* and *Hex*),** gamma index **(*Iγ*)** and annual gonadal equivalent dose ***(AGED)*** were calculated by using the gamma spectrometry using ***NaI(Tl)*** scintillation well-shaped detector (Canberra Inc).

II. Materials and methods:

Study of area

Oued Mouilah is situated in the North West of Algeria (Tlemcen) with area of **2650 Km^2**. It runs along **124 Km**. rinsing at an altitude of **1250 m** in Algeria then flowing into Morocco. It is ephemeral; perennial flow sets in near Oujda (Morocco) below which it re-enters Algeria near Maghnia [4].

Sample collection and preparation:

A total of 16 sediment and soil samples were collected from different places at the area of Oued Mouilah in Hammam Boughrara dam in Tlemcen, the North West of Algeria.

We dried each sample in an oven at about **110°C** to ensure that moisture was completely removed. The samples were crushed, homogenized and sieved through a **2 mm** which is the optimum size enriched in heavy minerals. Weighed samples were placed in polyethylene beaker. These beakers were completely sealed for 4 weeks in order to reach secular equilibrium where the rate of decay of the progeny becomes equal to that of the parent (radium and thorium). This step is necessary to ensure that radon gas confined within the volume and the progeny will remain in the sample [5, 6].

Table 1: Morphological characteristics of basin flow of Oued Mouilah [4].

Area (Km^2)	**2650**
Perimeter (Km)	**230**
Minimal altitude (m)	**285**
Maximal altitude (m)	**1430**
Average altitude (m)	**746**
Indices of density	**1.25**
Indices of rock slope	**0.11**
Indices of global slope (m/km)	**11.59**

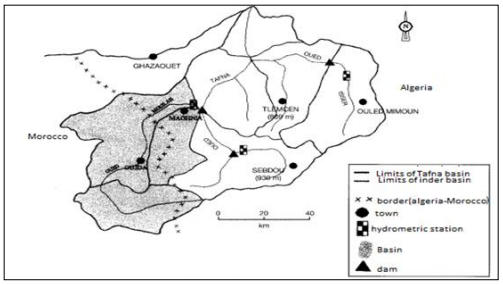

Fig. 1: the site of the basin that flows into Oued Mouilah [4].

III. Instrumentation

In order to measure the natural radioactivity of sediment and soil samples, the background spectrum was measured for **24h** in an empty cylindrical plastic container under identical geometry.

All samples were placed on the well of detector and counted for **24 h (86400 s)**, the background spectra were used to correct the net peak area of gamma rays of measured isotopes, and its spectrum was stored in a PC-based multichannel analyzer **(MCA)**. The gamma-spectrometric measurements were performed with **NaI(Tl)** well detector **2x2 inch** with a software program **Genie 2000** from Canberra has carried out the online analysis of each measured gamma ray spectrum) [7].

For the determination of the specific activity concentration of **^{226}Ra**, **^{232}Th** and **^{40}K**, the analysis of **^{40}K** was based upon its single peak of **1460.8 keV**. Whereas the analysis of **^{226} Ra** and **^{232}Th** depended upon the peaks of the daughter products in equilibrium with their

parent nuclides, the concentration of ^{226}**Ra** was determined from the average concentrations of ^{214}**Pb** (**352keV**) and ^{214}**Bi** (**609. 1120** and **1765keV**), and that of ^{232}**Th** was determined from the average concentrations of ^{212}**Pb** (**239keV**), ^{208}**Tl** (**583. 2615keV**) and ^{228}**Ac** (**338.3. 911. 969.11keV**) in each sample under study (Table2).

IV. Method of calculations:

Each radionuclide has efficiencies used to estimate its activity concentration in the samples (Table 2). The detection efficiency of the system was determined using several calculations including linear attenuation coefficient, geometric and intrinsic efficiencies for well type **2x2 NaI(Tl)** detector. The well-shaped detectors are of higher efficiency for the same volume of detector. This particular characteristic allows almost a 100 percent efficiency (so called 4π geometry). And for low gamma-emitting test sources that can fit the well shape (Knoll. 1989; Gilmore and Hemingway1995; Nicholas. 1995).

