Assessment of population doses and radiation hazards from rock samples collected in the South West of Cameroon

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Abstract

The concentrations of the natural radionuclides: ^{226}Ra, ^{232}Th, ^{40}K, have been determined in syenites which are typical rocks from Awanda, Bikouéssi, and Ngombas, localities in the South West of Cameroon. A HPGe p-type detector coupled to a multichannel analyzer was used to perform measurements and data processing. The highest average value of ^{226}Ra concentration (4026±270 Bq/kg) was observed at Ngombas whereas the highest average value of ^{232}Th concentration (1385±176 Bq/kg), were detected at Awanda. However ^{40}K contributes significantly to increase the radioactivity in rocks from the three locations studied in this work. The radium equivalent activity Ra_{eq}, the external hazard index H_{ex}, the outdoor absorbed dose rate D in air and the annual effective dose equivalent E were evaluated to external radiation exposure for people living to studied areas. The mean absorbed dose rate in air is 1427.49 n Gy/h at 1.0 m above the soil.

Keywords: Natural radioactivity, syenite, rock, HPGe detector, effective dose, Cameroon.

Introduction

Radioactivity in the environment consists of the three well known radioactive series of uranium, thorium and actinium which respectively originate from radionuclides ^{238}U, ^{232}Th and ^{235}U. Several singly occurring radionuclides are also present in the environment but the most important of them is ^{40}K. The members of the radioactive decay chain of ^{232}Th (14%), ^{235}U and ^{238}U (55.8%) along with ^{40}K (13.8%) are responsible for the main contributions to the dose from natural radiation[1]. The average annual effective dose associated with external exposure to terrestrial radionuclides is about 0.46 mSv shared as follow: 0.12 mSv due to the ^{40}K, 0.21 mSv for the ^{232}Th series and 0.13 mSv for ^{238}U series[2]. The ratio of ^{235}U to ^{238}U is less than 1% hence the contribution of ^{235}U to the environmental dose is very small[3].

Radionuclides are present in rocks in varying amounts, and they are easily mobilized into the environment. Radioactivity in soil results from the rocks from which they were derived. The distributions of naturally occurring radionuclides depend on the distribution of rocks from which they originate and the processes which result to their removal from the soil and migrate them. Therefore, the natural environmental radioactivity mainly depends on geological and geophysical conditions[4]. The concentration of natural radionuclides in the rock varies considerably depending on the rock formation and lithologic character [3,5-7]. The high radiometric values in soils are good indicators of enriched source rocks[8]. It is therefore important to measure the concentration of radionuclides in the environmental rocks in order to assess the radiological risks on humans.

In Cameroon, all rocks of sedimentary origin during Precambrian became subjected to folding, metamorphism and volcanic eruptions. The formations of the studied area are Palaeoproterzoic and belong to the Nyong Group and are cross-cut by Pan-African intrusive shown in the fig.1. Radioactive source rocks of this area comprise syntectonic syenites which appear to be linked with shear zones[8]. The objective of this present work is to determine the specific radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in rock samples collected from locations in the South West of Cameroon with the aim to assess their contribution in the external dose exposure and the radiation hazards associated for population inhabiting investigated areas. The average radium equivalent activity (Ra_{eq}), the total absorbed dose rate (D), the external hazard index (H_{ex}) and the annual external effective dose rate (E**)** have been estimated and compared with the recommended limits from UNSECAR data.

Material and Methods

Physiographic sitting: Studied area is located on Akongo-Lolodorf syenitic axis extending on about 75 km in the South West of Cameroon, from geological period of Precambrian. From 1978 to 1985, airborne and scintillametric surveys, coupled with geochemical studies carried out by the French office of geological and mining research (BRGM) revealed about fifteen radiometric anomalies identified in the localities of Ngombas (11°06'E; 3°25'N), Awanda (10°59' 30"E; 3°22'N), Bikoué (10°51' E; 3°21'N), and Madong (10°44'E; 3°17'N). The radiometric anomalies zones are concentrated along the Lokoundje River Basin which dissects through radioactive syenite sources[8].

