

Radiological safety assessment of soil samples at Kankara mining sites, Kankara Local Government Area of Katsina State, using Gamma-ray spectrometry

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ABSTRACT: The levels of natural radioactivity of ^{40}K , ^{226}Ra and ^{232}Th were investigated to check the safety limit with the aim of identifying and quantifying prominent Gamma Emitting Norms associated with mining sites. The specific activity concentration, gamma index, alpha index and hazards indices were calculated and compared to the world recommended value using Gamma Spectroscopy with NaI (TI) detector. The Specific activities of ^{40}K varied from 68.93098 to 98.74651 with a mean value of 84.720414; for ^{232}Th , 19,65476 to 48.29756 with a mean value of 35.192296; and for ^{226}Ra , 25.85647 to 41.89947 with a mean value of 34.50852. The mean values obtained for external hazard (H_{ex}), internal hazard (H_{in}), gamma index and alpha index were 0.245, 0.338, 0.634 and 0.170. Generally, with the exception of ^{232}Th and ^{226}Ra , the activity levels of the radionuclides investigated were low and within the world average value. The calculated hazard indices were lower than international recommended limit. The alpha index was found to be lower with the exception of gamma index.

Keywords: Gamma index, Kankara, Katsina State, radioactivity, soil.

INTRODUCTION

Soil is a major constituent of the earth making it suitable for living inhabitants to live. It constitutes 80 to 95% of the plant's growth (Veihmeyer and Hendrickson, 1927). It is vital for all known forms of life and man uses soil for various reasons such as agriculture and other domestic activities. Hence, its availability and quality as regard radiological, microbiological, chemical and any other form of contamination are delicate and vital issues. Natural radioactive activity has been in existence in our environment for decades ever since the creation of the universe. Occurring radioactive materials are widely spread in the earth's environment and exist in various geological formations such as rocks, soils, vegetation, water and air (Innocent et al., 2013). The natural occurring radioactive materials became the focus of regulatory interest with the publication of the International Atomic Energy Agency (Augustine and Aku., 2013). The levels of radionuclides distribution in the environment provide

essential radiological information when sites are being investigated. One thing to note is that soil from abandoned site may contain naturally occurring radionuclides in significant have been subject for study in the field of radiation protection and measurement (Faweya and Babalola, 2010). Workers been exposed to naturally occurring radioactive material (NORM) will continue to be an issue to the industry. For example, in the extraction of rare earths, the ^{226}Ra , ^{232}Th and ^{40}K . According to United Nations Scientific Committee on the effects of atomic radiation, about 13 million workers are exposed to natural sources of radiation (Innocent et al., 2013).

Radiation is a natural part of the environment in which we live. All people receive exposure from naturally occurring radioactivity in soil, water, air and food. One of the parts is that most of the abandoned mines are being turned into economics sites by some of the villages who engage in activities such as molding of mud bricks,

washing of motorcycles, some directly wash themselves and using some as relaxation joints by herdsmen with their cattle, thereby spending most of their time there almost every day. It is important to monitor the terrestrial background radiation mainly due to natural radio-nuclides in soil of those abandoned mine sites to ascertain the level of associated hazard indices (Innocent et al., 2013).

Radium (²²⁶Ra) is an alkaline earth metals which has a half-life of 1600 years and it is a decay product of ²²⁸Ra. Thorium (²³²Th) is a weakly radioactive chemical element, a decay product of Uranium (²³⁸U) and having a half-life of 14 billion years. Potassium (⁴⁰K) is a radioactive isotope of potassium which is an alkali metal and having half-life of 1.3 billion years. One of the interests of this work is to evaluate the radiological hazard indices using the activity concentration of the prominent gamma emitting NORMs (⁴⁰K, ²²⁶Ra, ²³²Th) associated with mine sites. With great hope that this research work will add to the database of some related studies carried out in other parts of the world. Also, the research assesses the measurement of ²²⁶Ra, ²³²Th and ⁴⁰K concentration level in five selected mining areas in Kankara, Katsina (Nigeria).

