

Natural Radioactivity Levels and Radiogenic Heat Production in River Sediments from Gulu and Amuru Districts, Northern Uganda

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Abstract

The activity concentrations (*AC*) of ²³⁸U, ²³²Th and ⁴⁰K in sediments from Gulu and Amuru districts, Uganda were determined using NaI(Tl) detector at Physics Department, Makerere University, Kampala to obtain; radioelement concentrations (*RC*), radiogenic heat production (*RHP*) and associated heat flow (*H_f*). The *AC* varied from (36.1 ± 2.3 - 261.2 ± 15.3) for ²³⁸U, (97.4 ± 13.5 - 334.2 ± 36.6) for ²³²Th, and (47.5 ± 3.9 - 1442.3 ± 58.9) Bq kg⁻¹ for ⁴⁰K, with averages of 89.6 ± 6.3, 168.6 ± 17.9, and 275.4 ± 14.0 Bq kg⁻¹, respectively, above the world limits of 35, 30, and 400 Bq kg⁻¹, for ²³⁸U, ²³²Th and ⁴⁰K. The *RC* varied from (2.9 ± 0.2 - 20.8 ± 1.2) ppm for ²³⁸U, (23.9 ± 3.3 - 82.0 ± 9.2) ppm for ²³²Th, and (0.2 ± 0.01 - 3.2 ± 0.1) % for ⁴⁰K, with averages of 7.1 ± 0.5 ppm, 41.3 ± 4.3 ppm, and 1.1 ± 0.05%, respectively. The ²³⁸U and ²³²Th averages were above the Earth's crust values of 3 ppm and 12 ppm, respectively. The ⁴⁰K average is below the Earth's crust value of 2.33%. The *RHP* varied from (2.5 ± 0.6 - 10.2 ± 2.6) µW m⁻³ with an average of 4.5 ± 1.1 µW m⁻³, above the world average of 4µW m⁻³. High and moderate *RHP*, each account for 50% of the samples. The overall *RHP* mainly depended on ²³²Th amounts, with 60% contribution. However, an increase in the *AC* of ²³⁸U, ²³²Th and ⁴⁰K reflected the integrated effect of *RHP*. The *H_f* varied from (19.6 ± 1.0 - 80.0 ± 4.0) mW m⁻² with an average of 35.3 ± 1.8 mW m⁻². The high *RHP* and *H_f* values indicates feasibility for geothermal exploration. This calls for further studies to validate these findings.

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1. Introduction

The natural radioactivity levels are useful in delineating the health effects of prolonged exposure to the source on human, animal and the environment [1, 2, 3]. The natural radioactivity of significance in this study are ²³⁸U, ²³²Th and ⁴⁰K [4, 5]. These isotopes have half-lives comparable to the age of the Earth (about 10¹⁰ years) and are still extant as significant heat sources in the Earth's crust [6, 7, 41:243]. The ²³⁸U, ²³²Th and ⁴⁰K have average abundances of about 3 ppm, 12 ppm, and 120 ppm in the Earth's crust [8, 9]. The decay of ²³⁸U, ²³²Th and ⁴⁰K in the Earth's crust emits energetic particles (α -particles, and β -particles) and γ -rays. Except for the energy taken by the neutrino, the whole decay is converted to heat [10]. The reactions are as illustrated in Equations (1) – (4).

$${}^{238}_{92}U \rightarrow {}^{206}_{82}Pb + 8{}^{4}_{2}H_{e} + 6e + 6anti - v + 51.667 \, MeV, (1)$$

$${}^{232}_{90}Th \rightarrow {}^{208}_{82}Pb + 6{}^{4}_{2}H_{e} + 4e + 4anti - + 42.793 MeV, (2)$$

$${}^{40}_{19}K \rightarrow {}^{40}_{20}C_{a} + anti - v + e + 1.321 \, MeV \, (89\%), (3)$$

$${}^{40}_{19}K + e \rightarrow {}^{40}_{18}Ar + v + 1.513 \, MeV \, (11\%), (4)$$

The decay of 238 U and 232 Th are the prevalent heat producers today with heat production constants of 9.52 x 10^{-5} w kg⁻¹ and 2.56 x 10⁻⁵ w kg⁻¹, respectively [11]. The 40 K is the least heat producer compared to 238 U, and 232 Th with heat production constant of $3.48 \times 10^{-9} \text{ w kg}^{-1}$ [12]. The heat produced in the mantle and crust of the Earth during the decay of ²³⁸U, ²³²Th and ⁴⁰K is importantly called the Radiogenic Heat Production or *RHP* [9,13]. The *RHP* determines the heat flow in the basement rocks from which river sediments are formed and has been used for the assessment of geothermal resources in many parts of the world [14, 15]. River sediments pile and transfer contaminant within the geographic region and is considered the environmental host of the waste discharged by natural or man-made processes in our surroundings [6, 16, 17]. Most river sediments are formed when the rock and organic materials are broken into small pieces by fluvial means which enhance the levels of natural radioactivity in river sediments [17, 18]. The RHP is influenced by the sediments' lithological and geochemical features as reported by [9, 19]. Natural radioactivity has been intensively studied [12, 15, 17, 20]. Sathish and colleagues [12] recently assessed the natural radiation hazards and function of heat production rate in lake sediments, India using NaI(Tl) detector. The study attributed the radioactive heat production ranges of $0.06 - 0.23 \ \mu W \ m^{-3}$ to the high concentration and density of radioactive minerals in the lake sediments. In determination of natural radioactivity concentration and radiogenic heat production in selected quarries in Ondo State, Nigeria, by Gamma Surveyor, Asere and colleagues [15] reported total heat production and heat flow ranges of $0.97 - 5.3 \mu$ W m⁻³ and 7.63 - 42.12 mW m⁻², respectively. The study recommended further probe for potential geothermal exploration. In 2018, Ghania, Fatima & Mourad [17] determined the levels and effects of natural radionuclides in sediment banks of Rhumel River (Northeast Algeria) using high resolution HPGe detector. Results of this study shows that the mean AC were; 26.64 Bq Kg⁻¹ for ²²⁶Ra, 25.95 Bq Kg⁻¹ for ²³²Th and 164.50 Bq Kg⁻¹ for ⁴⁰K. The variations in the AC were attributed to inhomogeneous samples composition.

Natural radioactivity levels and associated radiation hazards in Nile river sediments, Egypt were also investigated using NaI(Tl) detector by El-Taher & Adel [16]. The higher concentrations of the radionuclides were attributed to industrial wastes. Biira, Kisolo & D'ujanga [20] determined the concentration levels of radon in Tororo and Busia districts, Eastern Uganda using activated charcoal canisters and NaI(Tl) detector. It was found that the mean radon concentrations values were below the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) and International Commission of Radiological Protection (ICRP) recommended radon action level of 200 Bq m⁻³. Similar assessments for the dormitories of secondary schools in Otuke district, Uganda based on survey meters and activated charcoal canister were made by Oruru, Todo & Kisolo [10]. The measured radon concentrations were below the World Health Organization (WHO) action level of 100 Bq m⁻³. A study by [33] in selected quarries in Northern Uganda using NaI(Tl) detector, reported the average AC as: 57.1 \pm 4.7 for ²³⁸U, 122.3 \pm 11.1 for ²³²Th, and 914.2 \pm 30.3 Bq kg⁻¹ for ⁴⁰K, well above the global limits. It was concluded that the workers and inhabitant were exposed to significant background radiations from quarries. The determination of RHP in river sediments for the assessment of geothermal resources in the districts of Gulu and Amuru, Uganda have not yet been done, but very significant. A study by Ojara & Odongkara [21] reported the needs to develop geothermal energy as an alternative energy source to hydro and others to meet the energy demand of the locals.

This paper, therefore, reports the results of: (i) the *AC* of ²³⁸U, ²³²Th and ⁴⁰K; (ii) the *RC* of ²³⁸U, ²³²Th and ⁴⁰K; (iii) the *RHP* and (iv) H_f in 30 representative sediment samples from Gulu and Amuru districts, Northern Uganda, in order to estimate the area's geothermal potential. The results of this study may be used by Uganda Geological Survey and Mines (*UGSM*) for geothermal exploration and eventual development of geothermal energy project. Also, Gulu and Amuru districts could use these findings to raise awareness on the viable local geothermal sources and support its development as a proxy for energy source. However, this study did not consider the energies produced by α and β particles which are significant radiation dose to lungs and other respiratory organs.