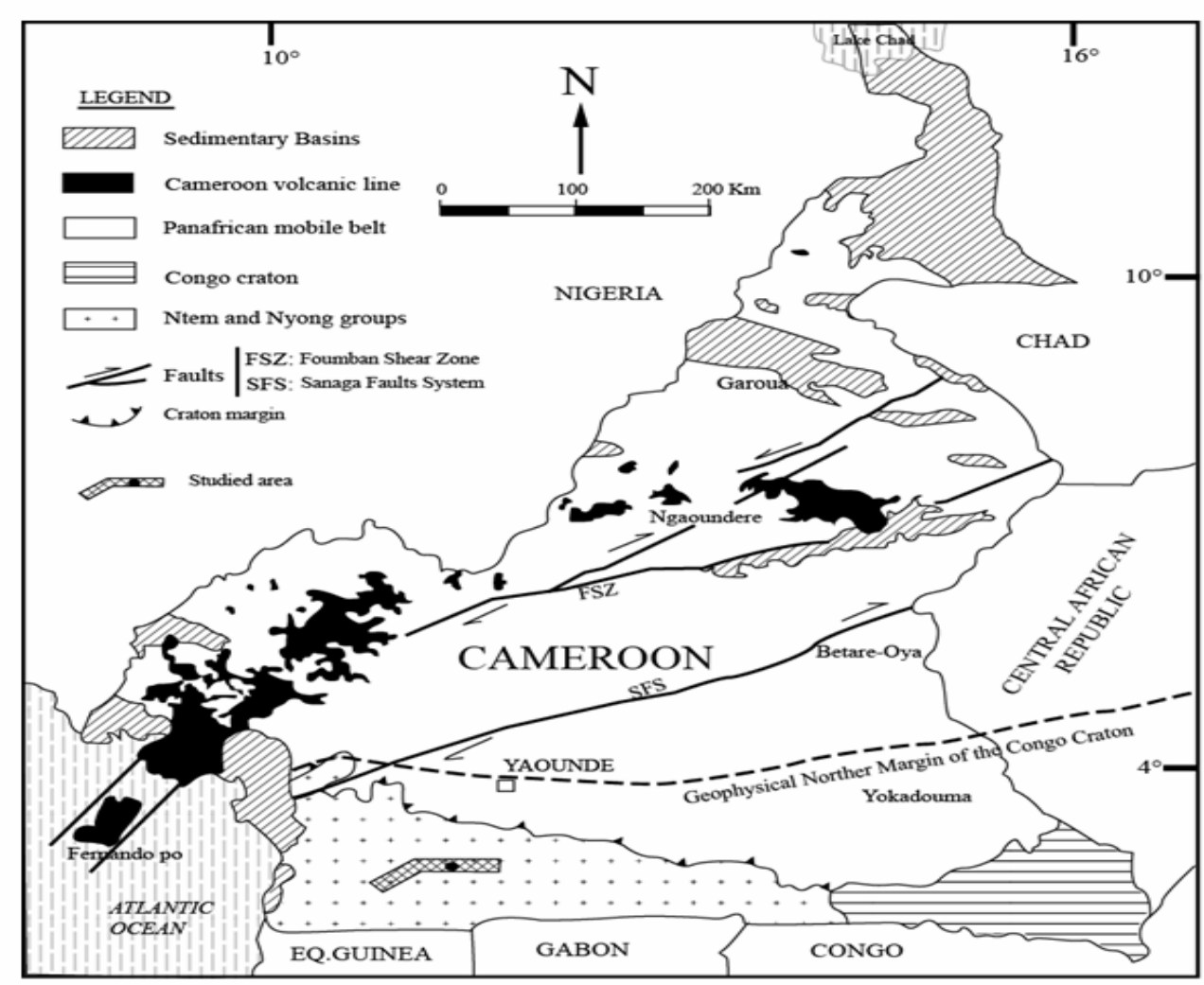

Figure 1: Geological map of Cameroon showing localisation of studied area

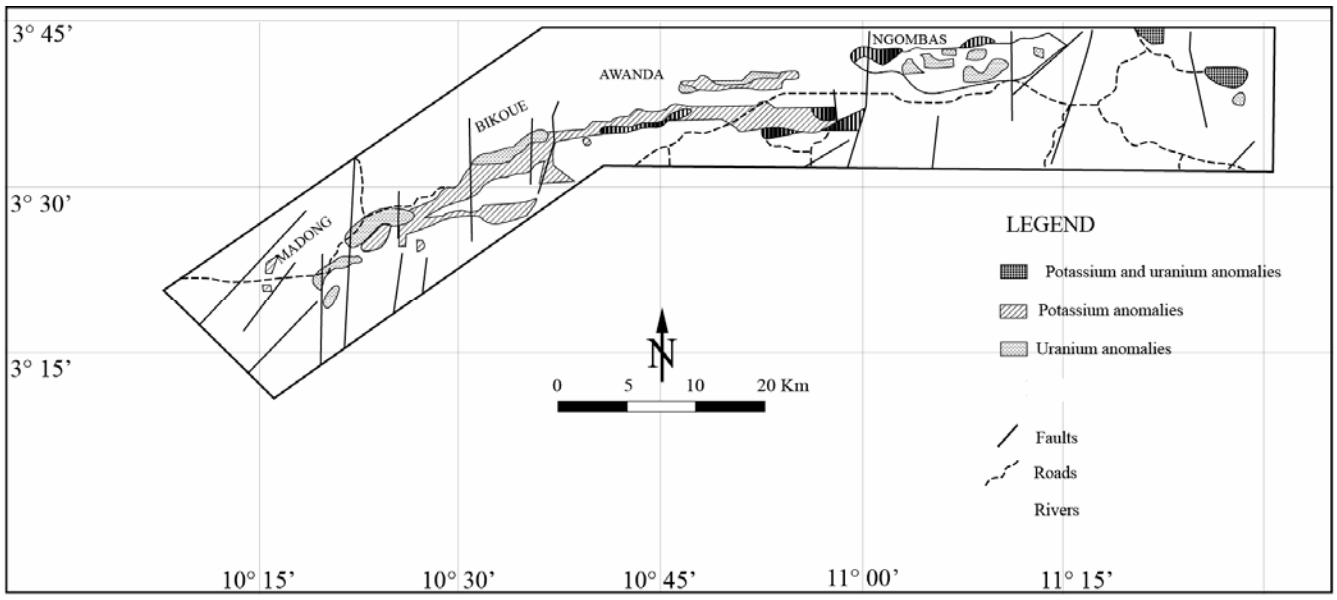

Figure 2: Airborne radiometric anomalies map showing sampling locations in South West of Cameroon 1-Ngombas; 2-Awanda; 3-Bikoué

Fig.2 shows the spatial distribution of radiometric anomalies on the studied area. Some anomalies are narrow thin blades not exceeding a few hundred meters thickness. Surface areas investigated are inside the equatorial forest where people leave and carry out agricultural activities. Our investigations in this work were limited to the first three localities because of their high population density.

Sample collection and preparation: Fifteen samples from fixed rocks were collected in the localities of Ngombas, Awanda and Bikoué. Identification of the rock samples was done at the Earth Science Department of the University of Yaoundé I and the Institute of Geological and Mining Research. Typical litholigic formations from studied areas are constituted by intrusive rocks, specifically alkalines syenites[8]. Syenite is a coarse-grained intrusive igneous rock of the same general composition as granite but with the quartz either absent or present in relatively small amounts (<5%)[9].

The in situ background radiation of each sampling point has been probed before sampling by the ambient radiometer Graetz x 5 DE, which has been calibrated in the dosimetry laboratory of the "Centre National de l'Energie des Sciences et des Techniques Nucléaires (CNESTEN)" in Morocco. Values of background measured are ranging from about 1.20 to 2.01 µSv/h in Ngombas, 0.82 to 1.40 µSv/h in Awanda, 0.20 to 0.70 µSv/h in Bikoué. The detection limit