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Measurement of Surface Dose Rate of Nuclear Radiation in Coastal Areas of Akwa Ibom State, Nigeria

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Abstract In this study, the surface doses rate measurements were done in-situ using dose rate meters. The analysis of naturally occurring radionuclides (^{40}K, ^{238}U and ^{232}Th) has been carried out in soil samples collected from Eastern Obolo, Ikot Abasi, and Ibeno local government areas of Akwa Ibom State, using gamma spectroscopy operated on a Canberra vertical high purity 2''×2'' NaI(Tl) model 802 detector. The activity concentration (Bq.kg^{-1}) of the samples ranges from 34.65±2.56 to 214.12±4.34 with mean value 94.60±3.42 for ^{40}K, 5.12±0.38 to 38.5±2.38 with mean value 15.16±1.83 for ^{238}U and 0.03±0.77-30.59 with mean value 15.40±0.73 for ^{232}Th. This study also examines some radiation hazard indices, the mean values obtained are 44.46 Bq.kg^{-1}, 20.52 nGy.h^{-1}, 0.02 mSv, 0.17 Bq.kg^{-1}, 0.20 Bq.kg^{-1} for Radium Equivalent activity (Ra_{eq}), Absorbed Dose rate (D), Annual Effective Dose Rate (E_{eff} Dose), External Hazard index (H_{ex}) and internal Hazard index (H_{in}) respectively. These calculated hazard indices to assess the potential radiological health risk in soil and the dose associated with it are well below their permissible limit. The soil and sediments from the study area provide no excessive exposures for inhabitants and can be use as construction material without posing any immediate radiological threat to the public. However, the public is cautioned against excess exposure to avoid future accumulative dose of these radiations.

Keywords: *natural radioactivity, radiological hazards, NaI (Tl) Detector, gamma-ray spectrometry*

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1. Introduction

The stellar material, from which the earth was formed, about 4.5 billion years ago, contained many unstable nuclides [1]. Some of the original primordial nuclides, whose half-lives are about as long as the earth's age, are still present [2]. The exposure of human beings to ionizing radiation from natural sources is a continuing and inescapable feature of life on earth; for most individuals, this exposure exceeds that from all man-made sources combined [3]. Over 60 radionuclides (radioactive element) can be found in nature, and they can be placed in three general categories i.e. Primordial – formed before the creation of the earth, Cosmogenic- formed as a result of cosmic ray interactions and Human produced- enhanced or formed due to human actions (minor amounts compared to natural) [4]. The natural terrestrial gamma radiation dose rate is an important contribution to the average dose rate received by the world's population [5,6]. Estimation of the radiation dose distribution is important in assessing the health risk to a population and serve as reference in documenting changes to environmental radioactivity in soil due to anthropogenic activities [7]. Human beings are exposed outdoors to the natural terrestrial radiation that originates predominantly from the upper 30cm of the soil [8]. Radionuclides with half-lives comparable with the age of the earth or their corresponding decay products existing in terrestrial material such as ^{232}Th, ^{238}U and ^{40}K are of great interest. More specifically, natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world [10,11]. Naturally occurring radioactive material (NORM) found in the earth's crust, largely in the form of ^{226}Ra and their associated radionuclides, is brought to the surface during gas and oil production processes. The NORM represents a potential internal radiation exposure hazard to both workers and members of the public through the inhalation and ingestion of radionuclides [11]. The major potential hazard from the natural radiation is from external exposure either by direct exposure to the soil or as they enter in many building material [12]. The specific levels of terrestrial environmental radiation are related to the geographical composition of each lithologically separated area, and to the content of the rock from which the soils originated in each area in the radioactive elements of Thorium (Th), Uranium (U) and Potassium (K). It is well known, for instance, that, igneous rocks granite composition are strongly enriched in Th and U as compared to rocks of basaltic or ultramafic composition [9,12,13]. There are exceptions, however, as some shale and phosphate rocks have relatively high content of those radionuclide [9,10]. Based on these facts, one can certify that the knowledge of natural occurring radionuclide materials (NORMs), such as ^{238}U, ^{232}Th and ^{40}K, is an

important pre-requisite for evaluation of the rate of exposure absorbed dose by the population in order to estimate their radiological impacts and to establish a data base which will be used as reference to radiation observer in the studied area [15].