1.1. Description and Geology of Study Area

This study was conducted in the districts of Gulu (2°10' and 03°6' N, 32°10' and 33°30' E) and Amuru (2°35' and 03°45' N, 31°30' and 32°10' E) in Northern Uganda, illustrated in Figure 1 and Table 1, respectively. The districts were chosen due to the rapid growth in agriculture, commerce, tourism, industry and lucrative trade links with South Sudan. The selected areas had total population of 187,124 persons (constituting 30% of the population of the two districts). The relief consists of complex low landscape with relatively uniform topography and altitude ranging from 775 - 1106 m above sea level. The temperature varied from (18 – 30) ⁰C [21]. The unit is mainly composed of Neoarchean gneissose-migmatic rocks and granitoids with the quartzo-feldspathic veins cut by colored layers of granite rich in Quartz (*SiO*₂), plagioclase (*NaAlSi*₃*O*₈), feldspar (*KAlSi*₃*O*₈), iron oxide (*Fe*₂*O*₃), opaque minerals, biotite K (*Mg*, *Fe*⁺²)₃(*Al*, *Fe*⁺³) *Si*₃*O*₁₀(*OH*, *F*)₂, and muscovite *KAl*₂(*AlSi*₃) *O*₁₀(*OH*)₂ [22]. Due to the up and down wrapping of the basement rocks consisting of uniform rock structure, the drainage pattern formed network of rivers which drain their waters into Albert Nile, Victoria Nile and Achwa River at acute angles [21]. The network of rivers is analogous to branches of trees and formed V-shaped patterns. The said rivers offer immense environmental and socio economic values and services to the

population such as: crop farming, brick-laying, pottery, sand mining, irrigation development, commercial tree seedlings, horticulture and fish farming. The rivers traversing the two districts include among others: Unyama, Ayugi, Omee, Aswa, Tangi, Ayago, Oitino, Abera, Ziola, Awicpalaro, and Tochi. The reader is referred to illustration in Figure 1 for further details.



Figure 1: Map of Gulu and Amuru districts showing selected rivers and sample locations [23].

2. Materials and Methods

2.1 Sample Collection and Preparation

The sample locations, as in Table 1 and Figure 1, were identified from the Nile river major tributaries in Gulu and Amuru districts, 30 km apart in which river sand were mined, highly populated, and representative enough of the major river sediments types in the area. Different rivers dissecting heterogeneous geological settings were chosen to obtain representative number of sediment samples with various physical, chemical and geochemical behaviors to better understand the distribution pattern of 238 U, 232 Th and 40 K in the sediment samples as stated by Laith *and colleagues* [9]. At each site, a sample point from near the river bank of 10 – 100 cm was carefully selected where the survey meters indicated high dose rate as pointed in the study by Keser *and colleagues* [8], Oruru, Todo & Kisolo [10], and Atibu *and colleagues* [24]. The distance of 10 – 100 cm from the river bank was used so that samples could be taken from undisturbed area (closed system).

This was to limit the factors that could obscure the experimental outcome. Coordinates were recorded by high sensitivity (etrex) Garmin Global Position Satellite (*GPS*). A more complete details of the sampling sites and GPS coordinates can be found in Table 1.

Site	GPS Coordinate			Elevation	Population
ID	Latitude	Longitude	Sampling Site	(<i>m</i>)	(persons)
S_1	30 30' 53.2506" N	320 5' 13.002" E	Bibia	980	7,934
S_2	30 24' 53.0094" N	320 8' 30.7536" E	Pacilo	775	4,143
S_3	30 19' 15.6864" N	320 2' 29.5146" E	Pogo	783	5,968
S_4	30 14' 1.5174" N	320 4' 33.4482" E	Palukere	808	2,633
S_5	30 16' 35.8458" N	320 12' 1.7598" E	Okidi	811	3,515
S_6	30 9' 49.7766" N	320 2' 0.1896" E	Pogo	783	5,968
S_7	30 8' 6.1116" N	320 7' 11.6538" E	Parubanga	1010	6,696
S_8	30 4' 28.689" N	320 8' 12.9876" E	Parubanga	903	6,696
S_9	30 1' 31.1766" N	320 5' 52.5516" E	Kal	953	4,980
S_{10}	20 55' 25.824" N	320 7' 51.204" E	Coke	1036	4,151
S_{11}	20 52' 25.1326" N	310 39' 40.4064" E	Pailyec	1073	15,042
S_{12}	20 51' 43.6356" N	310 44' 22.2072" E	Pailyec	1039	15,042
S_{13}	20 50' 59.193" N	310 59' 46.7052" E	Toro	1090	5,664
S_{14}	20 46' 2.9208" N	310 57' 48.0522" E	Paminrut	783	9,194
S_{15}	20 48' 50.8104" N	320 1' 35.4714" E	Pamuca	1049	5,383
S_{16}	20 59' 32.7336" N	320 31' 18.2598" E	Paibona	972	5,480
S_{17}	20 53' 27.3222" N	320 31' 49.8648" E	Kal Alii	1006	6,614
S_{18}	20 38' 10.6836" N	320 40' 49.1874" E	Lukwor	1052	6,390
S_{19}	20 28' 32.9484" N	320 25' 47.6112" E	Patek	1062	4,835
S_{20}	20 33' 59.691" N	320 22' 19.6428" E	Paidwe	1071	8,663
S_{21}	20 38' 28.7622" N	320 18' 49.4244" E	Labwoc	1090	5,676
S_{22}	30 4' 9.1554" N	320 26' 13.6824" E	Owalo	998	6,189
S_{23}	20 53' 47.0754" N	320 18' 28.9584" E	Oding	1043	3,524
S_{24}	20 38' 18.7152" N	320 15' 30.9816" E	Kal	1106	5,658
S_{25}	20 43' 17.835" N	320 16' 17.8422" E	Abwoc	1083	3,929
S_{26}	20 44' 44.235" N	320 39' 9.7374" E	Binya	1017	10,122
S_{27}	20 48' 31.0608" N	320 24' 44.694" E	Anyaya	1081	4,009
S_{28}	20 52' 28.0734" N	320 13' 22.44" E	Pabwo	1031	3,458
S_{29}	20 37' 58.962" N	320 29' 31.4334" E	Parwech	1005	4,384
S_{30}	20 37' 58.962" N	320 12' 13.2258" E	Onyona	1097	5,184

Table 1: Sampling site and GPS coordinates of the sampled rivers in Gulu and Amuru district.

A sample of about 1 kg wet weight was then manually collected from each site at a depth of 5 - 10 cm from the river bed using a hand shovel within a sampling area of 1 m² [5, 7, 18, 25, 26]. The 1 kg sample was then put into a polyethene bag, sealed, and labeled with a file name to avoid cross contamination until analyzed [9]. The process was repeated for all the selected rivers until thirty (30) representative sediment samples were obtained. Stones and organic materials were removed from the sediment samples and then oven dried to 120 °C for nine hours until constant weight was attained [28]. This was done to ensure that moisture was completely removed from the sample in order to avoid clamping of the sample particles during crushing [6, 16, 29]. The sample was then grounded in a mortar using a pestle to powder form in order to increase the surface area and sieved through a 200 µm mesh sieve to homogenize the contents [4, 17, 30]. The sieved sample was then filled in air tight standard 500 ml plastic Marinelli beaker (to minimise radon leakage); labeled with a file name and weighed using a digital meter to determine the dry mass. The digital meter registered an average of 570 ± 0.1 g for each prepared samples [31, 32]. The air tight sealed sample was stored for four weeks (30 days), approximately 7 times the half-lives of the daughter nuclide before counting to obtain secular equilibrium between 226 Ra and decay products of 222 Rn (214 Pb & 214 Bi) [1, 2, 6, 33]. All the samples were then analyzed to show the geologic attributes of the study area.