of the radiometer is 0.06 µSv/h. A relatively high natural background radiation region was measured in Ngombas and Awanda than Bikoué. Therefore, a laboratory measurement of geologic samples from the studied area was needed to determine the relative concentration of radioactive elements ^{226}Ra, ^{232}Th and ^{40}K contributing to the high radiation level.

In the laboratory, rock samples were crushed to homogenize them, dried in a desiccator at 200° C for 12 hours and finally sieved using a sieve meshes lower than 2 mm. Each sieved sample was sealed and stored in cylindrical plastic container (SG 50, standardized by the CEA) of 50 ml. The sieved samples were sealed and stored for one month before the measurement to allow that parent radionuclides and their respective descendants to reach secular equilibrium[10].

Radioactivity measurements: The activity concentrations of rock samples were measured using a Canberra low-level gamma counting system (SAGA 0930-7), with a high-resolution HPGe detector (p-type, GR3019) coupled to an analyzer multi channel (Model DESA). The energy resolution of the 1332 keV line from ^{60}Co was found to be 1.93 keV at full width of half maximum (FWHM) with a relative efficiency of 30%. The data acquisition and analysis were carried out by the software package, GENIUS 2000. The gamma spectrum of each rock sample was determined by counting for 54 000 seconds. Samples were presented to the detector automatically; this configuration saves time and keeps the geometry sample-detector fixed during the counting. To reduce the background radiation, a 5-cm thick lead castle was used to shield the detector. This has been constructed to establish a low background radiation[11].