IV.1) Calculating of detector

Counting efficiency DE:

There are three factors G.I and M that affect the efficient absorption of the photons emitted by the sources. Their product is the detector counting efficiency DE (Bicron Saint Gobain Cristals. 2004).

$$DE = G \times I \times M \qquad (1)$$

where $1\text{-}G = (\pi r^2) / (4\pi R^2)$.

$I = \exp^{-(\mu_I \times d)}$. $M = 1 - \exp^{-(\mu_I \times d)}$

Linear attenuation coefficient

Calculations:

For calculation of detecting efficiencies we try to find the values of μ_I for each aluminum and **NaI** (crystal). Firstly we investigate the reference in this item and compare between them to take the main values (Hubbell and Seltzer.2004), and we calculate μ_I for mixture NaI using the following formulas:

$$\mu_m(NaI) = \sum \mu_i.w_i = (\mu_1.w_1)_{Na} + (\mu_2.w_2)_I .$$

$$\mu_l(NaI) = \mu_m(NaI).\rho$$

$$A = N / (T \times I_\gamma \times \varepsilon \times W) \qquad (2)$$

where **N** is net peak counts (background subtracted), **T** is the measured time **(sec)**, **and ε** is the efficiency of detector. I_γ is the branching ratio of gamma emission for decay mode and **W** is the sample weight.

Finally each radionuclide has gamma energy so we can calculate the **DE** of our detector and the activity concentration for the natural radionuclides (^{40}**K.** ^{232}**Th.** ^{238}**U** and ^{226}**Ra**) present in the sol and sediment samples under study (Table 2) (Eq 2) and it was estimated for the detected photopeaks in the spectra.

IV.2) Determination of radiological hazards:

IV.2.1) The Radium equivalent dose (Ra_{eq}) [8]: It was calculated through the following equation:

$$Ra_{eq}\ (Bq/kg) = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_K \qquad (3)$$

where **CRa,** C_{Th} and **CK** are the activity concentration of ^{226}**Ra,** ^{232}**Th** and ^{40}**K i**n **Bq/Kg** respectively. It has been assumed that **370 Bq/Kg** of ^{226}**Ra**, **259 Bq/Kg** of ^{232}**Th** and **1410** of ^{40}**K** produce the same gamma dose rate.

IV.2.2) The absorbed dose rates (D) [8]:

The external terrestrial γ-radiation absorbed dose rate in air at a height of about 1m above the ground was calculated by using the conversion factor of **0.042 nGyh^{-1}/Bqkg^{-1}** for ^{40}**K**, **0.462 nGyh^{-1}/Bqkg^{-1}** for ^{226}**Ra** and **0.604 nGyh^{-1}/Bqkg^{-1}** for ^{232}**Th.**

$$D\ (nGyh^{-1}) = 0.462\ C_{Ra} + 0.604\ C_{Th} + 0.042CK \qquad (4)$$

IV.2.3) Annual gonadal equivalent dose (AGED) [9]

The bone surface cells, the gonads, the bone marrow are considered as organs of interest by UNSCEAR (2000) because of their sensitivity to radiation. An increase in **AGED** has been known to affect the bone marrow, causing destruction of the red blood cells that are then replaced by white blood cells.

This situation results in a blood cancer called leukemia which is fatal. The **AGED** for the resident using such material for building by evaluated by the following equation (Avwiri, et al,2012)

AGED (μSvy^{-1}) C = 3.09 C_{Ra} + 4.18 C_{Th} + 0.314C_K (5)

where, C_{Ra}, C_{Th}, and C_K are the radioactivity concentration of ^{226}Ra, ^{232}Th and ^{40}K in soil samples or water samples.

IV.2.4) External radiation hazard (H_{ex}) [8]: It was calculated by the following equation:

Hex = C_{Ra} /370 + C_{Th}/259 + C_K/4810 $\leq$1 (6)

IV.2.5) Internal hazard index (H_{in}) [9]:

The internal exposure to **^{222}Rn** and its radioactive progeny is controlled by the internal hazard index (H_{in}) which is given by:

H_{in} = C_{Ra}/ (185) + C_{Th}/ (259) + C_K/ (4810) $\leq$1 (7)

For the safe use of a material in the construction of dwellings, index **(H_{in})** should be less than unity.