1.1. Description of Study Area

The soil samples for this research were collected within Akwa Ibom State, an oil producing state in the Niger Delta region of Nigeria. The Niger Delta is situated in the Gulf of Guinea between latitude 3° and 6°N and longitude 5° and 8°E. It is the largest delta in Africa, and is very rich in hydrocarbons, covering an area of about 7500km^2 [16]. The first set of samples were collected from Eastern Obolo (04°30'9"−04°31'3") N, (007°45'3"−007°45'3") E within an abandoned oil operational area and around estuary where dredging activities were carried out. The second set of samples were collected from Ikot Abasi (04°32'6"−04°33'3") N, (007°32'6"−007°34'0.02") E around Uta Ewa beach and Aluminum Smelting Plant. The third set of samples were collected from Ukpenekang in Ibeno (04°32'23"−04°34'9") N, (008°009'8"−007°59'8") E where some oil spill traces were observed.

2. Materials and Methods

2.1. Sample Collection and Treatment

Table 1. In- situ Measurement

SAMPLE CODE	LOCATION		SURFACE DOSE RATE	
	NORTH	EAST	RADALERT^{100} (mR/hr)	DIGILERT^{TM} (cpm)
EO1	04^{0}30'40.9''	007^{0}45'11.3''	0.023	5.25
EO2	04^{0}30'41.9''	007^{0}45'11.4''	0.012	10.67
EO3	04^{0}30'43.9''	007^{0}45'11.7''	0.011	14.00
EO4	04^{0}30'44.8''	007^{0}45'07.8''	0.012	13.00
EO5	04^{0}30'48.7''	007^{0}45'02.6''	0.011	9.67
EO6	04^{0}31'19.3''	007^{0}45'17.3''	0.014	20.50
IK1	04^{0}32'32.6''	007^{0}32'51.6''	0.011	8.33
IK2	04^{0}32'55.7''	007^{0}32'55.3''	0.018	8.00
IK3	04^{0}32.843'	007^{0}32.825'	0.013	9.67
IK4	04^{0}33.692'	007^{0}32.568'	0.012	9.33
IK5	04^{0}33'58.3''	007^{0}34'02.2''	0.010	11.67
IB1	04^{0}32'23''	008^{0}009'8.8''	0.017	14.67
IB2	04^{0}34'12.2''	007^{0}59'16.2''	0.011	9.67
IB3	04^{0}34'14.0''	007^{0}59'16.6''	0.011	10.00
IB4	04^{0}34.306'	007^{0}59.218'	0.016	16.67
IB5	04^{0}34'21.9''	007^{0}59'12.8''	0.012	11.67

The samples were collected and prepared according to the method reported by Agbalagba and Onoja [17]. The top surfaces of the soils at all the soil sampling sites were scraped off to remove stones, vegetation and organic debris [18]. Thereafter, about 5kg weight of field samples of the soil at each of the sites were collected at a depth of 5-15 cm, thoroughly mixed and loaded in a labeled black polyethylene bags. Each of the twenty soil samples collected was a composite of 5 subsamples. At all the sample locations, Global Positioning System (GPS) was used in locating the coordinates of each sample station. Surface dose measurements were performed at each sampling point using two survey meters, $Radalert^{100}_{TM}$ and Digilert rate meters. The soil samples collected from the field were quartered and exposed to ambient air. The soil samples were then oven dried to a constant weight at 60 − 80°C for about 24 hours in a monitored KETONG 101 oven. The dried samples were ground with mortar and pestle and then passed through a 2 mm mesh sieve and weighed. 500 grams of each soil samples was weighed and wrapped in labeled black polyethylene bag for easy transportation to the laboratory for analysis. The sample locations and descriptions are given in Table 1.