LITERATURE REVIEW

Radioactivity

Radioactivity is a keyword used to describe the disintegration of an atom. It involves the nucleus of an atom and not its extra nuclear electrons as in chemical changes as it is well known that some atoms are naturally unstable. These atoms in an attempt to become more stable emit particles and release energy (Duncan and Kennett, 2002). This phenomenon is called radioactivity and the unstable atoms involved are called radionuclides. The disintegration process proceeds at a definite rate through a certain number of stages until a stable end-product is formed.

Radioactive decay law

Radioactive lead to the emission of alpha, beta particles and gamma-rays due to the disintegration obey the statistical law of chance though it cannot be ascertained which particular atom is likely to disintegrate next. The law states that a radioactive substance decays exponentially with time (Nelkon and Parker, 1995). It is expressed mathematically as;

$$N_t = N_0 e^{-\lambda t} \text{-----1}$$

Where: N_0 = initial number of nuclei present at time, t equal zero, N_t = number of nuclei present after time t and λ is the radioactive decay constant.

Activity

The activity of a radioactive source (A) is the number of

disintegrations per second which is measure in Becquerel and expressed by calculus notation as described by Duncan and Kennett (2000);

$$A_t = \frac{dN_t}{dt} = -\lambda N_t \text{-----2}$$

Substituting for N_t from equation (1) into equation (2) gives

$$A_t = A_0 e^{-\lambda t} \text{-----3}$$

Where: A_t = Activity of the radioactive substance at time t and A_0 = Activity of radioactive substance at time t = 0.

MATERIALS AND METHOD

Location of the study area

Kankara is a local government area in Katsina State, Nigeria. It is located in Northern part of Nigeria and its coordinates are 11° 55'48" N and 07°24'25" E (Figure 1). It has an area of 1,462 km² and a population of 245,739 at the 2006 census (Population Census, 2006). The local government has 11 political wards which include; Burdugau, Danmurabu, Gundawa, Gurbi, Kankara, Ketare, Kukasheka, Mabai, Yargoje, Yar'Tsamiya and Zango. The people are predominantly farmers, cattle rearers and traders.

Geology of the study areas

Generally, the survey area lies in the Northwestern part of Nigeria (Figure 2). It is underlain by rocks of the basement complex, consisting mainly of migmatites, granites gnesis, biotitegnesis, metasediments of generally amphibolitesfacies metamorphism and granitic rocks of the older granite's suite augengnesis and mica schist were present, with mica schist dipping generally SE at angles varying from 27 to 43° (Hussaini and Daniel., 2016).

Materials used

The materials employed for this research work include cylindrical plastic containers, petroleum jelly, candle wax, matchbox, masking tape, wooden block, sieve, digital weighing balance, oven, NaI(Tl) detector, lead shielding, PC-based data acquisition system and printer.

Sample collection

The rock samples were collected by random sampling method from the mine sites at selected locations in five mine area of Kankara local government areas of the state. The soil samples were packed in plastic containers from



Figure 1. Map of Katsina state showing Kankara Local Government Area.

the areas of surveillance, properly sealed and labelled for easy identification and then transported to the Environmental Laboratory at Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria.

Sample preparation

In the laboratory at CERT Zaria, the soil samples were

placed in an oven and set to a temperature of 105°C to allow for drying overnight in order to remove any available moisture. The dried samples were gently crushed and sieved with a sieve of mesh size 2 mm to remove organic materials. Then after, the samples were homogenized and packed to fill cylindrical plastic containers of height 7 cm by 6 cm diameter to fit adequately into the lead shield housing the counting detector. This satisfies the selected optimal sample container height for the NaI(Tl) analytical instrument according to Ibeanu et al. (2000). The samples