IV.2.6) Annual effective dose (AEDE) [8]:

Annual estimated average effective dose equivalent received by a member was calculated using a conversion factor of **0.7 Sv/Gy**$^{-1}$ which was used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of **20%** and **80%** for indoor .

It was determined as follows

AEDE = D x T x F (8)

where **D** is the calculated dose rate (in **nGyh**$^{-1}$), **T** is the outdoor occupancy time (**0.2x24 h x 365.25 d≈1753 hy**$^{-1}$), and F is the conversion factor (**0.710**$^{-6}$**SvGy**$^{-1}$)

IV.2.7) Excess lifetime cancer risk (ELCR) [8]:

It was calculated using the following equation

ELCR= AEDE x DL x RF (9)

where **AEDE**, **DL** and **RF** is the annual effective dose equivalent, duration of life (**70 year**) and risk factor(**Sv**), fatal cancer risk per Sievert. For stochastic effects, ICRP60 uses values of **0.05** for the public.

IV.2.8) Gamma index (Iγ) [9]:

This radiological index is used to estimate the gamma radiation hazard associated with the natural radionuclide in samples and it was estimated as follow (Avwiri et al, 2013)

Iγ = C_{Ra}/150+ C_{Th}/100+ C_K /1500) ≤ 1 (10)

V) Results and discussion:

In the current study the obtained result of activity concentration of natural radionuclides (^{238}U, ^{232}Th, ^{40}K) in soil samples were ranged from 0.09 to 1.627Bq/kg ,0.09 to 1.75 Bq/kg, and 0.067 to 0.504 Bq/kg respectively .
The activity concentration of ^{238}U, ^{232}Th, ^{40}K of sediment samples were varied from 0.108 to 9.190 Bq/kg, 0.09 to 17.7 Bq/kg and 0.055 to 0.874 Bq/kg respectively.
The radionuclide activity concentrations are shown in figure 3 for ^{238}U, in figure 4 for ^{232}Th and in figure 4 for ^{40}K.

The highest value of radionuclide activity was measured in sediment samples H2 (Fig.3), in sediment samples B2 (Fig.4) and in sediment samples C2 (Fig.5) for ^{238}U, ^{232}Th and ^{40}K respectively.
The world average concentration of ^{328}U, ^{232}Th are reported by UNSCEAR 2000 as 35, 30Bq/Kg [10] so the level of radioactivity in Oued Mouilah was permissible with world values.
In order to determinate this activity we use a gamma spectrometry technique with well shaped **2×2 NaI(Tl)** detector characterized by high efficiency and low resolution and we have calculate the efficiency of each gamma ray emitted by radionuclide (Table 2) using equation (2), after this results we can stimulate the natural radioactivity using equation (1).
The radiological and harmful hazard indices are calculated and comparable with world average, the radium equivalent dose is shown in (Fig.6), the absorbed dose rate is shown in in (Fig.7), the annual gonadal equivalent dose (AGED) is

shown in (Fig.8), the Excess lifetime cancer risk (ELCR) is shown in (Fig.9)

There were lower than the world average.

In this study we have to point out these values were not the representative values for the Algeria mentioned, but only for the region from where the samples were collected (North West Algeria «Oued Mouilah»).

VI) Conclusion:

Both of soil and sediment samples were analyzed for the present of natural radionuclides ^{238}U, ^{232}Th and ^{40}K.

The study shows that all samples of soil and sediment represent a reasonable level of activity concentration radioisotopes and the distribution of this level is not uniform, that is due to the difference of the geographical and geological nature of these regions.

The radiological hazard indices (radium equivalent activity (Ra_{eq}), the absorbed dose rates (D), Annual gonadal equivalent dose (AGED), External radiation hazard (H_{ex}), Internal hazard index (H_{in}), Annual effective dose (AEDE), Excess lifetime cancer risk (ELCR), Gamma index (Iγ)) are in the world average and within acceptable limits.