2.2 Sample Analysis

2.2.1 Detector System and Calibration

The gamma ray detector used in this study is a Gammadata Matteknik (GDM) 20 Teledye NaI Iodide model, (version 1.2, 1997). The NaI(Tl) is the most widely used detector because it is the most efficient inorganic scintillator, cheap and readily available. The detector unit has a height of 94 cm, diameter 42 cm and weighs 350 kg. It has a 3 x 3 – inch active area with 14-pin Photo Multiplier Tube (*PMT*). It offers an energy resolution of <7% Full Width at Half Maximum (FWHM) at the 662 keV ¹³⁷Cs line with cylindrical lead shielding of 10 cm thick and removal pellets of 315 kg. The detector unit consists of a high voltage supply D234 of 0 - 1500 V which is connected to a power source of $\pm 15V$, 500 mA. The whole system is connected to a computer interface, IBM computer-IAEA16867 (SS01/005), which has an inbuilt autoDAS software, (version 3.16). The energy and efficiency calibrations of the detector were done by common radioactive ¹⁵²Eu standard source from Gammadata Matteknik AB, Sweden (model number 26-Jan-1993 Europium-152). The source weighs 54 g with activity of 1370 ± 80 Bq and half-life of 3.2 years. The ¹⁵²Eu is a frequently used calibration source due to its wide range of gamma energies from 122 keV to 1460.8 keV [8, 9]. A spectrum for ¹⁵²Eu was collected for 5000 seconds and saved in the computer hard disc [20]. The peaks of known energies were analyzed for energy and efficiency calibrations. The energy resolution of the detector was also determined and found to be 6.2% at 662 keV. The background of the laboratory environment was determined by placing an empty Marinelli beaker on the detector with the same period and operating voltage used to determine the energy spectra of the samples [20, 32]. The background spectrum was later subtracted from the gamma ray spectra of the samples to obtain the net counts.

2.2.2 Activity Concentrations of ²³⁸U, ²³²Th and ⁴⁰K

When the analyzed sediment sample attained a state of secular equilibrium between ²³⁸U and ²³²Th with their progenies [18], the activity concentration (*AC*) of ²³⁸U in the samples was obtained from the average energies of 351.9 keV of ²¹⁴Pb, and 609.3 keV of ²¹⁴Bi, respectively. Similarly, the *AC* of ²³²Th was obtained from the average energies of 238.6 keV of ²¹²Pb, and 583.2 keV of ²⁰⁸Tl, respectively [20, 34]. The *AC* of ⁴⁰K was determined directly from its 1460.8 keV gamma-ray peaks following the decay of ⁴⁰K [35]. Spectra peaks were analyzed one at a time using auto*DAS* command, whereby the cross was placed immediately at the left edge of the peak and *L* was typed to define the cross of the lower mark. The cross of the upper mark was similarly placed at the right edge of the same peak using command *U*. This was followed by typing *CEN* command which gave centroid, standard deviation, *FWHM*, sum between markers and rate [20, 33]. The *AC* of *i*th nuclides from the respective energy peak was computed using the Equation (5) as stated by the authors in [9, 33, 36]:

$$AC_i(Bq \ kg^{-1}) = \frac{N_e}{M_i T \varepsilon \rho}, (5)$$

where AC_i = Activity concentrations $(Bqkg^{-1})$ in the *i*th sample. N_e = net peak area of energy, e, (Bq). M_i = dry mass of the *i*th sample (kg). T = the sample measurement time (seconds). ε =efficiency of the detector (%). ρ = branching ratio (absolute gamma emission probability) (%).

2.3 Radioelement Concentrations (RC)

The *RC*, in parts per million (ppm) of 238 U, and 232 Th, and in (%) of 40 K in the sediment samples were numerically deduced from their measured *AC* values by the use of the Equations (6), (7) and (8) reported by [36, 37, 38], as;

$$U(ppm) = \left(\frac{C_U M_U}{N_A I n 2}\right) t_{1/2}, (6)$$
$$Th (ppm) = \left(\frac{C_{Th} M_{Th}}{N_A I n 2}\right) t_{1/2}, (7)$$
$$K(\%) = \left(\frac{C_K M_K}{N_A I n 2}\right) t_{1/2}, (8)$$

where: C_U , C_{Th} , and C_K are activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K, respectively in Bq g⁻¹, M_U , M_{Th} , and M_K are molecular weights of ²³⁸U, ²³²Th and ⁴⁰K, respectively in g mol.⁻¹, N_A is Avogadro's number (6.02 x 10²³) and $t_{1/2}$ is the half-life of each of the natural radionuclides expressed in seconds as reported by Okeyode [39].

2.4 The Radiogenic Heat Production (RHP)

The *RHP* defines the amount of heat liberated in a unit time per unit volume of sediment by the decay of unstable radioactive isotopes; in unit of μ W m⁻³. The *RHP* is high in the upper crust and always low at the mantle reaching 1 – 2 μ W m⁻³. Murugesan *and colleagues* [13], and Okeyode [39] noted that high *RHP* are targets for geothermal exploration and production. The *RHP* in the studied samples was determined by Equation (9) as proposed by Rybach [19]. It should be appreciated that this model has been widely used and accepted beyond question by various scholars, including among others Sokari *and colleagues* [36], Omosule & Adelowo [38], and Murugesan *and colleagues* [40].

$$RHP (\mu W m^{-3}) = \beta \rho (f_U C_U + f_{Th} C_{Th} + f_K C_K), (9)$$

where: β is constant (10⁻⁵), ρ is the average samples density (kg m⁻³), C_U , and C_{Th} are ²³⁸U and ²³²Th concentrations (weight *ppm*), and C_K is the total ⁴⁰K concentrations (weight %). The numerical constants f_U , f_{Th} , and f_K used in the conversion were obtained by Rybach [19] and later on used in the study by Sokari *and colleagues* [36]. The constant for ²³⁸U (9.52) is more than double the constants for ²³²Th (2.56) and ⁴⁰K (3.48), reflecting the significant role that uranium has in producing heat compared with thorium or potassium [9]. The amount of heat that flows per second across a square meter of surface (H_f) was calculated using Equation (10) as introduced by Turcotte & Schubert [41] and later used for river sediment samples by Asere *and colleagues* [15] as;

$$H_f = -k\frac{dT}{dz} = \frac{RHP}{A}(M), (10)$$

where H_f is the heat flow in (mW m⁻²), k is thermal conductivity (Wm⁻¹K⁻¹), $\frac{dT}{dz}$ is geothermal gradient (K km⁻¹) M is the mass of the mantle plus crust (4.0 x 10²⁴ kg), and A is the Earth's surface area (5.1 x 10⁸ km²). On the basis of geochemical studies, the core can't contain a significant fraction of the heat-producing elements. Hence, the mass of the mantle is used instead of the Earth. According to Turcotte & Schubert [41], the mean H_f for all continents is 65 ± 1.6 mW m⁻² with 37 mW m⁻² as the fraction attributed to the decay of ²³⁸U, ²³²Th and ⁴⁰K. The remaining 28 mW m⁻² was attributed to basal heating of the continental lithosphere by mantle convection. Determination of heat flow by Fourier's law ($-k\frac{dT}{dz}$) is beyond the scope of this work which requires further studies. The negative sign is needed to account for the direction of the heat flow; if temperature increases in the downward direction of the z-axis, the flow of heat from high to low temperature is upward.

3. Results

Table 2: The AC, RC and RHP of 238 U, 232 Th and 40 K in the sediment samples.