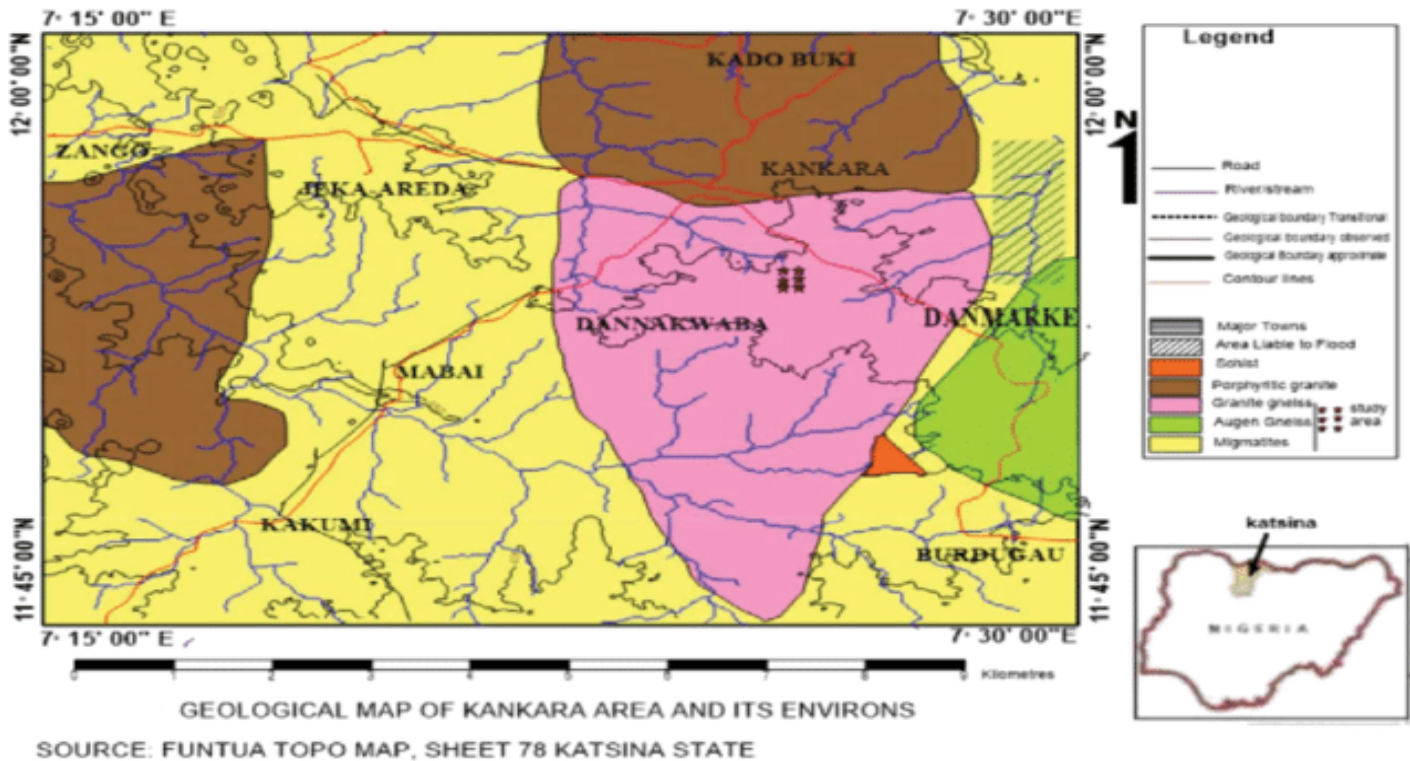


Figure 2. Geological Map of Kankara Area and its environs (Hussaini and Daniel., 2016).

were carefully sealed using petroleum jelly, candle wax and masking tape in order to prevent trapped radon gas from escaping. The sealed samples were kept for a minimum period of 30 days so as to allow for ²²⁶Ra and its short-lived progenies to reach secular radioactive equilibrium before gamma counting.

Experimental set-up

The gamma-ray spectrometry set-up consists of a 7.62 cm by 7.62 cm NaI(Tl) detector housed in a 6 cm thick lead shield and lined with cadmium and copper sheets in order to assist in the reduction of background radiation (Umar et al., 2012). In addition, a computer based multichannel analyzer (MCA) Maestro Programme from ORTEC was used for the data acquisition and analysis of gamma spectra.

Sample counting

The soil samples were placed on the detector surface and each counted for a lifetime of 29,000 s (8 h, 3 min) in reproducible sample detector geometry. The configuration and geometry were maintained throughout the analysis, as previously characterized based on well-established protocol of the laboratory at the Centre for Energy

Research and Training, Zaria. The background count was also measured for the same period of lifetime. The 1.764 MeV γ -line of ²¹⁴Bi for ²²⁶Ra was used in the assessment of the activity concentration of ²³⁸U while 2.615 MeV γ -line of ²⁰⁸Tl was used for investigating the activity concentration of ²³²Th while the single 1.46 MeV γ -line of ⁴⁰K was used for its content evaluation (Umar et al., 2012).

