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ARTICLE

Determination of ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U in soil samples from bauxite core deposits in western Cameroon

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Abstract – The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U in soil samples from a bauxite ore deposit site in Western Cameroon were determined by gamma spectrometry using a Broad-Energy Germanium (BEGe6350) detector. The average activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U were found to be 124.8 ± 58.1, 157.3 ± 67.3, 670.9 ± 272.3 and 5.8 ± 2.7 Bq kg^{-1}, respectively. The measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were compared with available data in some countries and the world average activity concentration of soil. The radiological hazard indices of radioactivity were calculated, with average values of 416.90 Bq kg^{-1} for the radium equivalent activity, 188.20 nGy h^{-1} for the gamma radiation dose rate, 0.23 mSv y^{-1} for the external outdoor annual effective dose, 0.92 mSv y^{-1} for the indoor annual effective dose, 1.02 mSv y^{-1} for the total annual effective dose and 1.13 for the external hazard index, respectively. These radiological hazard index values were also compared with the internationally approved values. Consequently, use of concrete bricks made of soil as building materials might lead to an increased radiation risk for the population of Fongo-Tongo.

Keywords: radioactivity / BEGe6530 detector / public external radiation dose / soil / bauxite

1 Introduction

Terrestrial radionuclides are present in the Earth's environment (soil, rocks, plants, water and air). The members of the radioactive decay chains of ^{232}Th and ^{238}U, along with ^{40}K, are the major contributors to the average annual radiation dose received in soft tissues by humans (UNSCEAR, 2000). The activity concentrations of these radionuclides in soil depend on the activity of the parent rock as well as the type of formation and transfer processes that are involved. In cores of such rock, weathering and soil formation, and chemical and geochemical interactions influence the distribution of primordial radionuclides and their decay products and ^{40}K. The geological and hydrogeological conditions of these rocks can sometimes lead to their enrichment in the environment, creating mineral deposits over a geological time scale. Because of the large deposits of minerals, wide deviations of natural radioactivity exist (UNSCEAR, 2000).

The western region of Cameroon is known to have large bauxite ore deposits. Since 1950, investigations have been carried out by the Foreign Office of Geological Mining Research. The objective of these investigations was to evaluate the mineral potential of the western region of Cameroon. As a result, an estimation of 54 000 000 tons of bauxite ore deposits can be found in the Menoua division of the western region. The project of exploiting the site is likely to become a reality soon. A study of the radiological impact of the bauxite ore deposits has not yet been undertaken. The population living in that area use bricks made of soil for house construction. Many studies worldwide reveal the radiological impact of natural mineral mining and milling on the environment (Winkelmann *et al.*, 2001; Carvalho *et al.*, 2007; Saïdou *et al.*, 2011; Ndontchueng *et al.*, 2014).

However, this impact cannot be well established without performing natural radioactivity measurements on site and in the vicinity of the site prior to mining activities. In addition, natural radioactivity measurements in soil using gamma spectrometry allow the implementation of precautionary measures whenever the dose is found to be above the recommended limits. The purpose of this work is to study the radioactivity level in top surface soil samples using the gamma spectrometry technique. Radium equivalent activity (Ra_{eq}) was calculated to assess the risk of using concrete bricks for house construction. The individual dose and risks to members of the public from an external exposure scenario (living in a house built with concrete bricks made with soil and near the bauxite ore deposit zone) given in UNSCEAR (2000) were also assessed as part of this study.