ID	238 U (<i>Bq kg</i> ⁻¹)	232 Th (<i>Bq kg</i> ⁻¹)	40 K (<i>Bq kg</i> ⁻¹)	²³⁸ U (<i>ppm</i>)	²³² Th (<i>ppm</i>)	Th/U	⁴⁰ K (%)	RHP($\mu W m^{-3}$)
S_I	68.3 (5.6)	130.0 (27.6)	287.0 (12.3)	5.4 (0.5)	32.0 (3.8)	5.93	1.1 (0.04)	3.5 (0.9)
S_2	73.0 (5.7)	155.3 (15.9)	106.8 (10.0)	5.8 (0.5)	38.1 (3.9)	6.57	0.4 (0.03)	3.9 (1.0)
S_3	133.8 (13.8)	253.6 (27.4)	187.6 (6.2)	10.7 (1.1)	62.2 (6.9)	5.81	0.7 (0.02)	6.7 (1.7)
S_4	77.6 (6.6)	169.7 (18.1)	135.7 (8.0)	6.2 (0.5)	41.6 (4.5)	6.71	0.5 (0.03)	4.2 (1.1)
S_5	63.7 (5.9)	135.2 (19.5)	169.6 (9.1)	5.1 (0.5)	33.3 (4.8)	6.53	0.6 (0.03)	3.4 (0.9)
S_6	148.9 (5.6)	334.2 (36.6)	1442.3 (58.9)	11.9 (0.5)	82.0 (9.2)	6.90	5.5 (0.20)	8.6 (2.2)
S_7	63.9 (5.4)	138.9 (20.0)	189.9 (8.6)	5.1 (0.4)	34.1 (4.9)	6.69	0.7 (0.03)	3.5 (0.9)
S_8	88.5 (6.3)	154.5 (16.7)	247.9 (14.9)	7.0 (0.5)	36.8 (4.1)	5.26	0.9 (0.05)	4.2 (1.1)
S_9	77.9 (5.5)	132.0 (20.1)	284.9 (14.4)	6.2 (0.4)	32.4 (4.9)	5.23	1.1 (0.05)	3.7 (0.9)
S_{10}	75.0 (6.0)	124.2 (15.8)	575.0 (26.2)	6.0 (0.5)	30.5 (3.9)	5.08	2.2 (0.08)	3.5 (0.9)
S_{11}	36.1 (2.3)	151.1 (3.7)	222.2 (13.7)	2.9 (0.2)	37.1 (0.9)	12.80	0.8 (0.04)	3.1 (0.8)
S_{12}	156.2 (7.2)	288.0 (9.7)	339.9 (21.4)	12.4 (0.6)	70.6 (2.4)	5.69	1.3 (0.07)	7.6 (1.9)
S_{13}	101.7 (6.0)	181.5 (13.3)	234.0 (6.3)	8.1 (0.5)	44.5 (3.3)	5.49	0.9 (0.02)	5.0 (1.3)
S_{14}	261.2 (15.3)	319.5 (16.8)	218.2 (14.5)	20.8 (1.2)	78.4 (4.1)	3.77	0.8 (0.05)	10.2 (2.6)
S_{15}	64.7 (5.8)	128.0 (15.1)	105.2 (6.8)	5.1 (0.5)	31.4 (3.7)	6.16	0.4 (0.02)	3.2 (0.8)
S_{16}	78.6 (6.2)	114.6 (21.3)	164.8 (8.1)	6.3 (0.5)	28.1 (5.2)	4.46	0.6 (0.03)	3.4 (0.9)
S_{17}	83.7 (7.0)	162.5 (16.4)	295.9 (10.8)	6.7 (0.6)	39.9 (4.0)	5.96	1.1 (0.03)	4.3 (1.1)
S_{18}	43.5 (2.5)	101.1 (14.2)	172.8 (12.4)	3.5 (0.2)	24.8 (3.5)	7.09	0.7 (0.04)	2.5 (0.6)
S_{19}	88.3 (6.1)	161.2 (20.1)	201.5 (9.2)	7.0 (0.5)	39.5 (5.0)	5.64	0.8 (0.03)	4.4 (1.1)
S_{20}	99.9 (6.6)	195.0 (11.9)	105.5 (5.7)	8.0 (0.5)	47.8 (2.9)	5.98	0.4 (0.02)	5.0 (1.3)
S_{21}	112.3 (4.8)	198.0 (19.6)	188.5 (7.5)	8.9 (0.4)	48.6 (4.8)	5.46	0.7 (0.02)	5.4 (1.4)
S_{22}	72.7 (5.0)	134.7 (12.0)	213.7 (15.2)	5.8 (0.4)	33.0 (3.0)	5.69	0.8 (0.05)	3.6 (0.9)
S_{23}	67.0 (5.3)	134.2 (19.4)	370.4 (22.3)	5.3 (0.4)	32.9 (4.8)	6.21	1.4 (0.07)	3.5 (0.9)
S_{24}	86.7 (4.9)	164.4 (26.0)	829.1 (39.7)	6.9 (0.8)	40.3 (6.4)	5.84	3.2 (0.10)	4.5 (1.1)
S_{25}	61.4 (5.7)	115.8 (19.7)	185.5 (11.5)	4.9 (0.5)	28.4 (4.8)	5.80	0.7 (0.04)	3.1 (0.8)
S_{26}	47.1 (6.3)	97.4 (13.5)	169.6 (9.1)	3.8 (0.5)	23.9 (3.3)	6.29	0.6 (0.03)	2.5 (0.6)
S_{27}	54.8 (5.5)	108.2 (18.8)	190.7 (9.6)	4.4 (0.4)	26.5 (4.6)	6.02	0.7 (0.03)	2.9 (0.7)
S_{28}	92.9 (6.9)	161.4 (15.7)	47.5 (3.9)	7.4 (0.6)	39.6 (3.9)	5.35	0.2 (0.01)	4.4 (1.1)
S_{29}	120.0 (5.8)	228.5 (16.9)	217.3 (11.5)	9.6 (0.5)	56.0 (4.2)	5.83	0.8 (0.04)	6.0 (1.5)
S_{30}	88.6 (5.8)	186.5 (15.4)	162.4 (11.2)	7.1 (0.5)	45.7 (3.8)	6.44	0.6 (0.04)	4.8 (1.2)
Ave	89.6 (6.3)	168.6 (17.9)	275.4 (14.0)	7.1 (0.5)	41.3 (4.3)	6.09	1.1 (0.05)	4.5 (1.1)
Mi.	36.1 (2.3)	97.4 (13.5)	47.5 (3.9)	2.9 (0.2)	23.9 (3.3)	3.77	0.2 (0.01)	2.5 (0.6)
Ma.	261.2 (15.3)	334.2 (36.6)	1442.3 (58.9)	20.8 (1.2)	82.0 (9.2)	12.80	5.5 (0.20)	10.2 (2.6)

Where: Ave is average; Mi. is minimum; Ma. is maximum.

The AC, RC and RHP of ²³⁸U, ²³²Th and ⁴⁰K in the experimentally analyzed samples were computed using

Equations (5) to (9) and are given in Table 2. The results show that ²³⁸U, ²³²Th and ⁴⁰K were the only radionuclides detected in all the samples but with uneven occurrences. As shown in Table 2, the *AC* varied from (36.1 ± 2.3 - 261.2 ± 15.3) Bq kg⁻¹ for ²³⁸U, (97.4 ± 13.5 - 334.2 ± 36.6) Bq kg⁻¹ for ²³²Th, and (47.5 ± 3.9 - 1442.3 ± 58.9) Bq kg⁻¹ for ⁴⁰K, with averages of 89.6 ± 6.3, 168.6 ± 17.9, and 275.4 ± 14.0 Bq kg⁻¹, respectively. Figure 2 shows bar graphs of the *AC* of ²³⁸U, ²³²Th and ⁴⁰K in the samples. The data shows that the *AC* of ²³⁸U is higher than that of ⁴⁰K in samples S_{14} and S_{28} . Also, the *AC* of ²³²Th is highest in samples S_2 , S_3 , S_{14} , S_{15} , S_{20} , S_{21} , S_{28} , S_{29} , and S_{30} compared to ²³⁸U, and ⁴⁰K. In 70% of the samples, the *AC* were in the order ⁴⁰K > ²³²Th > ²³⁸U. The *AC* of ²³⁸U, ²³²Th and ⁴⁰K. In 70% of the samples, the algobal averages of 35, 30, and 400 Bq kg⁻¹ by factors of 4.3, 11.1, and 3.6, for ²³⁸U, ²³²Th and ⁴⁰K, respectively. The samples S_{10} (575.0 Bq kg⁻¹), and S_{24} (829.1 Bq kg⁻¹) had *AC* of ⁴⁰K higher than world average value of 400 Bq kg⁻¹ by factors of 1.44 and 2.07, respectively. It is seen that 90% of the samples had *AC* of ⁴⁰K lower than world average value of 400 Bq kg⁻¹.

As illustrated in Table 2, The *RC* of ²³⁸U, ²³²Th and ⁴⁰K varied from $(2.9 \pm 0.2 - 20.8 \pm 1.2)$ ppm, $(23.9 \pm 3.3 - 82.0 \pm 9.2)$ ppm, and $(0.2 \pm 0.01 - 3.2 \pm 0.1)$ %, with averages of 7.1 ± 0.5 ppm, 41.3 ± 4.3 ppm, and 1.1 ± 0.05% for ²³⁸U, ²³²Th and ⁴⁰K, respectively. The ranges for ²³⁸U and ²³²Th obtained in this study are above the normal continental crust ranges of 2 - 3 ppm, and 8 – 12 ppm, respectively [9, 42]. The minimum *RC* of ⁴⁰K (0.2%) is less than the crustal abundance of 2% and the maximum of ⁴⁰K (3.2%) is above the crustal abundance of 2.5%. In 29 samples (97%), the *RC* of ²³⁸U and ²³²Th were all above the normal continental crust average values of 3 ppm for ²³⁸U and 12 ppm for ²³²Th, respectively. In sample *S*₁₁, only ²³²Th had *RC* higher than the normal crustal average value. In 28 samples (93%), the *RC* of ⁴⁰K were below the crustal average value of 2.33%.