The detector system was systematically calibrated using a standard multi-gamma reference sample (N° 71863/3) of 50 ml which contains certified concentrations of following radionuclides: ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{137}Cs, ^{88}Y, ^{113}Sn, ^{85}Sr, ^{51}Cr and ^{60}Co and emitting gamma rays in the energy interval 59-1900 keV. All spectrometric measurements of this work were carried out in the "Unité de Surveillance de l' Environment (USE)". For a nuclide having more than one peak in the spectrum, the activity is obtained as the weighted average activity obtained at each peak. With the full energy gamma-ray peaks of ^{40}K and radionuclides in the ^{238}U and ^{232}Th decay series, their activities were calculated. Then the activities of the parents were obtained using the weighted average of the daughter activities assumed to be in secular equilibrium[12]. The gamma-transitions of 186.3 keV (^{226}Ra), 351.9 keV (^{214}Pb), 609.3 keV (^{214}Bi), 768.4 keV (^{214}Bi), 1120.3 keV (^{214}Bi), 1238.0 keV (^{214}Bi) and 1764.0 keV (^{214}Bi) were used to determine the concentrations of the ^{226}Ra. The γ-transitions of 238.0 keV (^{212}Pb), 338.0 keV (^{228}Ac), 583.1 keV (^{208}Tl), 911.2 keV (^{228}Ac), 968.3 keV (^{228}Ac) and 974.2 keV (^{228}Ac) were used to determine the concentration of the ^{232}Th. The 1460.0 keV gamma-transition of ^{40}K was used to determine the concentration of ^{40}K[13].

Results and Discussion

Activity concentrations: The specific activities (in Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K for rocks samples collected from Ngombas, Awanda, Bikoué in South western Cameroon are listed in table 1. The average concentration of ^{226}Ra was found to be 4026±270 Bq/kg in Ngombas (with a minimum of 704±50 and a maximum of 7256±364 Bq/kg), 317±31 Bq/kg in Awanda (with a minimum of 61±4 and a maximum of 836±58Bq/kg), 402±34 Bq/kg in Bikoué (with a minimum of 70±6 and a maximum of 570±40 Bq/kg).

The average concentration of ^{232}Th was found to be 1044±75 Bq/kg in Ngombas (with a minimum of 633±43 and a maximum of 1423±99 Bq/kg), 1385±176 Bq/kg in Awanda (with a minimum of 90±6 and a maximum of 5498±390 Bq/kg), 683±58 Bq/kg in Bikoué (with a minimum of 261±18 and a maximum of 1128±85 Bq/kg) while the normal average value is 50 Bq/kg[2,14]. Fig. 3 shows a histogram representing the average concentration of ^{226}Ra, ^{232}Th and ^{40}K from rocks at different locations. It can be viewed that the average values of activity in different locations are much higher than normal and are not uniformly distributed in rocks from one location to another. Rocks from Ngombas are more enriched with uranium than thorium while rocks from Awanda and Bikoué are more enriched with thorium than uranium. ^{40}K contributes significantly to increase the radioactivity in rocks.

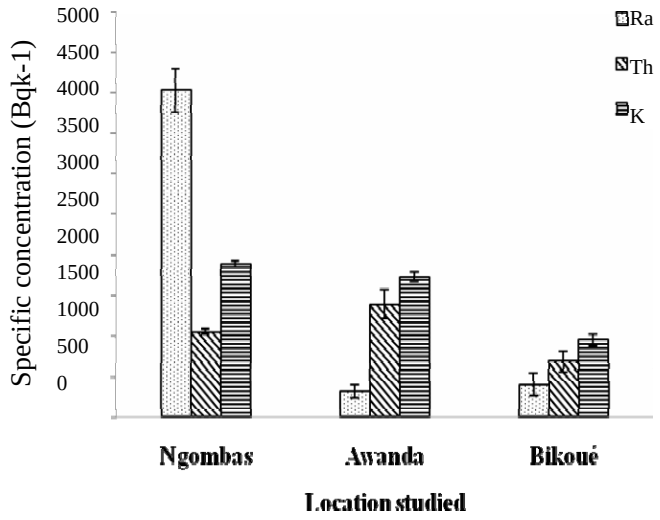

Figure 3: Histogram representing the average concentration of ^{226}Ra, ^{232}Th and ^{40}K from rocks at different locations

The average concentration of ^{40}K was found to be 1879±134 Bq/kg in Ngombas (with a minimum of 542±37 and a maximum of 2505±192 Bq/kg), 1725±124 Bq/kg in Awanda (with a minimum of 1470±100and a maximum of 2214±160 Bq/kg), 941±68 Bq/kg in Bikoué (with a minimum of 747±53and a maximum of 1340±96 Bq/kg) while the normal average value is 500 Bq/kg[2]. The total concentration values of, ^{226}Ra ^{232}Th, ^{40}K in all rock samples collected are respectively: 1582 ±158 Bq/kg**,** 1037±116 Bq/kg and 1515 ±113 Bq/kg.