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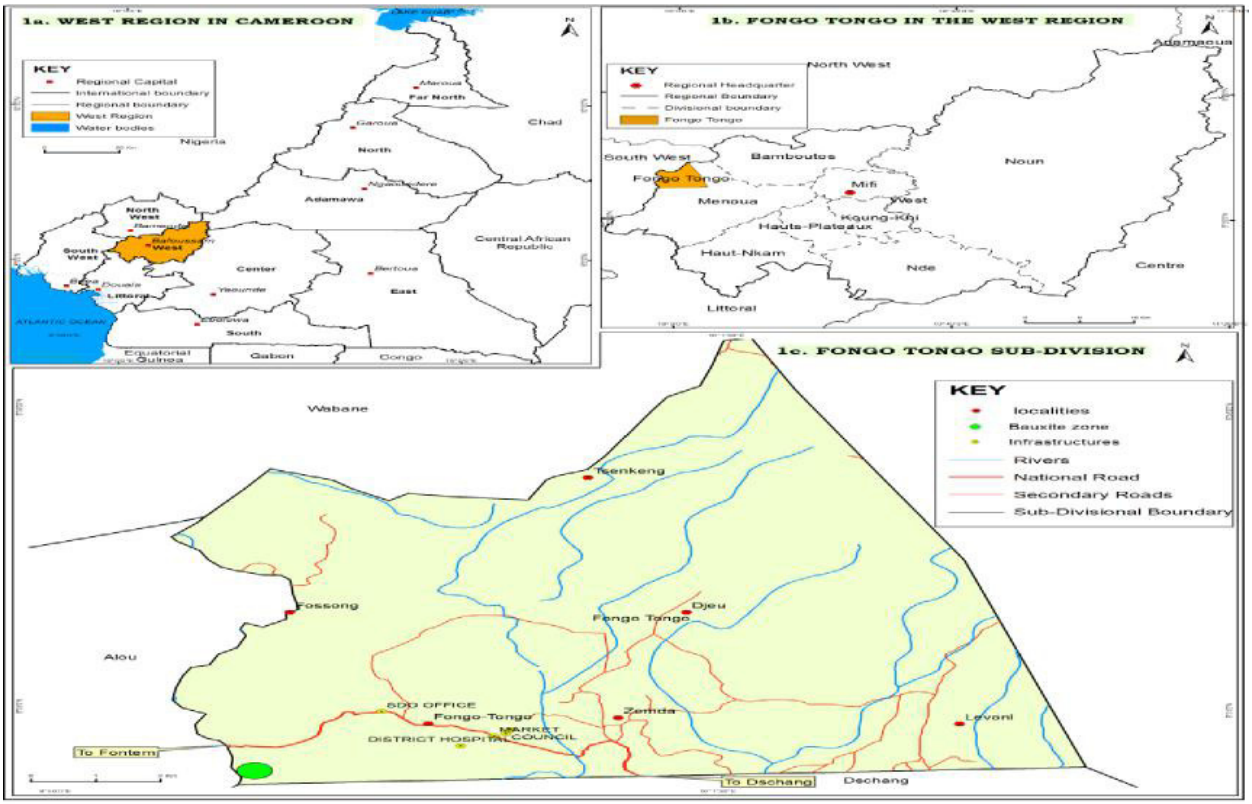

Figure 1. Map of the study area.

2 Materials and methods

2.1 Geology of the area and type of bauxite ore deposits

The study site was Fongo-Tongo, located in the highlands of western Cameroon, specifically the south-western side of Bambouto Mountains. The area is characterized by low altitude (1300−1600 m) and medium altitude (1600−2000 m). The vegetation is strongly influenced by anthropic activities and the crops cultivated include maize, beans, potatoes and tea (Wouatang *et al.*, 2014). The soils are ferralatic andic and ferralitic battleships according to CPCS (Commission de Pédologie et de Cartographie des Sols) classification. The petrography of this location consists of trachytes, welded ignimbrite, non-welded ignimbrite, basalts and granites. Rock types in the area are essentially the basement rocks (orthogneiss, gneiss, migmatites and amphibolites) and various volcanic rocks (basalts, trachytes, phonolites, tuffs, breccia and ignimbrites). The bauxite ore deposits in this area are developed exclusively from the Miocene aphyric or porphyric basalts. The mean chemical composition of these basalts is as follows: 15.9% of Al_2O_3, 13.5% of Fe_2O_3 and 44.6% of SiO_2 (Nouazi *et al.*, 2012).

2.2 Sample collection and preparation for radiological analysis

The field reconnaissance survey was based on the topography. This enabled us to identify the bauxite ore deposit site in the region. A total of twenty-five (25) composite soil samples were randomly collected at a typical depth of about 10 cm from the top surface layer. In order to cover the study area and to observe the significant local spatial variation in terrestrial radioactivity, each sampling location was considered as being overlaid by a 25 × 25 m grid, subdivided into 15 cells of 3 × 5 m, with a minimum distance between them of 1000 m. Within each grid, different samples were randomly collected and mixed to obtain composite samples. Table 1 presents the composite sample obtained in each grid. Each of these grids was marked using a global positioning system (GPS) as shown in Table 1. The samples were dehumidified for about 24 h at ambient temperature to provide a stable homogeneous mixture and then the same protocol was followed as described by Ndontchueng *et al.* (2014).