Computation of parameters

Measurement of external hazards

External radiation hazard index (H_{ex}) is a widely used hazard index which reflects the external exposure level due to gamma radiation. It is estimated from the relation as described by UNSCEAR (2000):

$$H_{ex} = \frac{ARa}{370} + \frac{Ath}{259} + \frac{AK}{4810} \tag{4a}$$

Measurement of internal hazards

The internal exposure to radiation is quantified by the internal hazard index (H_{in}) as defined by UNSCEAR (2000):

$$H_{in} = \frac{ARa}{185} + \frac{Ath}{259} + \frac{AK}{4810} \tag{4b}$$

Table 1. Activity concentration of the soil sample.

S/No	Sample ID	K-40 (Bq/Kg)	Ra-226 (Bq/Kg)	Th-232 (Bq/Kg)
1	S-G 1	98.74651	34.95463	28.98498
2	S-G 2	91.94968	29.95635	32.38547
3	S-G 3	68.93098	39.87568	46.63871
4	S-G 4	76.87628	25.85647	19.65476
5	S-G 5	87.09862	41.89947	48.29756
6	Minimum	68.93098	25.85647	19.65476
7	Maximum	98.74651	41.89947	48.29756
8	Mean	84.720414	34.50852	35.192296

Table 2. GPS location of the of the mining site of each sample collected.

Samples	Latitude	Longitude
S-G 1	11.94022 N 11° 56' 24.066"	7.414404 E 7° 24' 50.55"
S-G 2	11.94037 N 11° 56' 25.314"	7.414408 E 7° 24' 50.688"
S-G 3	11.94107 N 11° 56' 27.84"	7.41417 E 7° 24' 51.006"
S-G 4	11.94257 N 11° 56' 33.246"	7.41358 E 7° 24' 48.888"
S-G 5	11.94268 N 11° 56' 33.63"	7.41395 E 7° 24' 50.214"

Measurement of gamma index

The representative level of gamma index (I) was calculated using the following equation (UNSCEAR, 2000):

$$I_{\gamma} = \frac{ARa}{150} + \frac{ATh}{100} + \frac{AK}{1500} \quad 5$$

Measurement for alpha index

The alpha index for the natural radionuclides in the measured samples were computed using the following relation:

$$I_{\alpha} = \frac{ARa}{200} \quad 6$$

RESULTS AND DISCUSSION

The results of the radiological safety assessment of soil samples from 5 locations at Kankara mining site in Kankara Local Government Area of Katsina State, using Gamma Ray Spectrometry are shown in Tables 1 and 2. The specific activities for ^{226}Ra , ^{232}Th and ^{40}K are reported in Bqkg⁻¹. The specific activity of ^{226}Ra , ^{232}Th and ^{40}K in various soil samples vary from 25 to 41.89947, 19 to 48.29756 and 68.93098 to 98.74651 respectively. The worlds mean values of ^{226}Ra , ^{232}Th and ^{40}K activity concentration are 35, 30 and 400 Bqkg⁻¹ (UNSCEAR, 2000). A correlation was observed between specific

activities of ^{226}Ra and ^{232}Th in the collected soil samples. This clearly indicates the presence of Monazite and Zircon sand in the samples. Any increase in Thorium is observed being associated with similar increase in Radium and Potassium. It should be noted that the measured specific activity for ^{40}K were analogous to the world values, which were determined for soil and building materials. However, specific activity of ^{226}Ra in all of samples especially S-G 5 having 41.89947 Bqkg⁻¹ and ^{232}Th at S-G 5 having 48.29756 was much higher than the recommended world values (UNSCEAR, 2000). Hence, these samples pose some health hazard for the workers and occupants residing in these regions and for those using sample for building construction. The natural radioactivity levels in the soil are said to be small degree higher than the acceptable limit. Consequently, permanent settlers of these regions are exposed to an increased level of natural ionizing radiation from these samples. The mean activity concentration for ^{40}K ranged from 68.93098 to 84.720414 Bqkg⁻¹ with sample S-G 1 leading the activity concentration in Table 1. Similarly, the mean activity concentration for ^{226}Ra ranged from 25.85647 to 41.89947 Bqkg⁻¹ with S-G 5 leading the activity concentration. Also, the mean activity for ^{232}Th ranged from 19.65476 to 48.29756 Bqkg⁻¹ having S-G 5 leading the activity concentration. The highest activity concentration of ^{226}Ra was found in S-G 5 with 41.89947 Bqkg⁻¹. Though the value is high compared to the world wide average of 35 Bqkg⁻¹ (UNSCEAR, 2000). For ^{232}Th , the highest activity was found in S-G 5 with 48.29756 Bqkg⁻¹ whose value also was found to be higher than the recommended value of 30 Bqkg⁻¹ (UNSCEAR, 2000), which could be due to presence of abundant