Figure 2: Bar graphs showing AC of 238 U (blue), 232 Th (red) and 40 K (yellow) in the samples.

Country/Study Area	Average AC (Bq kg^{-1})			Reference	
	²³⁸ U	²³² Th	40 K		
Italy (Calabria Rivers)	21.3	30.3	849.0	[5]	
India (Beach-Tamilnadu)	35.12	713.16	349.60	[51]	
India (Cauvery River)	5.3	34.1	401.1	[40]	
India (South Indian rivers)	9.81	36.49	742.46	[13]	
India (Cauvery and Palar rivers)	5.31	34.04	401.11	[11]	
India (Ponnaiyar river)	7.31	46.85	384.11	[25]	
Egypt (Red Sea Coast)	22.2	19.2	477.6	[30]	
Egypt (Aswan to El-Mania)	29.0	45.0	123.0	[16]	
Egypt (Minia Nile River)	46.91	32.35	255.88	[1]	
Iraq (Sulaymaniyah Rivers)	15.47	2.99	54.76	[35]	
Iraq (Tigris river-Mosul City)	13.15	24.68	258.0	[9]	
Iraq (Tigris River)	13.5	35.2	272.2	[28]	
Iraq (Tigris River Al-Amara City)	18.22	13.79	317.34	[57]	
Iran (Kerman Province hot springs)	60.78	51.43	394.37	[59]	
Iran (Soleymani Hot Springs)	1630.56	39.99	125.40	[27]	
Turkey (İkizdere Valley)	32.71	134.12	811.68	[8]	
Bangladesh (Jamuna river)	60.0	135.0	1002.0	[55]	
Russia (Potash Salts Deposit)	7.52	9.98	236.67	[58]	
Algeria (Rhumel River)	26.64	25.95	165.5	[17]	
Namibia (Orange River)	63.46	54.88	416.99	[56]	
Nigeria (Tuomo River)	43.89	99.21	793.99	[3]	
Nigeria (Imo River)	187.12	38.62	180.59	[18]	
Nigeria (Osun River)	23.9	17.5	205.6	[14]	
Nigeria (Yobe River)	60.0	45.0	324.0	[60]	
Nigeria (llobi and Erinja Rivers)	52.0	7.6	488.0	[31]	
Ghana (Tema Harbour)	34.0	30.0	320.0	[34]	
Sudan (River Nile)	17.5	16.1	386.0	[37]	
DR. Congo (Rivers in Mining Region)	378.0	30.0	202.0	[24]	
Kenya (Bungoma County Rivers)	2.0	123.0	148.0	[32]	
Kenya (Port Victoria Shoreline)	66.23	76.23	523.21	[7]	
Uganda (Gulu and Amuru Rivers)	89.6	168.6	275.4	Present Study	
UNSCEAR, 2000 (World limits)	35	30	400	[42]	

Table 3: Comparison of average AC of ²³⁸U, ²³²Th and ⁴⁰K in the samples with those in other Countries.

In order to determine the existing ratio between the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in the sediment samples, correlations between them were drawn using *MATLAB R*2022*b*. Figure 3 (a - c) show the correlations between the *AC* of ²³²Th and ²³⁸U, ⁴⁰K and ²³²Th, and ⁴⁰K and ²³⁸U, respectively, with a trend line drawn among the data points. In Figure 3 (a – c), linear and positive correlations were found. It can be seen that there is a good correlation between ²³²Th and ²³⁸U with correlation coefficient of ($R^2 = 0.7879$). The relation shows a slope of 3.689 with intercept 13.69 ppm. Similarly, Figure 3 (b) show a slope of 0.02922 with intercept of 0.1679% while Figure 3 (c) has 0.06951 and 0.5435% as slope and intercept, respectively. However, as is argued later in this paper, the most striking difference is the fact that no consistent correlations were observed between ⁴⁰K and ²³⁸U with weak correlation coefficients of ($R^2 = 0.1841$) and ($R^2 = 0.05483$), respectively.



Figure 3 (a): Comparison of ²³²Th with ²³⁸U.

Figure **3** (b): Comparison of 40 K with 232 Th.



Figure 3 (c): Comparison of ⁴⁰K with ²³⁸U.

In Table 4, the *RHP* due to the individual isotopes varied from $(0.7 \pm 0.06 - 5.0 \pm 0.5) \mu$ W m⁻³ for ²³⁸U, $(1.5 \pm 0.1 - 5.3 \pm 0.6) \mu$ W m⁻³ for ²³²Th, and $(0.006 \pm 0.0004 - 0.4 \pm 0.02000) \mu$ W m⁻³ for ⁴⁰K with averages of 1.7 ± 0.1 , 2.7 ± 0.3 and $0.07 \pm 0.0040 \mu$ W m⁻³, respectively. The total *RHP* values varied from $(2.5 \pm 0.6 - 10.2 \pm 2.6) \mu$ W m⁻³ with an average value of $4.5 \pm 1.1 \mu$ W m⁻³. The percentage contributions varied from $(22.6 \pm 1.1 - 49.0 \pm 2.5)$ % for ²³⁸U, $(50.0 \pm 2.5 - 77.4 \pm 3.9)$ % for ²³²Th, and $(0.1 \pm 0.005 - 4.7 \pm 0.24)$ % for ⁴⁰K with averages of 37.8 ± 1.9 %, 60.0 ± 3.0 %, and 2.2 ± 0.11 %, respectively. The H_f in the samples due to the decay of ²³⁸U, ²³²Th and ⁴⁰K varied from $(19.6 \pm 1.0 - 80.0 \pm 4.0)$ mW m⁻² with average of 35.3 ± 1.8 mW m⁻². Figure 4 illustrates a plot of total *RHP* and H_f producing a linear distribution segments with strong correlation coefficient of $R^2 = 0.9997$. The linear regression through the data points gives a slope of 7.836 km and vertical axis intercept of 0.0768 mW m⁻².