2.2. Sample Analysis

The gamma spectrometric measurement was carried out using Gamma ray spectrometric system coupled with a NaI (Tl) model 802 detector at the National Institute of Radiation Protection and Research, University of Ibadan Campus, Ibadan. The detector is mounted vertically coupled with 8K PC based Multi- Channel Analyzer (MCA) and the detector is enclosed in a massive lead shield to reduce background of the system. The detector was calibrated with point sources Co-60, Cs-137, Am-241 and Na-22 for energy calibration and the efficiency calibration of the detector was done with volume source, IAEA-385. The detector which was well calibrated used Menager [14] as its operating software in the analyses of various energies of K-40, U-238 and Th-232. Each sample was sealed in an already washed Marinelli beaker for twenty eight days in order for it to attain secular equilibrium (to allow buildup of radionuclide in the beaker) before placing in the shielded detector. The counting time for the samples was 36,000 seconds. Each sample was counted for 36,000 seconds to reduce the statistical uncertainty. An already washed empty Marinelli beaker was also placed in the detector for the same counting time (36,000 seconds) under identical geometry to determine the background radiation level of the laboratory environment. It was later subtracted from the measured γ−ray spectra of each sample. At the end of the measurement, the various region of interest which was deducted from the background reading was computed with a specialized template involving the energy, percentage error, count, uncertainty, Activity concentration, uncertainty in activity, Gamma probability, uncertainty in Gamma probability, Efficiency and uncertainty in Efficiency was used to determine the radionuclide concentration in each sample.

According to published reports, the activity concentration, A, in unit of Bq.kg^{-1}, for a radionuclide with a detected photopeak at energy E, is obtained from the following equation given by Awudu *et al*. [19] and Faanu *et al*. [20]:

$$A = \frac{N}{\xi \times t \times \gamma \times M} \quad (1)$$

where N is the net peak-area of the radionuclide, ξ is the detector energy-dependent efficiency, t is the counting live time in seconds, γ is the gamma-ray yield per disintegration of the nuclide, and M is the mass of the sample measured in kilograms.

3. Analysis of Results and Discussion

3.1. Radiological Hazard Indices

The activity concentration of ^{238}U, ^{232}Th and ^{40}K measured in each of the studied samples indicate the quantity of radioactivity present but do not provide a measure of radiation hazard. The hazard associated with radioactivity in samples is assessed through hazard indices. A hazard index is a parameter that is represented by a single value that takes into account the measured activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the sample. The various types of radiological hazard indices are the radium equivalent activity, external and internal hazard indices, radioactivity level index, dose rate, annual effective dose and total absorbed dose rate.

Radium equivalent: To represent the activity levels of ^{238}U, ^{232}Th and ^{40}K which take into account the radiological hazards associated with them, a common radiological index has been introduced. This index is called radium equivalent activity (Ra_{eq}) and is mathematically defined by [10].

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \quad (2)$$

Where A_U, A_{Th} and A_K represent the activity concentrations of uranium, Thorium and Potassium respectively. The permissible limit of Ra_{eq} is 370Bq.kg^{-1} in soil that contain ^{238}U, ^{232}Th and ^{40}K measured in Bq.kg^{-1} [10].

Hazard Index: A widely used hazard index (reflecting the external exposure) called the external hazard index, H_{ex} is defined as follows [10].

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

In addition to external hazard index, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index, H_{in}, which is given by the equation [2],

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (4)$$

Where A_U, A_{Th} and A_K are the specific activities of ^{238}U, ^{232}Th and ^{40}K, respectively.

The values of the indices (H_{ex}, H_{in}) must be less than unity for the radiation hazard to negligible.

Annual Effective Dose Rate (AEDR): To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7Sv.Gy^{-1}) and outdoor occupancy factor (0.2) (i.e. people on the average spent 20% of the time outdoor) proposed by UNSCEAR [10] are used. Therefore, the annual effective dose rate (mSv.yr^{-1}) was calculated using the following formula [10].

$$\begin{aligned} AEDR(mSv/yr) &= D(nGy/h) \times 8760h/yr \times 0.7 \\ &\quad \times (10^3 mSv/10^9) nGy \times 0.2 \\ &= D \times 1.21 \times 10^{-3} (mSv/yr) \end{aligned} \quad (5)$$

To estimate the annual effective dose, the conversion coefficient from absorbed dose in air to effective dose (0.7Sv.Gy^{-1}) and indoor occupancy factor (0.8), (i.e. people on the average spent 80% of the time indoor) proposed by UNSCEAR [10] are used. Hence, the annual effective dose (mSv.yr^{-1}) was calculated using the following formula [10].

$$\begin{aligned} AEDR(indoor) &= D(nGy/h) \times 8760h/yr \times 0.7 \\ &\quad \times (10^3 mSv/10^9) nGy \times 0.8 \\ &= D \times 4.91 \times 10^{-3} (mSv/yr) \end{aligned} \quad (6)$$

Absorbed Dose Rate: the absorbed dose rate due to gamma radiation of naturally occurring radionuclide (^{238}U, ^{232}Th and ^{40}K) were calculated on guidelines provided by UNSCEAR [10].

$$D(nGy/h) = 0.462A_U + 0.621A_{Th} + 0.0417A_K \quad (7)$$

The maximum permissible dose rate is 51nGy/h [6].