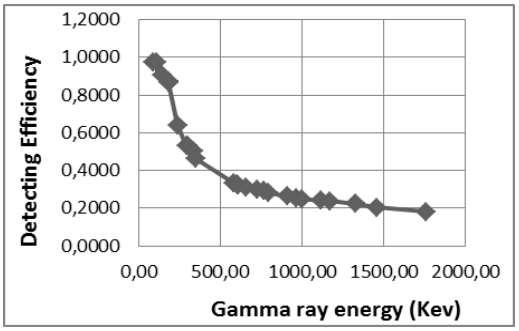

Fig.2: Efficiencies of well-Shaped 2×2 NaI (Tl) detector

Table 2: Efficiencies of well-shaped 2×2 NaI (Tl) **detector.**

Radio nuclides	Decay series	Photopeak energy (KeV)	I	M	G	DE
Ra^{226}	Bi^{214}	**609.30**	**0.975309912**	**0.336304179**	**0.983**	**0.32236311**
		1120.3	**0.996107595**	**0.246494955**	**0.983**	**0.24113213**
		1764.5	**0.996854956**	**0.185569411**	**0.983**	**0.18182091**
	Pb^{214}	**295.20**	**0.992776217**	**0.542600005**	**0.983**	**0.529571059**
		351.90	**0.993024400**	**0.47247744**	**0.983**	**0.461100321**
Th^{232}	Pb^{212}	**238.60**	**0.992701762**	**0.656766395**	**0.983**	**0.640889613**
	Ac^{228}	**338.30**	**0.993173407**	**0.51378197**	**0.983**	**0.5015525**
		911.60	**0.995649447**	**0.268633078**	**0.983**	**0.262920121**
		969.10	**0995734124**	**0.261316696**	**0.983**	**0.25577852**
	Tl^{208}	**583.00**	**0.994639419**	**0.343270417**	**0.983**	**0.33512621**
		2614.0	**0.99865091**	**0.180515668**	**0.983**	**0.174875589**
K^{40}		**1460.8**	**0.996854956**	**0.223101565**	**0.983**	**0.20225655**
Cs^{137}		**661.70**	**0.995111985**	**0.318827703**	**0.983**	**0.31187569**
Co^{60}		**1173.2**	**0.996107595**	**0.242165306**	**0.983**	**0.237121914**
		1332.5	**0.996381562**	**0.228025684**	**0.983**	**0.22333897**
U^{235}		**143.80**	**0.990421172**	**0.930190419**	**0.983**	**0.905618519**
		185.70	**0.991412088**	**0.893957571**	**0.983**	**0.871213117**

Table 3: Activity concentration of ^{238}U, ^{232}Th, ^{40}K and Ra_{eq} for soil and sediment samples

Samples	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Ra_{eq} (Bq/kg)
A1 (soil)	**1.019**	**0.385**	**0.485**	**1.607**
A2 (sediment)	**0.504**	**0.485**	**0.346**	**1.224**
B1 (soil)	**0.953**	**0**	**0**	**0.953**
B2 (sediment)	**0.212**	**17.7**	**0**	**25.515**
C1 (soil)	**0.271**	**0.168**	**0.257**	**0.531**
C2 (sediment)	**0.838**	**0.481**	**0.874**	**1.593**

D1 (soil)	1.627	0.819	0	2.798
D2 (sediment)	0.108	0.606	0.356	1.003
E1 (soil)	1.099	0.392	0.067	1.665
E2 (sediment)	0.732	0.090	0.055	0.865
F1 (soil)	0.999	0.364	0.504	1.559
F2 (sediment)	0.505	0.740	0	1.566
G1 (soil)	0.090	1.75	0	2.59
G2 (sediment)	0.875	0.997	0	2.261
H1 (soil)	0.484	0.759	0	1.570
H2 (sediment)	9.190	0.680	0	10.162
Average	1.157	1.65	0.189	3.58

Table 4: Radiation Hazard parameters (Hin, Hex and Iγ) for soil and sediment samples