ID	RHP (U)	RHP (Th)	RHP (K)	²³⁸ U (%)	²³² Th (%)	⁴⁰ K (%)	$H_f(mWm^{-2})$
S_1	1.3 (0.1)	2.1 (0.3)	0.08 (0.0050)	37.1 (1.9)	60.0 (3.0)	2.9 (0.14)	27.5 (1.4)
S_2	1.4 (0.1)	2.5 (0.3)	0.03 (0.0020)	35.9 (1.8)	64.1 (3.2)	0.8 (0.04)	31.9 (1.6)
S_3	2.6 (0.2)	4.0 (0.5)	0.05 (0.0030)	38.8 (1.9)	59.7 (3.0)	1.5 (0.08)	52.5 (2.6)
S_4	1.5 (0.1)	2.7 (0.3)	0.04 (0.0020)	35.7 (1.8)	64.3 (3.2)	1.0 (0.05)	32.9 (1.6)
S_5	1.2 (0.09)	2.1 (0.3)	0.05 (0.0030)	35.3 (1.8)	61.8 (3.1)	2.9 (0.14)	26.7 (1.3)
S_6	2.9 (0.3)	5.3 (0.6)	0.4 (0.02000)	33.7 (1.7)	61.6 (3.1)	4.7 (0.24)	67.5 (3.4)
S_7	1.2 (0.09)	2.2 (0.2)	0.05 (0.0030)	34.3 (1.7)	62.9 (3.1)	2.8 (0.14)	27.5 (1.4)
S_8	1.7 (0.1)	2.4 (0.3)	0.07 (0.0040)	40.5 (2.0)	57.1 (2.9)	2.4 (0.12)	32.9 (1.6)
S_9	1.5 (0.1)	2.1 (0.2)	0.08 (0.0050)	40.5 (2.0)	56.8 (2.8)	2.7 (0.14)	29.0 (1.5)
S_{10}	1.5 (0.1)	2.0 (0.2)	0.2 (0.01000)	42.9 (2.1)	57.1 (2.9)	0.6 (0.03)	27.5 (1.4)
S_{11}	0.7 (0.06)	2.4 (0.3)	0.01 (0.0004)	22.6 (1.1)	77.4 (3.9)	0.2 (0.01)	24.3 (1.2)
S_{12}	3.0 (0.3)	4.6 (0.6)	0.01 (0.0060)	39.5 (2.0)	60.5 (3.0)	0.1 (0.005)	59.6 (3.0)
S_{13}	2.0 (0.2)	2.9 (0.3)	0.07 (0.0040)	40.0 (2.0)	58.0 (2.9)	1.4 (0.07)	39.2 (2.0)
S_{14}	5.0 (0.5)	5.1 (0.6)	0.06 (0.0040)	49.0 (2.5)	50.0 (2.5)	0.6 (0.03)	80.0 (4.0)
S_{15}	1.2 (0.09)	2.0 (0.2)	0.03 (0.0020)	37.5 (1.9)	62.5 (3.1)	0.9 (0.05)	25.1 (1.3)
S_{16}	1.5 (0.1)	1.8 (0.1)	0.05 (0.0030)	44.1 (2.2)	52.9 (2.6)	3.0 (0.15)	26.7 (1.3)
S_{17}	1.6 (0.1)	2.6 (0.3)	0.08 (0.0050)	37.2 (1.9)	60.5 (3.0)	2.3 (0.12)	33.7 (1.7)
S_{18}	0.8 (0.06)	1.6 (0.1)	0.05 (0.0030)	32.0 (1.6)	64.0 (3.2)	4.0 (0.20)	19.6 (1.0)
S_{19}	1.7 (0.1)	2.6 (0.3)	0.06 (0.0040)	38.6 (1.9)	59.1 (3.0)	2.3 (0.12)	34.5 (1.7)
S_{20}	1.9 (0.2)	3.1 (0.4)	0.03 (0.0020)	38.0 (1.9)	62.0 (3.1)	0.6 (0.03)	39.2 (2.0)
S_{21}	2.2 (0.2)	3.1 (0.4)	0.05 (0.0030)	40.7 (2.0)	57.4 (2.9)	1.9 (0.10)	42.4 (2.1)
S_{22}	1.4 (0.1)	2.1 (0.2)	0.06 (0.0040)	38.9 (1.9)	58.3 (2.9)	2.8 (0.14)	28.2 (1.4)
S_{23}	1.3 (0.1)	2.1 (0.2)	0.1 (0.00600)	37.1 (1.9)	60.0 (3.0)	2.9 (0.15)	27.5 (1.4)
S_{24}	1.7 (0.1)	2.6 (0.3)	0.22 (0.0200)	37.8 (1.9)	57.8 (2.9)	4.4 (0.22)	35.3 (1.8)
S_{25}	1.2 (0.09)	1.8 (0.1)	0.05 (0.0030)	38.7 (1.9)	58.1 (2.9)	3.2 (0.16)	24.3 (1.2)
S_{26}	0.9 (0.07)	1.5 (0.1)	0.05 (0.0030)	36.0 (1.8)	60.0 (3.0)	4.0 (0.20)	19.6 (1.0)
S_{27}	1.1 (0.08)	1.7 (0.1)	0.05 (0.0030)	37.9 (1.9)	58.6 (2.9)	3.5 (0.18)	22.7 (1.1)
S_{28}	1.8 (0.2)	2.6 (0.3)	0.01 (0.0080)	40.9 (2.0)	59.1 (3.0)	0.3 (0.02)	34.5 (1.7)
S_{29}	2.3 (0.2)	3.6 (0.4)	0.06 (0.0040)	38.3 (1.9)	60.0 (3.0)	1.7 (0.09)	47.1 (2.4)
S_{30}	1.7 (0.1)	3.0 (0.4)	0.05 (0.0030)	35.4 (1.8)	62.5 (3.1)	2.1 (0.11)	37.6 (1.9)
Aver.	1.7 (0.1)	2.7 (0.3)	0.07 (0.0040)	37.8 (1.9)	60.0 (3.0)	2.2 (0.11)	35.3 (1.8)
Min.	0.7 (0.06)	1.5 (0.1)	0.01 (0.0004)	22.6 (1.1)	50.0 (2.5)	0.1 (0.005)	19.6 (1.0)
Max.	5.0 (0.5)	5.3 (0.6)	0.4 (0.02000)	49.0 (2.5)	77.4 (3.9)	4.7 (0.24)	80.0 (4.0)

Table 4: *RHP* due to individual radioelements, total *RHP*, percentage contribution, and $H_f(mWm^{-2})$.



Figure 4: Comparison of *RHP* with *H*_f.

As can be inferred from Table 5 and Figure 5 (a), the analyzed samples were subdivided into three categories according to their *RHP*: low *RHP* samples (*RHP* < 2 μ W m⁻³), moderate *RHP* samples (2 < *RHP* < 4 μ W m⁻³) and high *RHP* samples (*RHP* > 4 μ W m⁻³) as proposed by Ethinola *and colleagues* [43] and later used by Laith

and colleagues [9] and Adagunodo *and colleagues* [44]. These assemblages show the significance of the *RHP* to the thermal regime of the region. 50% of the samples showed moderate *RHP*, another 50% showed high *RHP* and 0% showed low *RHP*.

Table 5: Classification	n of the studied	l sample in term	s of RHP.
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Low RHP Samples	Moderate RHP Samples	High RHP Samples
Zero samples.	$S_1, S_2, S_5, S_7, S_9, S_{10}, S_{11}, S_{15}, S_{16}, S_{18}, S_{22}, S_{23},$	S_3 , S_4 , S_6 , S_8 , S_{12} , S_{13} , S_{14} , S_{17} , S_{19} , S_{20} , S_{21} ,
	S_{25} , S_{26} , and S_{27} (15 Samples).	S_{24} , S_{28} , S_{29} , and S_{30} (15 Samples).
0%	50%	50%

Figure 5 (b) illustrates the percentage contributions of 238 U, 232 Th and 40 K to the total *RHP*. It is very evident in Figure 5 (b) that 232 Th was the major contributor to total *RHP* with maximum value of 77.4%, followed by 238 U with maximum percentage of 49%. The 40 K contributed the least maximum percentage of 4.7%.



Figure 5 (a): Classification of the total *RHP*.

Figure 5 (b): Contributions of each isotope to total RHP.

4. Discussions

4.1 Activity Concentrations of ²³⁸U, ²³²Th and ⁴⁰K

The primeval radioactive elements ²³⁸U, ²³²Th and ⁴⁰K were present in all the sediment samples at varying amount. These variations may be due to the drainage pattern of the study area. It is likely that the physical and chemical (*pH*, organic content and redox) properties could have changed along the basins of the studied rivers. Similar results were also reported by the authors in [17, 24, 45] using gamma ray spectrometer techniques, who attributed them to huge variations in chemical and mineralogical properties of the provenance area. This study also found that the *AC* of ²³⁸U were higher than that of ⁴⁰K in samples *S*₁₄ and *S*₂₈. This may be due to the solubility and mobility of uranyl, $U(VI)O_2^{2^+}$ in water, as has also been reported by the authors in [13, 31, 46]. Under oxidizing conditions, uranium is found in soluble U(VI) oxidation state and remains dissolved in water [12, 26, 47]. In river water, U(VI) can be reduced to insoluble U(IV) under anoxic condition and precipitate to sediments and therefore increase *AC* of ²³⁸U in the sediment. This confirms the findings by the authors in [18, 24, 27, 48]. In the study by Atibu *and colleagues* [24], high *AC* of ²³⁸U was attributed to artisanal mining activities while Reza *and colleagues* [27] attributed it to the existence of hot springs. Sample *S*₁₁ had the lowest *AC* of ²³⁸U of 36.1 Bq kg⁻¹, which is 1.031 times higher than the world average value of 35 Bq kg⁻¹ for river sediments set by *UNSCEAR* [42]. Uranium may have become immobile under the reducing environment, due to