Table 1. Geographical coordinates of grid sampling.

Sample ID	Latitude	Longitude
SO1, SO2, SO3	05°31′49.9″N	09°58′31.4″W
SO4, SO5, SO6, SO7	05°31′50.7″N	09°58′35.7″W
SO8, SO9, SO10, SO11	05°31′53.2″N	09°58′36.9″W
SO12, SO13, SO14, SO15	05°31′46.8″N	09°58′33.1″W
SO16, SO17, SO18	05°32′40.4″N	09°59′45.3″W
SO19, SO20, SO21, SO22	05° 32’38.6”N	09° 59’24.2”W
SO23, SO24, S025	05° 32’33.5”N	09° 51’07.1”W

2.3 Instrumentation

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U in soil samples were measured using a high-resolution gamma-ray spectrometric system. The system consisted of a characterized Broad-Energy (BE6530) germanium detector shielded with lead to reduce background with an active volume of 6500 mm^3, relative efficiency of 60% at 1.33 MeV (^{60}Co line) and a resolution of 2.2 keV in the same line.

2.4 Calibration of the instrument and activity concentration

The energy calibration of the system was carried out using point sources of ^{109}Cd, ^{57}Co, ^{137}Cs and ^{54}Mn. The counting time was adjusted to record higher counts for each full-energy peak in order to minimize the statistical counting error. The absolute efficiency of the system was performed using ISOCS/LabSOCS mathematical calibration software with Monte Carlo, as described by Ndontchueng *et al.* (2014, 2015). Validation of the efficiency calibration files was done at the IAEA laboratory in Vienna through technical cooperation. To avoid error due to extrapolating the curve, the calibration curve was plotted in dual mode with cross-over energy at 165.85 keV (^{139}Ce).

To measure the activity concentration of nuclides in samples, each sample was counted for a time ranging between 24 and 48 h for effective peak area statistics of above 0.1%. Spectra were analyzed off-line using Genie 2000 Version V.3.2.1, including peak search nuclide identification activity and uncertainty calculation, and Minimum Detection Limit (MDL) calculation module software based on the equation taken from Aoun *et al.* (2015). Minimum detection activities of isotopes were 26.05 Bq kg^{-1} for ^{40}K, 7.55 Bq kg^{-1} for ^{226}Ra and 10.25 Bq kg^{-1} for ^{232}Th, respectively. The activity concentrations of the measured radionuclides were determined as described by Ndontchueng *et al.* (2014, 2015).

2.5 Dose calculations

As a result of non-uniform distribution of ^{226}Ra, ^{232}Th and ^{40}K in soil, uniformity with respect to exposure to radiation was defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. This was evaluated based on equations taken from Gonzalez-Fernandez *et al.* (2012); Uosif *et al.* (2015).

Assuming that the ^{235}U decay series can be neglected as it contributes very little to the total dose from the environmental background, the absorbed gamma dose rate outdoors at a height of 1m above the ground surface can be evaluated from the activity concentrations of the ^{226}Ra, ^{232}Th and ^{40}K measured in soil samples and the conversion factors of 0.0414 nGy h^{-1}/Bq kg^{-1} for ^{40}K, 0.623 nGy h^{-1}/Bq kg^{-1} for ^{232}Th and 0.461 nGy h^{-1}/Bq kg^{-1} for ^{226}Ra using the formula given in UNSCEAR (2000).

$$D(\text{nGy h}^{-1}) = 0.461\, A_{226\text{Ra}} + 0.623\, A_{232\text{Th}} + 0.0414\, A_{40\text{K}}.$$

As the population in the area uses concrete bricks as building materials, two external exposure scenarios were considered (outdoor (living within and near the study area) and indoor (living in a house built with concrete bricks made of soil) exposure). The outdoor and indoor annual effective doses (OAED and IAED) in units of mSv per year were then computed using conversion coefficient factors from the absorbed dose in air to the effective dose taken from UNSCEAR (2000). The total annual effective dose was then computed from the outdoor and indoor annual effective doses. The external hazard index (H_{ex}) was calculated using an equation taken from UNSCEAR (2000).