Samples	$H_{ex}(10^{-}3)$	$H_{in}(10^{-3})$	Iγ	Samples	$H_{ex}(10^{-3})$	$H_{in}(10^{-3})$	Iγ
A1	4.34	7.10	0.011	E1	4.50	7.47	11.29
A2	3.30	4.67	8.44	E2	2.34	4.32	0.0058
B1	2.58	5.15	6.35	F1	4.21	6.91	10.64
B2	68.90	69.47	178.37	F2	4.22	5.59	10.77
C1	1.43	2.16	3.66	G1	7	7.24	18.1
C2	4.30	6.57	10.98	G2	6.11	8.37	15.54
D1	7.56	11.95	19.03	D2	2.71	3.00	7.02
Average	7.979	9.722	19.443				

Table 5: Radiation Hazard parameters (D, AGED,AEDE and ELCR) for soil and sediment samples

Samples	$H_{ex}(10^{-}3)$	$H_{in}(10^{-3})$	Iγ	Samples	$H_{ex}(10^{-3})$	$H_{in}(10^{-3})$	Iγ
A1	4.34	7.10	0.011	E1	4.50	7.47	11.29
A2	3.30	4.67	8.44	E2	2.34	4.32	0.0058
B1	2.58	5.15	6.35	F1	4.21	6.91	10.64
B2	68.90	69.47	178.37	F2	4.22	5.59	10.77
C1	1.43	2.16	3.66	G1	7	7.24	18.1
C2	4.30	6.57	10.98	G2	6.11	8.37	15.54
D1	7.56	11.95	19.03	D2	2.71	3.00	7.02
Average	7.979	9.722	19.443				

Samples	D (nGy^{-1})	AGED($ms.y^{-1}$)	AEDE (outdoor)(μsv^{-1})	AEDE (indoor)(μsv^{-1})	ELCR (10^{-3})
A1 (soil)	1.552	4.911	0.887	3.548	0.003
A2 (sediment)	0.540	3.691	0.662	2.650	0.002
B1 (soil)	0.440	2.944	0.540	2.160	0.002
B2 (sediment)	10.786	74.622	13.092	52.912	0.0458
C1 (soil)	0.237	1.619	0.291	1.163	0.001
C2 (sediment)	0.714	4.875	0.876	3.505	0.003
D1 (soil)	1.246	8.450	1.528	6.113	0.005
D2 (sediment)	0.431	2.982	0.529	0.605	0.002
E1 (soil)	0.748	5.055	0.916	3.666	0.003
E2 (sediment)	0.395	2.656	0.484	0.553	0.002
F1 (soil)	0.483	4.768	0.592	2.369	0.002
F2 (sediment)	0.680	4.655	0.8398	3.359	0.003
G1 (soil)	1.1	7.59	1.35	7.7	0.004
G2 (sediment)	0.989	6.750	1.212	4.848	0.004
H1 (soil)	0.682	4.670	0.837	3.347	0.0029
H2 (sediment)	4.656	31.2	5.71	22.842	0.02
Average	1.48	10.7	1.899	7.583	0.0065

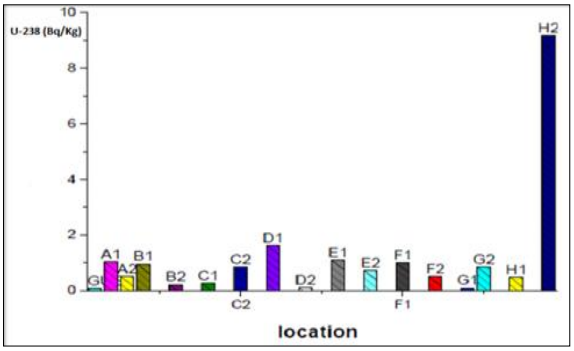

Fig. 3: the activity concentration of ^{238}U.

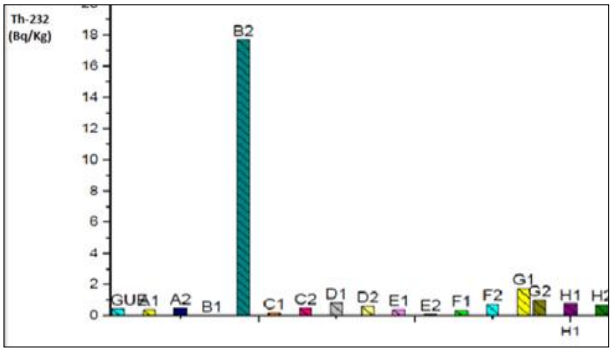

Fig. 4: the activity concentration of ^{232}Th.