the insoluble nature of uraninite (UO₂) and coffinite (USiO₄). The sample S_{11} also showed high AC of ²³²Th (151.1 Bq kg⁻¹) and low AC of 40 K (222.2 Bq kg⁻¹). Due to the high ionic potential of Th⁴⁺, 232 Th remains in solution for a short while before being precipitated as hydroxylates or being adsorbed on clay surfaces. It is important to bear in mind, though, that, while this is true for ²³²Th, it is not necessarily the case for ²³⁸U and ⁴⁰K. This unique feature of ²³²Th might have led to its significantly high AC in S_{11} compared to ²³⁸U and ⁴⁰K. Recent studies by Sindani and colleagues [32], and Satyanarayan and colleagues [49] also showed AC of ²³²Th greater than ⁴⁰K and they attributed the trend to the geological formation and type of rocks across the selected rivers. Although S_{11} showed high AC of ²³²Th, it should not be forgotten that the AC of ²³²Th was also higher than ²³⁸U and 40 K in 33.3% of the samples (S_2 , S_3 , S_4 , S_{14} , S_{15} , S_{20} , S_{21} , S_{28} , S_{29} , and S_{30}). The low AC of 40 K in the said samples was somewhat unexpected as previous studies showed that ⁴⁰K is the most common radionuclides with abundance of 0.012% [33]. Nevertheless, ²³²Th has very low solubility in almost all aqueous environment. Due to the great insolubility of ThO_2 & $ThSiO_4$, ²³²Th does not usually spread quickly through the environment when released. To a large degree, the Th^{4+} ion is insoluble, especially in acidic river sediments, and in such conditions, ²³²Th concentration can be higher. This finding is consistent with those of Suresh Gandhi and colleagues [50, 51] who found that AC of ²³²Th was 20.31 times higher than ²³⁸U and 2.04 times higher than ⁴⁰K. They attributed these anomalies to the presence of the loamy sediments at Thiruvanmiyur which had relatively high amount of ²³²Th. In contrast, sample S_{26} presented the lowest AC of ²³²Th of 97.4 Bq kg⁻¹, which is 3.25 times higher than the world average of 30 Bq kg⁻¹ for river sediments set by UNSCEAR [42]. This means that S_{26} had low amount of thorium bearing minerals. It is evident that all the analyzed samples had AC of ²³⁸U and ²³²Th higher than the global limits. This means that, the samples were rich in uranium and thorium bearing minerals. This undoubtedly confirms the earlier findings by Westerhof and colleagues [22] and Olanya and colleagues [33] for North Uganda terrane. This study also found that in 70% of the samples, the AC were in the order ${}^{40}\text{K} > {}^{232}\text{Th} > {}^{238}\text{U}$, similar to the results obtained by the authors in [5, 7, 32, 52, 53]. This may be attributed to the fact that ⁴⁰K is the most abundant in the Earth's crust as an essential component of many light minerals. Ugbede & Akpolile [3], Abu, Abbady & Harb [6] and Elena, and colleagues [54] also argued in favor of 40 K being a naturally occurring radionuclide which abounds in the earth crust, with an abundance of 0.012% (120 ppm). Also, the AC in 70% of the samples show that the underlying bedrock composed of surficial materials. It was also discovered that samples S_6 (1442.3 Bq kg⁻¹), S_{10} (575.0 Bq kg⁻¹), and S_{24} (829.1 Bq kg⁻¹) had AC of ⁴⁰K higher than world average value of 400 Bq kg⁻¹ by factors of 3.61, 1.44 and 2.07, respectively. This means that, S_6 , S_{10} , and S_{14} had high potassium rich minerals such as feldspar, micas and clay minerals. The clayey samples were taken from rivers with weathered granitic rocks outcrops from Pogo, Coke and Kal sites, respectively. After weathering, ⁴⁰K could have been transported with the rock fragments by fluvial means from raised land to the sample points (S_6 , S_{10} , and S_{14}) as a dissolved phase. On the contrary, 90% of the samples had AC of ⁴⁰K lower than world average value of 400 Bq kg⁻¹ [42]. The primary argument in favor of the low AC of 40 K is that the 90% samples could have low content of feldspar, mica and clay minerals. A study by Sindani *and colleagues* [32] supports this observation and suggests that low AC of 40 K can be attributed to less amount of feldspar, mica and clay minerals. However, not all the analyzed samples followed this trend. In sample S_6 of Pogo site, for example, AC were higher than the global limits of 35, 30, and 400 Bq kg⁻¹ by factors of 4.3, 11.1, and 3.6, for 238 U, 232 Th and 40 K, respectively. Similar high AC were also detected in samples S_{14} of Paminrut site, and S_{20} of Paidwe site, respectively. The samples S_6 , S_{14} and S_{20} consisted of mainly fine granitic rock materials

and clay sediments. The high AC of the radionuclides may be due to the abundance of clay content in the studied sediments as reported by the authors in [17, 25, 47, 55, 56]. This was noted during sampling. The clay sediments, known to be a high emanator of radon, changed the water color to pale which could have increased sediment radioactivity levels. This observation is highly significant but caution is warranted as we could not scientifically link the change in water color to the detected high AC in the sediments. The above observation requires further investigations.

To gaze the natural radioactivity levels in the study area, the *AC* of ²³⁸U, ²³²Th and ⁴⁰K were compared with those in river sediments of other countries as presented in Table 3. The *AC* values in Table 3 are not the representative values for the respective countries but for the studied rivers. The values obtained in this study compare well with those reported by other authors; Ugbede [3], Masinde [7], Reza *and colleagues* [27] and Khalil *and colleagues* [55] and above the *UNSCEAR* [42] global average. Also, the *AC* values reported by the authors in [9, 14, 16, 17, 28, 32, 35, 37, 57, 58] were lower than the results presented in this paper and the global average. However, the current study found lower average *AC* of ²³⁸U compared to those obtained by Ononugbo, Avwiri & Ogan [18], Atibu *and colleagues* [24], and Reza *and colleagues* [27]. Also, the average *AC* of ²³²Th was lower than that of Suresh Gandhi *and colleagues* [51]. Furthermore, a significant discrepancy exists between the average *AC* of ²³⁸U and ²³²Th and the lower values obtained by the authors in [1, 9, 11, 16, 18, 24, 25, 30, 31, 34, 35, 37, 56, 57, 58, 59]. This discrepancy is due to the varied mineralogical and geochemical composition of parent rocks. The average of ⁴⁰K was lower than those reported by the authors in [3, 7, 8, 24, 55] but higher than those reported by authors in [14, 16, 17, 18, 27, 32, 35].

4.2 Radioelement Concentrations of ²³⁸U, ²³²Th and ⁴⁰K

The calculated average RC were 7.1 \pm 0.5 ppm, 41.3 \pm 4.3 ppm, and 1.1 \pm 0.05% for ²³⁸U, ²³²Th and ⁴⁰K, respectively. These were above the worldwide average concentration of 2.88 ppm for ²³⁸U, and 11.25 ppm for ²³²Th but smaller than 1.31% for ⁴⁰K in sediments as reported by UNSCEAR [42] and later on cited by Laith and colleagues [9]. This paper reported values higher than those of Laith and colleagues [9] and Adagunodo and colleagues [44]. This shows that the studied samples had high content of ²³⁸U and ²³²Th and relatively low content of ⁴⁰K. This reveals the geological, geochemical and geographical conditions of the area. The values of ²³⁸U and ⁴⁰K were lower than those found by Sabina and colleagues [60]. The value of ²³⁸U found by Sokari and colleagues [36] is 1.5 times higher than the results presented in this paper. To show the relative depletion or enrichment of ²³⁸U and ²³²Th in the studied samples, the elemental concentration ratios ²³²Th/²³⁸U was determined. According to UNSCEAR [42], the theoretical values of the elemental ratios ²³²Th/²³⁸U are expected to be 3.0 for normal continental crust. The values obtained for the ratio ²³²Th/²³⁸U in this study varied from 3.77 - 12.80 with average of 6.09. This means there was significant fractionation of both the elements during weathering in which both uranium and thorium were enriched. These results were higher than that of the continental crust by factor of 2 and less consistent with 0.96 for Saif Eldin and colleagues [37], 1.9 for Faweya [61], 0.67 for Rafat [2], and 0.90 for Sathish and colleagues [12], respectively. The average ratio ²³²Th/²³⁸U in this study is lower than 18.99 for SureshGandhi, and colleagues [51], but above the world average of 3.94 reported by Sathish and colleagues [12]. Hence, the study area, without exception have relatively high background levels of ²³⁸U and ²³²Th from its soil (low organic content) and rock outcrops. This finding needs

further studies.