3 Results and discussion

3.1 Activity concentrations

The activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th and ^{235}U measured in the soil samples together with the statistical error are given in Table 2. The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th ranged from 135.5 ± 2.2 to 1269.2 ± 127.9 Bq kg^{-1} with an average value of 670.9 ± 272.3 Bq kg^{-1}, 52.8 ± 1.6 to 288.3 ± 21.7 Bq kg^{-1} with an average value of 124.8 ± 58.1 Bq kg^{-1}, and 58.84 ± 1.74 to 272.3 ± 11.3 Bq kg^{-1} with an average value of 157.3 ± 67.3 Bq kg^{-1}, respectively. The activity concentration of ^{40}K was observed to be comparatively higher than that of both ^{226}Ra and ^{232}Th in all the soil sampling locations of the studied area. The natural radioactivity of soil depends on the activity of the parent rock as well as the type of formation and transfer processes that are involved. In cores of such rock, weathering and soil formation, and chemical and geochemical interactions influence the distribution of primordial radionuclides, their decay products and ^{40}K. The rock types in this area are essentially basement rocks (orthogneiss, gneiss, migmatites and amphibolites) and various volcanic rocks (basalts, granites, trachytes, phonolites, tuffs, breccia and ignimbrites). Some of these basement and volcanic rocks such as gneiss, granites, basalts and orthogneiss are known to have more primordial radionuclides than others. From Table 2, ^{235}U was detected in twelve (12) of them, with an average value of 5.8 ± 2.7 Bq kg^{-1}.

The specific radioactivity of ^{226}Ra, ^{232}Th and ^{40}K of the soil samples was compared with available data in some countries and the worldwide average values, as seen in Table 3. As can be seen in this table, the activity concentration of terrestrial radionuclides found in soils varies from country to country. However, the values reported in Table 3 are not representative of the countries mentioned but only of the regions in which the samples were collected. The measured values of primordial radionuclides in this study were relatively higher than the measured values in South India (Gudalore), Upper Egypt, Northern India (Upper Siwaliks) and Turkey, except for the measured values of ^{226}Ra in Germany (Ronneburg uranium mining zone) and in Portugal (uranium mining site) and those of ^{40}K in Upper Egypt. This variation in radioactivity could result from the type of mineral ore deposit and the geotechnical characteristics of the area. Comparing the average values of ^{40}K, ^{226}Ra and ^{232}Th of the soil samples with the current worldwide mean, the means obtained are 73.56% more for ^{226}Ra, 71.39% more for ^{232}Th and 38.59% more for ^{40}K than the average worldwide.

Table 2. Activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th and ^{235}U measured in soil samples (Bq kg^{-1}).

Sample code	^{40}K	^{226}Ra	^{232}Th	^{235}U
SO1	1269.2 ± 127.9	86.9 ± 4.1	95.3 ± 4.3	4.3 ± 1.5
SO2	998.2 ± 29.4	94.6 ± 2.5	130.3 ± 3.9	5.6 ± 1.1
SO3	1004.9 ± 41.3	88.2 ± 4.2	88.9 ± 2.8	3.9 ± 1.4
SO4	1200.4 ± 98.3	119.7 ± 5.8	166.4 ± 7.1	5.3 ± 1.3
SO5	135.5 ± 2.2	154.6 ± 2.9	144.1 ± 4.9	6.9 ± 0.4
SO6	240.4 ± 4.3	97.7 ± 4.9	78.5 ± 6.1	5.6 ± 1.1
SO7	615.5 ± 82.7	116.0 ± 4.3	58.9 ± 6.5	4.5 ± 0.3
SO8	315.7 ± 12.1	62.2 ± 1.9	117.1 ± 2.8	3.2 ± 0.9
SO9	652.1 ± 25.1	199.3 ± 14.3	181.2 ± 7.4	ND
SO10	609.4 ± 22.1	175.4 ± 12.4	217.9 ± 4.2	ND
SO11	867.6 ± 45.3	118.0 ± 3.0	231.1 ± 5.4	ND
SO12	798.9 ± 41.2	288.2 ± 21.7	256.1 ± 10.6	ND
SO13	830.8 ± 39.2	106.1 ± 4.8	192.3 ± 4.7	ND
SO14	678.5 ± 21.8	172.4 ± 7.7	199.9 ± 9.4	ND
SO15	800.8 ± 41.3	108.01 ± 4.8	206.3 ± 8.5	ND
SO16	549.4 ± 29.3	117.7 ± 5.2	272.3 ± 11.3	5.4 ± 0.4
SO17	498.9 ± 20.1	209.6 ± 9.1	236.1 ± 4.9	8.9 ± 1.4
SO18	500.4 ± 30.1	113.1 ± 4.9	250.9 ± 5.5	6.1 ± 0.4
SO19	964.7 ± 57.5	127.3 ± 3.3	271.1 ± 11.3	6.3 ± 3.8
SO20	898.4 ± 50.5	91.8 ± 4.2	175.9 ± 7.3	ND
SO21	909.9 ± 52.4	153.3 ± 6.9	212.9 ± 8.5	ND
SO22	698.3 ± 49.0	107.8 ± 3.5	174.2 ± 4.4	ND
SO23	809.3 ± 40.4	270.3 ± 20.2	212.9 ± 8.5	ND
SO24	727.9 ± 39.1	150.3 ± 12.8	86.8 ± 2.1	ND
SO25	867.6 ± 49.8	52.8 ± 1.6	58.8 ± 1.7	ND
Min	135.5	52.8	58.8	ND
Max	1269.2	288.2	272.3	8.9
St. Dev.	272.3	58.1	67.3	2.7
Average val.	670.9	124.9	157.3	5.8