Table 2. Activity concentration, Radium Equivalent (Raeq) and Absorbed Dose (D)

Sample Code	specific activity (Bq/Kg)			Ra_{eq}. (Bq/Kg)	D (nGy/h)
	^{40}K	^{238}U	^{232}Th		
EO1	147.17±3.69	14.61±1.92	19.99±0.77	54.53	25.3
EO2	136.80±3.42	12.47±1.75	14.65±0.60	43.95	20.56
EO3	122.01±3.23	9.50±1.39	12.87±0.67	37.3	17.47
EO4	137.30±3.71	10.80±1.45	12.92±0.74	39.85	18.74
EO5	214.12±4.34	7.84±1.32	14.71±0.66	45.36	21.69
EO6	63.75±3.55	16.36±1.78	0.03±0.77	21.31	10.24
IB1	62.69±4.00	38.50±2.38	30.59±0.82	87.07	39.4
IB2	65.33±2.95	13.74±1.98	16.43±0.70	41.81	19.28
IB3	75.15±2.64	11.42±1.75	12.19±0.58	34.64	15.98
IB4	60.70±3.75	28.12±2.94	29.64±0.94	75.2	33.93
IB5	47.81±3.29	17.10±2.57	18.49±0.74	47.22	21.38
IK1	101.91±3.24	13.90±1.98	14.96±0.65	43.14	19.96
IK2	80.31±3.49	13.65±1.76	17.15±0.70	44.36	20.42
IK3	34.65±2.56	5.12±0.38	4.31±1.15	13.95	6.49
IK4	77.86±2.93	11.89±1.89	14.97±0.62	39.29	18.04
IK5	86.06±3.96	17.56±2.12	12.42±0.64	41.95	19.41
Min.	34.65±2.56	5.12±0.38	0.03±0.77	13.95	6.49
Max.	214.12±4.34	38.5±2.38	30.59±0.82	87.07	39.4
Mean	94.60±3.42	15.16±1.84	15.40±0.73	44.26	20.37

Table 3. The hazard index (External, H_{ex}, and Internal, H_{in}) ($Bq.kg^{-1}$) and annual effective dose rate (AEDR) (Indoor and outdoor) for various L. G. A. in the study area

Sample Code	AEDR (Indoor)	AEDR (Outdoor)	H_{ex}	H_{in}
EO1	0.12	0.03	0.14	0.89
EO2	0.1	0.02	0.42	0.15
EO3	0.08	0.02	0.1	0.12
EO4	0.09	0.02	0.1	0.13
EO5	0.1	0.02	0.12	0.14
EO6	0.05	0.01	0.05	0.1
IB1	0.19	0.02	0.23	0.33
IB2	0.09	0.01	0.11	0.15
IB3	0.07	0.04	0.09	0.12
IB4	0.16	0.02	0.2	0.27
IB5	0.1	0.02	0.12	0.17
IK1	0.09	0.02	0.44	0.15
IK2	0.1	0.01	0.11	0.15
IK3	0.03	0.02	0.03	0.05
IK4	0.08	0.02	0.39	0.13
IK5	0.09	0.02	0.06	0.16
Min.	0.03	0.01	0.03	0.05
Max.	0.19	0.04	0.44	0.89
Mean	0.10	0.02	0.17	0.20