Table 3. Activity concentrations, external hazard, internal hazard, gamma index and alpha index.

S/No	Sample ID	K- 40 (Bq/Kg)	Ra-226 (Bq/Kg)	Th-232 (Bq/Kg)	H _{ex}	H _{int}	I _γ	I _α
1	S-G 1	98.74651	34.95463	28.98498	0.227	0.322	0.579	0.175
2	S-G 2	91.94968	29.95635	32.38547	0.225	0.306	0.575	0.140
3	S-G 3	68.93098	39.87568	46.63871	0.302	0.410	0.778	0.199
4	S-G 4	76.87628	25.85647	19.65476	0.152	0.222	0.420	0.129
5	S-G 5	87.09862	41.89947	48.29756	0.317	0.430	0.820	0.209
6	Minimum	68.93098	25.85647	19.65476	0.152	0.222	0.420	0.129
7	Maximum	98.74651	41.89947	48.29756	0.317	0.430	0.820	0.209
8	Mean	84.720414	34.50852	35.192296	0.245	0.338	0.634	0.170

radioactive Thorium minerals such as Monazite, Zircon and thorianite (Okeyode and Akanni., 2009).

Table 3 shows the activity concentration, external hazard, internal hazard, gamma index and alpha index. For the radiation hazard to be acceptable, it is recommended that the H_{ex} be less than unity. The estimated H_{ex} for all the soil sample produced varies from 0.152 to 0.317 with the highest value noted in S-G 5, whereas the lowest value reported in S-G 4 as shown in Table 3. This highest value from the present study is lower than the recommended value which is 1.0. It is estimated using Equation (4b). For the safe use of the soil for construction purposes, H_{in} should be less than unity. The calculated values are shown in Table 3, ranging between 0.222 to 0.430 and the mean value of 0.338 for internal hazard (H_{in}) for soil samples in Kankara. The obtained results for H_{in} are below limit of unity which is 1.0. It is estimated using Equation (2). Table 3 presents the value for alpha index. It is noted that the upper limit of radon concentration (I_α) is equal to 1. The results of the present study shows that the radon concentration varies from 0.129 to 0.209 with average value of 0.170. Assessment of the alpha is another important aspect of hazard assessment that deals with the estimation of that excess alpha radiation due to radon inhalation originating from the soil. If the radon activity level in the soil exceeds the value of 200 Bqkg⁻¹, there is possibility that the radon exhalation from the soil could cause indoor radon concentration Bqm⁻³. The international commission on radiation protection recommends an action level of 200 Bqm⁻³ in dwellings. At the same time, if this Radium activity is below 100 Bqkg⁻¹, it shows that the radon exhalation from the soil may not likely cause indoor concentration greater than 200 Bqm⁻³. It is reported that the recommended exempted value and the recommended upper limit for radon concentration are 100 and 200 Bqkg⁻¹ respectively in the soil. The alpha index was estimated using Equation (4). Table 3 also presents the gamma activity index which is used to identify whether the dose criterion is met. The gamma index ranges from 0.420 to 0.820 with mean of 0.634. It is observed that the values are higher than the limit given as 1.0 (UNSCEAR, 2000). The gamma index was estimated using Equation (5). Table 2 shows the various sample ID