ND = Not Detected.

Table 3. Comparison of the activity concentration of terrestrial radionuclides with other published values.

	Activity concentration (Bq kg^{-1})			
Location	**^{226}Ra**	**^{232}Th**	**^{40}K**	**References**
South India (Gudalore)	17–62	19–272	78–596	Selvasekarapandian *et al.*, 1999
Northern India (Upper Siwaliks,)	28.3–81.0	61.2–140.3	363.4–1002.0	Singh *et al.*, 2005
Upper Egypt	31–40	52–61	3149–3210	Amin and Uosif, 2011
Camroon (volcanic area)	14.00	30	103	Ngachin *et al.*, 2008
Turkey	28.60	33	448.5	Turhan *et al.*, 2012
Portugal (uranium mining)	200.00	91	- - - -	Carvalho *et al.*, 2007
Eastern Germany (Ronneburg)	370.00	45	620	Winkelmann *et al.*, 2001
Cameroon (Fongo-Tongo)	52.8–288.2	58.8–272.3	135.5–1269.2	Present study
World average	16–116(33)	7–50(45)	100–700(420)	UNSCEAR, 2000

Table 4. Radiological health hazard parameters in the soil samples under investigation.

Sample code	Ra_{eq} (Bq kg^{-1})	Absorbed dose (nGy h^{-1})	OAED (mSv y^{-1})	IAED (mSv y^{-1})	H_{ex}
SO1	320.9	150.7	0.18	0.74	0.87
SO2	357.8	164.0	0.20	0.80	0.97
SO3	292.7	136.4	0.17	0.67	0.79
SO4	450.1	205.9	0.25	1.01	1.22
SO5	371.1	164.1	0.20	0.80	1.00
SO6	228.5	102.6	0.13	0.50	0.62
SO7	247.6	114.9	0.14	0.56	0.67
SO8	253.9	112.6	0.14	0.55	0.69
SO9	508.6	228.7	0.28	1.12	1.37
SO10	533.9	238.1	0.29	1.17	1.44
SO11	515.4	230.3	0.28	1.13	1.39
SO12	715.9	321.2	0.39	1.58	1.93
SO13	445.4	199.9	0.25	0.98	1.20
SO14	510. 5	228.7	0.28	1.12	1.38
SO15	464.7	207.9	0.25	1.02	1.25
SO16	549.3	241.7	0.30	1.19	1.48
SO17	585.7	260.3	0.32	1.28	1.58
SO18	510.3	224.6	0.28	1.10	1.38
SO19	589.2	262.8	0.32	1.29	1.59
SO20	412.6	186.2	0.23	0.91	1.11
SO21	527.8	237.4	0.29	1.16	1.43
SO22	410.7	184.2	0.23	0.90	1.11
SO23	637.1	287.2	0.35	1.41	1.72
SO24	330.5	152.2	0.19	0.75	0.89
SO25	203.7	96.1	0.12	0.47	0.55
Min	203.7	96.1	0.12	0.47	0.55
Max	715.9	321.2	0.39	1.58	1.93
St. Dev.	135.6	59.4	0.07	0.29	0.37
Geom. Mean	416.9	188.2	0.23	0.92	1.13
World Mean	370	60	0.07	0.46	