To determine the geochemical behavior of ²³⁸U, ²³²Th and ⁴⁰K in the studied sediments, correlation plots were done as shown by Figure 3 (a-c). The remarkable correlations between ²³²Th and ²³⁸U demonstrates that they originated from the same source, that is, the sediment samples were geochemically coherent. ²³⁸U & ²³²Th belong to actinide group of elements with similar radii ($U^{4+} = 1.05$ Å, $Th^{4+} = 1.1$ Å) and similar electron configuration ($z_u = 92$, $z_{Th} = 90$). This is consistent with the previous data reported by Khalil and colleagues [55], Ramadan and colleagues [62], and Tholkappian and colleagues [63], who attributed the good correlations to the likely presence of monazite minerals in the samples and that it may also demonstrate the property of the sediment samples in retaining ²³⁸U, ²³²Th and ⁴⁰K under varying weather conditions. Contradicting phenomenon was observed in the studies by Murugesan and colleagues [13], Al-Mur and colleagues [45], and Sylvanus and colleagues [56], who found weak correlations between 232 Th and 238 U of $R^2 = 0.426$, $R^2 = 0.419$, and $R^2 =$ 0.1528, respectively. Sylvanus and colleagues [56] attributed this to the weak natural abundance of ²³⁸U and ²³²Th over period of times through their decay process and Murugesan and colleagues [13] attributed the weak correlations to minimum contribution of monazite mineral in the sample. The weak relationship between ²³⁸U and ²³²Th ($R^2 = 0.451$) is in general agreement with the conclusions of Uosif and colleagues [30] in Marine sediments of Quseir City, Egypt using NaI(TI) detector. They attributed this to ²³⁸U and ²³²Th having some similarity in their environmental origin. The weak correlations between ⁴⁰K and ²³²Th of ($R^2 = 0.1841$) illustrated by Figure 3 (b), shows that ⁴⁰K had source that was independent of ²³²Th. During the weathering process, ⁴⁰K and ²³²Th could have reacted differently. As discussed by [56], ⁴⁰K is more soluble and is easily carried away in water, whereas ²³²Th tends to remain. The weak correlations between ⁴⁰K and ²³⁸U (R^2 = 0.05483) indicated by Figure 3 (c) is probably due to the fact that ²³⁸U is more soluble and mobile compared to 40 K. Such insignificant correlation between 238 U and 40 K ($R^2 = 0.2337$) and 232 Th and 40 K ($R^2 = 0.0604$) are conspicuous in the findings by Uosif and colleagues [30] and Sylvanus and colleagues [56], who attributed this to high potassium solubility and the anomaly in potassium existence on earth crust due to human activity. The current finding is less consistent with ($R^2 = 0.853$) for 40 K versus 238 U and ($R^2 = 0.826$) for 40 K versus 232 Th reported by Rafat [2] in sediment samples from Qarun Lake, Egypt analyzed by HPGe detector. They attributed the significant positive correlations between ²³⁸U, ²³²Th and ⁴⁰K to the agriculture and industrial wastes in the fine-grained sediments that were rich in organic matter content.

4.3 Radioactive Heat Production and Associated Heat Flow

To determine the geothermal potential of the study area, *RHP* of the samples were calculated. The total *RHP* values varied from $2.5 \pm 0.6 - 10.2 \pm 2.6 \,\mu\text{W} \,\text{m}^{-3}$ with an average value of $4.5 \pm 1.1 \,\mu\text{W} \,\text{m}^{-3}$. The average value is higher than those found by authors in [9, 11, 13, 36, 39, 40, 64, 65], but similar to those of Asere *and colleagues* [15] and Adagunodo *and colleagues* [44]. The samples were subdivided into three categories according to Ethinola *and colleagues* [43] as cited in Laith *and colleagues* [9] and Adagunodo *and colleagues* [44]. The first category shows 0% samples with low *RHP*. The second shows 50% samples with moderate *RHP*. The third shows the remaining 50% samples with high *RHP*. The most striking feature of this categorization scheme reveals that there is great heat production in the study area due to the decay of ²³⁸U, ²³²Th and ⁴⁰K. A common assumption is that high *RHP* in an area is associated with viable geothermal resources [9]. The

percentage contribution to the heat produced by ²³⁸U, ²³²Th and ⁴⁰K in the samples are summarized in Table 4 with averages of 37.8%, 60.0% and 2.2%, respectively. The ²³²Th contributed the highest percentage to the total heat produced in the samples. The contribution of ²³²Th to the total *RHP* was prominent in sample S_{II} of Pailyec site with the highest population of 15,042 inhabitants. As indicated in Table 4, the combined contributions of ²³²Th and ²³⁸U is 97.8% to the total *RHP*. No significant contributions to the total *RHP* came from ⁴⁰K, only 2.2%. The ²³⁸U and ²³²Th contributions may have come from the same source (probably from the basement rock) while that of ⁴⁰K may be due to mica, feldspar and clay minerals. However, the total *RHP* depicts the overall contributions from ²³⁸U, ²³²Th and ⁴⁰K for each sample and is a significant parameter reflecting the thermal regime of the region. This conclusion is in general agreement with the conclusions of Murugesan & Ravichandran [11], who studied sand samples from Cauvery and Palar rivers, India using NaI(Tl) detector. However, Sokari, Gbarato & Ononugbo [36] analyzed sediment samples from Okrika Local Government Area of Rivers state, Nigeria using NaI(Tl) detector and concluded that the *RHP* was mostly from ²³⁸U in the order of ²³⁸U > ²³²Th > ⁴⁰K. In contrast, the study by Omosule & Adelowo [38] in Osun State of Nigeria using NaI(Tl) detector discovered that ⁴⁰K was the major contributor to heat production compared to ²³⁸U and ²³²Th.

To determine H_f in the sampled points due to the decay of ²³⁸U, ²³²Th and ⁴⁰K, H_f was modeled from the values of *RHP*. The H_f in the samples varied from (19.6 ± 1.0 - 80.0 ± 4.0) mW m⁻² with average of 35.3 ± 1.8 mW m⁻². This average H_f is slightly higher than 21.6 mW m⁻² reported by Asere *and colleagues* [15] for quarry sites in Ondo State, Nigeria but less than the global value of 37 mW m⁻² stated by Turcotte & Schubert [41] for inactive regions. Katumwehe *and colleagues* [66] reported H_f for North Uganda Terrane of between 58 - 65 mW m⁻², while [67] reported 34 - 70 mW m⁻² for the Tanzanian craton. In comparison, the maximum H_f value (80 mW m⁻²) obtained in this study maybe be an outlier, not true value for the North Uganda terrane and is beyond the scope of this work. This requires further investigation using another method. Figure 4 shows the plot of the total *RHP* against H_f with significantly strong correlation coefficient of $R^2 = 0.9997$. As expected, *RHP* in the samples produced a significantly greater effect on H_f . The H_f values reflect the high *RHP* due to decay of ²³⁸U, ²³²Th and ⁴⁰K in the study area, indicating feasibility for geothermal exploration. This agrees with the findings of Turcotte & Schubert [41] for stable continental areas, where the H_f had a strong correlation with the surface *RHP*. Hence, this research shows that the study area is in thermally more quiescent regions.

5. Conclusions

The average *AC* of ²³⁸U, ²³²Th and ⁴⁰K, and the computed *AC* of ²³⁸U and ²³²Th in all the samples were higher than the global limits. Hence, the study area is rich in uranium and thorium bearing minerals. The *AC* in 33.3% of the samples were in the order of ²³²Th > ²³⁸U> ⁴⁰K, while in 70% of the samples were in the order ⁴⁰K > ²³²Th > ²³⁸U. The *AC* of ⁴⁰K in 90% of the samples were lower than the global limits.

The samples showed 50% moderate and 50% high *RHP* with ²³²Th contributing the highest percentage to total *RHP*. However, an increase in the concentrations of ²³⁸U, ²³²Th and ⁴⁰K reflected the integrated effect of *RHP*. Also, as *RHP* increases, there is observed concomitant increased in H_{f} . Hence the study area is suggested to have good potential for geothermal exploration and are underlain by stable continental crust.

The results of this study can be used as a baseline for further studies with special focus on the region's medium enthalpy resources such as Amoropii, Amuru and Panyimur. Particular attention should be given to parameters such as temperature, geochemistry, crustal heat flux, the origin and flow of the fluids.

6. Limitations and Recommendations

This research encountered some limitations. First, due to paucity of data on *RHP*, H_f and geothermal resources from the studied area, comparison and corroboration of previous data from the study area was difficult, though significant. To enrich the discussion and arrive at a better conclusion, the findings of this study was juxtaposed with those from other parts of the world. Therefore, this study serves as a baseline data and the basis for further research. Secondly, trace concentrations of ²³⁸U, ²³²Th and ⁴⁰K in the room furniture, floor, walls and possibly dust could have a significant influence on counting efficiency. This limitation was taken care of by; (i) installing the NaI(Tl) detector in the centre of the counting room, rather than around the walls to reduce background radiation from the walls, (ii) sealing the base of the detector with 15 cm wood that reduced the radiations from the floor by 20 - 25%. (iii) sealing the laboratory windows, ventilators and doors for the duration of the experiment to avoid ingress of dust. Further studies are necessary to validate the accuracy of our methods and assumptions.

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7. Conflict of Interest

The authors declare no conflicts of interest.

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