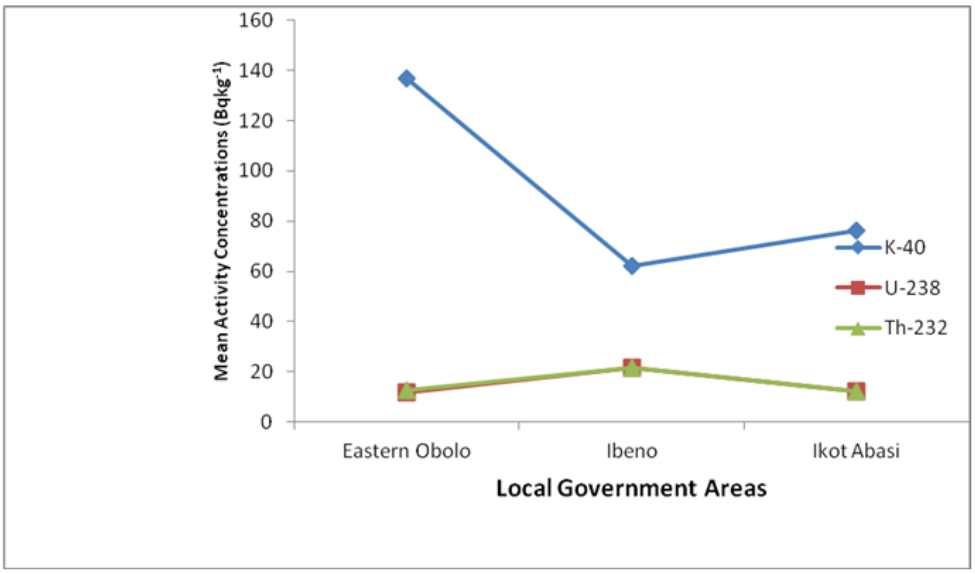

Figure 1. Mean activity concentration for ^{40}K, ^{238}U and ^{232}Th in top soil samples