3.2 Dose assessment

Table 4 presents the calculated data of the radium equivalent activity (Ra_{eq}), absorbed dose rate (AD), outdoor annual effective dose (AED), indoor annual effective dose (IAED) and external hazard index (Hex) evaluated for the soil samples of Fongo-Tongo. The radium equivalent activity (Ra_{eq}) due to the terrestrial radionuclides ^{226}Ra, ^{232}Th and ^{40}K varied from 203 to 715 Bq kg^{-1}, with an average value of 416 ± 135 Bq kg^{-1}. The average Ra_{eq} value of the present study is relatively higher than the criterion limit of 370 Bq kg^{-1}. This implies that using soils from this area as a building material might increase radiation exposure to the public. The outdoor gamma radiation dose rate in air at 1 m above the ground surface for the distribution of the primordial radionuclides (^{226}Ra, ^{232}Th and ^{40}K) varied from 96 to 321 nGy h^{-1}, with an average value of 188.2 ± 59.4 nGy h^{-1}, which is higher than the population-weighted average value of global primordial radiation of 60 nGy h^{-1} reported by UNSCEAR (2000). This difference in the absorbed dose rate from the values reported by UNSCEAR (2000) could be attributed to differences in the geology formation and geochemical structure of the sampling sites. The contribution of each primordial radionuclide to the average gamma absorbed dose rate is given in Table 5. It can be seen that ^{232}Th contributes more to the absorbed dose rate in air.

The values of the outdoor annual effective dose (OAED) varied from 0.12 to 0.39 mSv y^{-1}, with an average value of 0.23 ± 0.07 mSv y^{-1}. Similarly, the indoor annual effective dose rate (IAED) varied from 0.47 to 1.58 mSv y^{-1}, with an average value of 0.92 ± 0.29 mSv y^{-1}. The total annual effective dose rate derived from both the indoor and outdoor effective doses varied from 0.59 to 1.97 mSv y^{-1}, with an average value of 1.02 mSv y^{-1}. The recommended limits of the International Commission on Radiological Protection (ICRP, 1991) for individual members of the public is 1 mSv y^{-1}, the worldwide average annual effective dose is approximately 0.5 mSv

Table 5. Natural radionuclides in soil and the corresponding absorbed dose rate in air 1 m above the ground.

Nuclide	World average (UNSCEAR, 2008)		Mini-Matap (study area)		
	Concentration (Bq kg^{-1})	Dose rate (nGy h^{-1})	Concentration (Bq kg^{-1})	Dose rate (nGy h^{-1})	% Contribution
^{226}Ra	33	25	124.85	53.31	28.6
^{232}Th	45	45	157.27	104.11	55.85
^{40}K	420	30	670.92	28.98	15.55

and the results for individual countries are generally within the 0.3−0.6 mSv range for indoors. From the results obtained, the average indoor annual effective dose is slightly higher than the recommended limits for indoors. When comparing the observed annual effective dose with the worldwide limits, this was relatively equal to the ICRP limits for members of the public. The calculated external hazard index is given in Table 3. From a radiation protection point of view, this index value must be less than unity in order to keep the radiation hazard below that of significant health risks. The calculated values ranged from 0.55 to 1.93, with an average value of 1.1 ± 0.3. Since this average value is slightly higher than unity, soil from this region might not be safe and might increase the radiological threat to the population when used as a building material.

4 Conclusion

In this study, distribution of ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U in the surface soil samples of a bauxite ore deposit site in Western Cameroon was determined by gamma spectrometry using a Broad-Energy Germanium (BEGe6350) detector. The results were as follows:

- The average activity concentrations of the primordial radionuclides ^{226}Ra, ^{232}Th and ^{40}K were observed to be higher than the world average values reported by UNSCEAR.
- The results obtained show that the radiological hazard indices such as the radium equivalent activity and external hazard index are slightly higher than the world average values. This implies that gamma radiation from soil in this area might increase the radiological threat when used as a building material.
- The outdoor gamma dose rate for the soil samples in this study is higher than the world average value of 60 nGy h^{-1}.
- The results obtained show that the average outdoor and indoor effective annual doses due to the natural radioactivity of the soil samples are lower than the recommended value of 1 mSv y^{-1}.

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