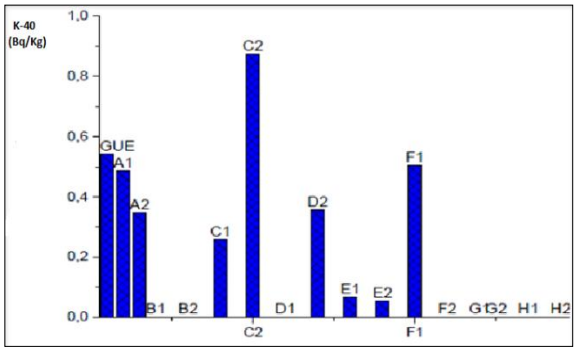

Fig. 5: the activity concentration of ^{40}K.

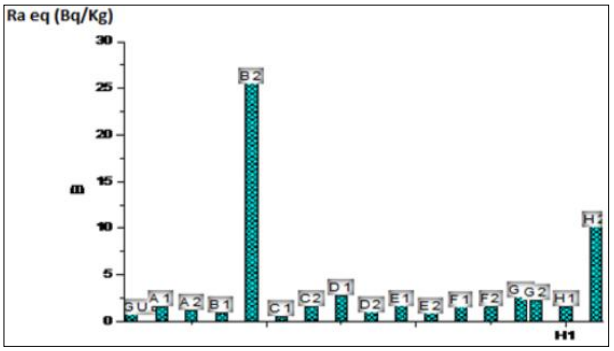

Fig. 6: the Radium equivalent dose (Ra_{eq}) of soil and sediment samples.

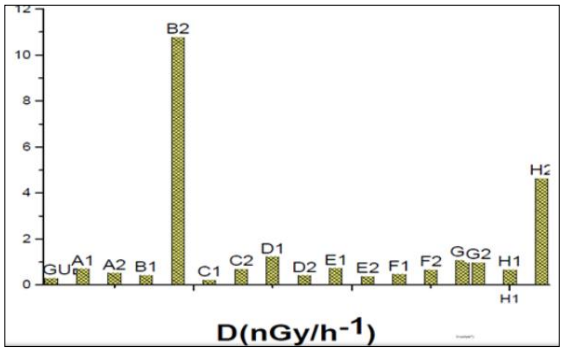

Fig.7: the absorbed dose rate of soil and sediment samples

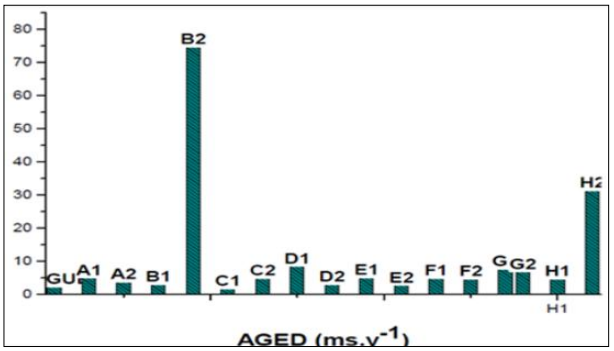

Fig. 8: the annual gonadal equivalent dose of soil and sediment samples.

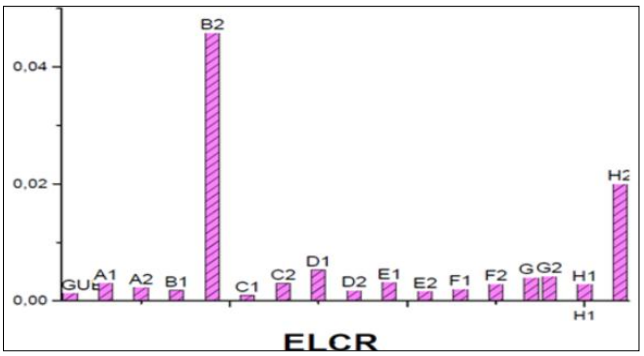

Fig. 9: the Excess lifetime cancer risk (ELCR) of soil and sediment samples.

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