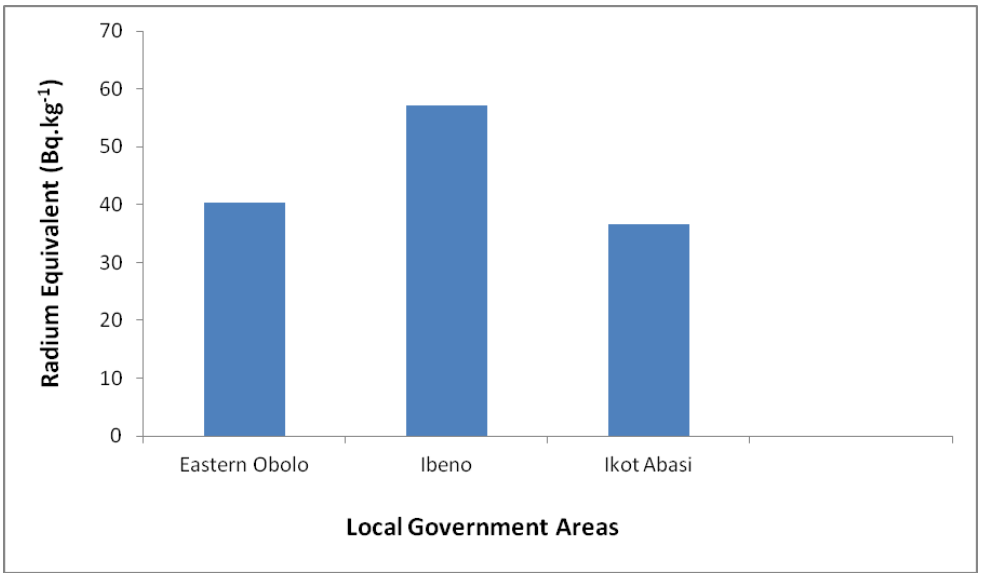

Figure 2. Mean Radium Equivalent in top soil samples

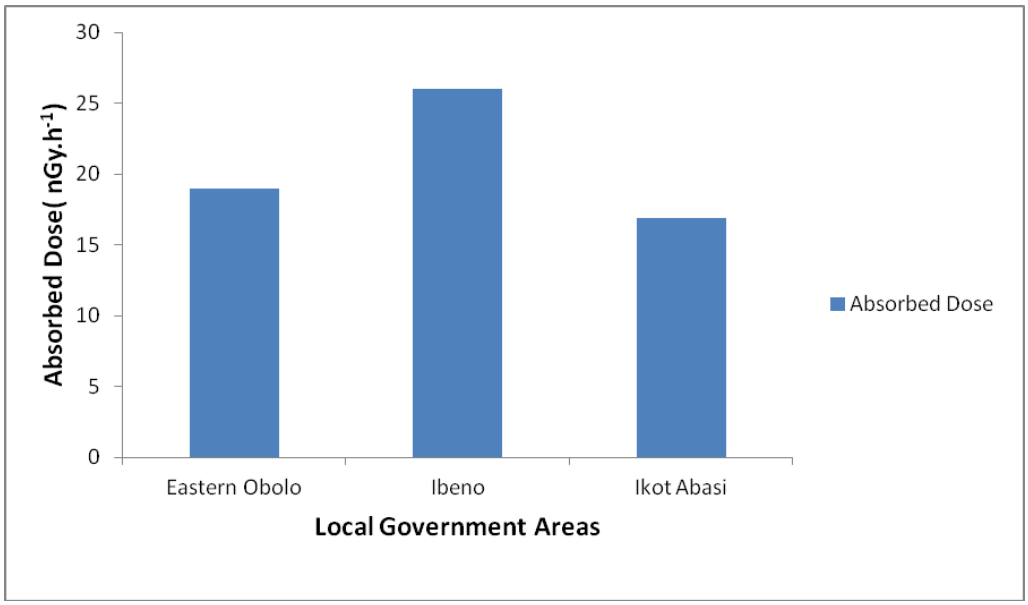

Figure 3. Mean Absorbed Dose Rate

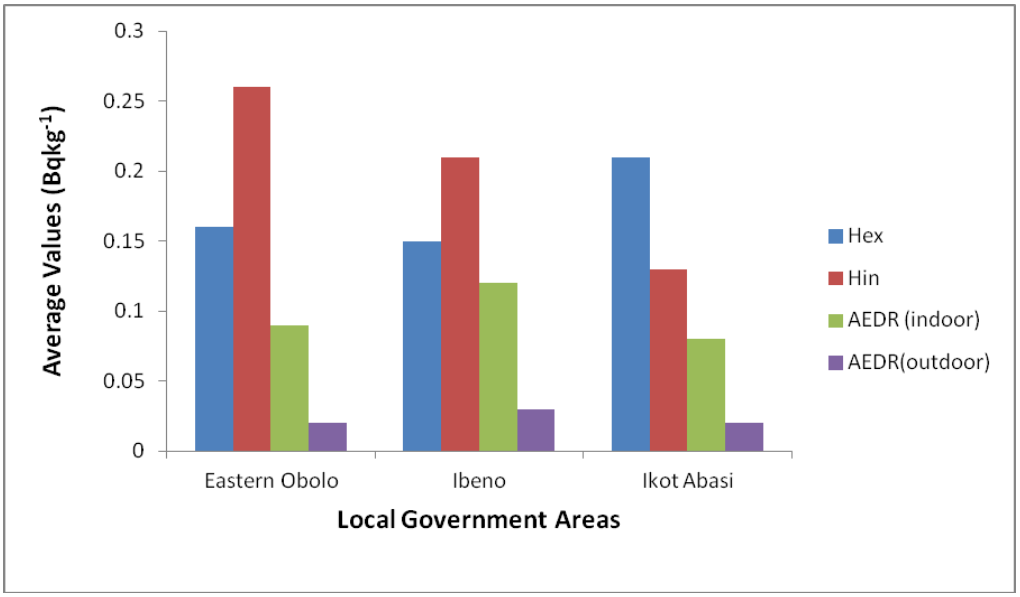

Figure 4. Hazard Index (Hex, Hin) and Annual Effective Dose Rate (indoor and outdoor)

4. Conclusion

The radioactivity concentrations of ^{40}K, ^{238}U and ^{232}Th in top soil samples collected from the study areas have been determined. The surface radiation dose rate at each sampling point has been measured. The Absorbed Dose, Annual Effective Dose, Radium Equivalent Activity and Radiation Hazard Index were estimated from the radioactivity concentrations measured in the study areas. From this study, the obtained values of gamma dose rate, radium equivalent activity, radiation hazard index and annual effective dose equivalent were found below the recommended safety limits. These are indications that the study area is safe for human activity. However, the data generated here may be useful for the introduction of radiation safety standards by the authorized organizations for the protection of general population from radiation hazards to terrestrial sources.

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