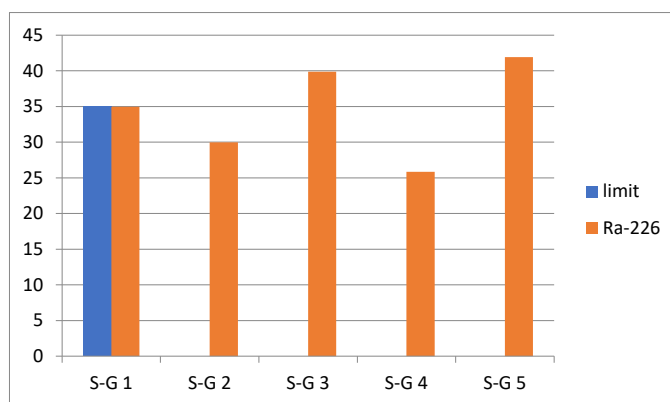


Figure 3. Chart of Ra-226 concentration.

with their coordinates.

Figure 3 gives a comparison of the measured values of the activity concentration for ²²⁶Ra with recommended limits which indicates that the concentration of ²²⁶Ra is greater at S-G 5 than the recommended value at some sampling sites, which shows that the dwellings is not considered safe. Similarly, a comparison of the measured value of ⁴⁰K activity concentration as shown in Figure 4 indicating the concentration of ⁴⁰K is lesser than the recommended value at every sampling site, which shows that the dwelling is considered safe. Figure 5 shows the comparison of the measured value of ²³²Th with recommended limits which indicates that the concentration is greater than the recommended value at some sampling sites, which indicates that the dwelling is not completely safe. Figure 6 shows the comparison of measured value of the external hazard index with recommended limits indicates the measured values is lesser than recommended value indicating the site is safe for dwelling. Figure 7 also gives a comparison of internal hazard index with its own recommended values which was found to be less than the recommended one indicating the site is safe for dwelling. Figure 8 shows the comparison of gamma index with its own recommended value which was found to exceed the recommended value indicating the site not good for dwelling due to its health effects. Figure 9 likewise

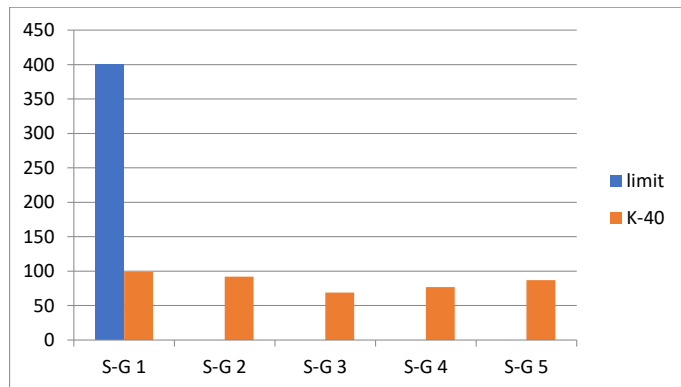


Figure 4. Chart of K-40 Concentration

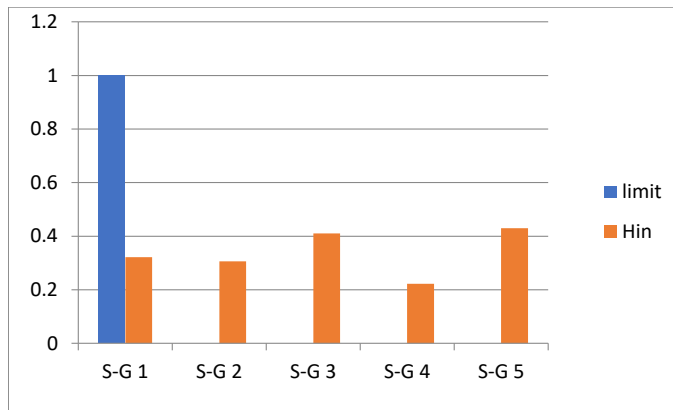


Figure 7. Chart of internal hazard.

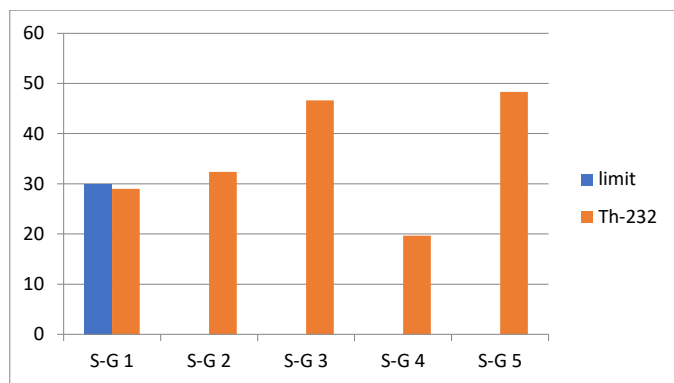


Figure 5. Chart of Th-232 concentration.

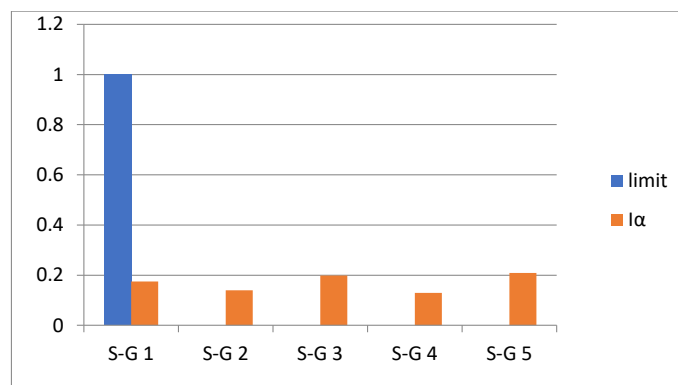


Figure 8. Chart of gamma index (I γ).

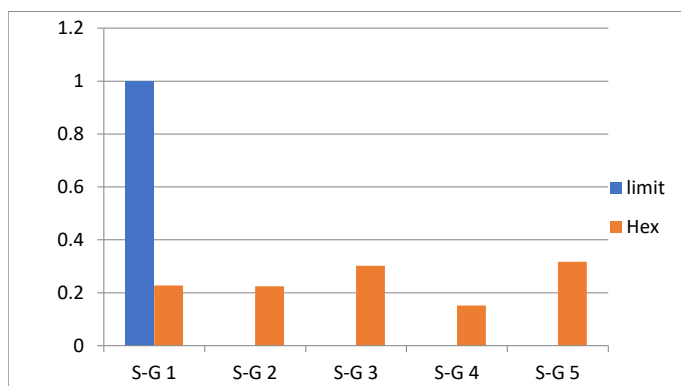


Figure 6. Chart of external hazard index.

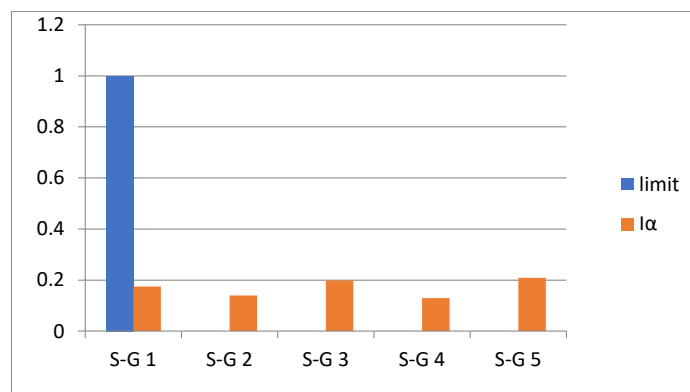


Figure 9. Chart of alpha index (I α).

shows the comparison of alpha index with its own recommended value which was found to be less than the recommended value indicating the site good for dwelling.

Conclusion

Conclusively, it was observed from the results that the

activity concentration of K40 in all the soil samples have values lower than the recommended value of 400Bq/Kg, for Ra226 two soil samples namely S-G3 and S-G5 have values higher than the recommended value of 35 Bq/Kg. Also, for Th232 three soil samples namely S-G2, SG3 and SG-5 have values higher than the recommended value of 30 Bq/Kg. All the calculated values for internal and external

hazard indices, Gamma index and Alpha index have values lower than the recommended value of 1.0. Since no documented research on the radioactivity measurement and radiological mapping of the soil samples in the area under investigation has been reported before now, the data generated here may be useful for the introduction of radiation safety standards by the authorized organizations for the protection of general population in the study area. Therefore, further research on this topic should be carried out using mining sites within the same region to validate the result obtained in this study.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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