Radium equivalent activity and external hazard index : To represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity that takes into account the radiation hazards associated with them, a common index has been introduced and is called "radium equivalent activity" (Ra_{eq}). It is defined as Beretka and Mathew[15]:

$$Ra_{eq} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_K, \quad (1)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K respectively. While defining Ra_{eq}, activity according to equation (1), it has been assumed that 1 Bq/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th and 13 Bq/kg of ^{40}K produce the same γ-ray dose rate [13]. The Ra_{eq} is related to the external gamma dose and the internal dose due to radon and its daughters [16]. In the present work, Ra_{eq} is estimated for the collected rock samples and are given in table 2. The average values of Ra_{eq} were found respectively to be 5663 Bq/kg, 24301 Bq/kg and 1451 Bq/kg for Ngombas, Awanda and Bikoué. It is clear that the estimated means values of Ra_{eq} in rocks from Ngombas, Awanda, Bikoué in the South West of Cameroon, are much higher than the recommended maximum value 370 Bq/kg[14,15]. This could be due to the enrichment of Ngombas rocks with ^{226}Ra, enrichment of Awanda and Bikoué rocks with ^{232}Th and relative high concentration of ^{40}K in all rock samples collected in the studied areas.

From measured activities of terrestrial radio-nuclides the external hazard index, H_{ex}, was calculated using the following model[17] as criterion. This model uses the external hazard index H_{ex} defined as:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively. This index must be less than unity so that the annual effective dose due to radioactivity in the material will be less or equal to 1.5 mSv/yr [15,18]. The values of H_{ex} obtained in this work are shown in table 2. The total average values of H_{ex} were found to be 8.60 with respective values of 15.30, 6.56, and 3.92 in Ngombas, Awanda and Bikoué. All sampling points had values higher than the recommended limit which may cause harm to people who are living and carrying out agricultural activities in these studied areas.

Absorbed dose and annual effective dose rate: The absorbed dose rate in air D (nGy/h) resulting from the natural specific activity concentration of ^{238}U, ^{232}Th and ^{40}K in Bq/kg, at a height of 1 m above the ground was calculated using the formula proposed by Beck et al[19] and UNSCEAR[20].

$$D = 0.427A_U + 0.662A_{Th} + 0.0432A_K \quad (3)$$

where A_U, A_{Th} and A_K (in Bq/kg) are the specific activities concentrations of ^{238}U, ^{232}Th, ^{40}K respectively[6].The absorbed dose rates in air for the areas under investigation are listed in table 2. The absorbed dose rates of all collected rocks varied from 209.88 to 4136.54 nGy/h with a total mean value of 1427.49 nGy/h which is much higher than the world average value of 55 nGy/h[20]. The means absorbed dose rate values from rock samples in the studied areas are: 2491.24 nGy/h, 1126.90 nGy/h, 664.42 nGy/h respectively in Ngombas, Awanda and Bikoué. All these values are relatively much higher than the world average value. Finally, to calculate the annual external effective dose rate E (mSv/yr), the conversion coefficient from the absorbed dose in air to the effective dose (0.7Sv/Gy)[20] and the outdoor occupancy factor of 20% were used. E was calculated using the following formula:

$$E = D \times O_f \times C_f \quad (4)$$

where D is given by Eq. (3). O_f is the occupancy factor for a year (0.2×365 d×24 h ≃ 1752 h/yr) and C_f is a conversion coefficient. From the table 2, the annual external effective dose rates varied from 0.26 to 5.07mSv/yr with a mean value of 1.75mSv/yr which is about 4 times higher than 0.46mSv/yr[2], the world average due to external exposure of terrestrial radionuclides. The annual external effective dose rate values in the studied areas are: 3.06 mSv/yr, 1.38 mSv/yr, 0.81 mSv/yr respectively in Ngombas, Awanda, Bikoué. The value of

the annual external effective dose rate from Ngombas and Awanda is respectively about 7 and 3 times higher than 0.46 mSv/yr[2]. From Bikoué, the annual external effective dose rate is still below of the regulatory public dose exposure which is 1 mSv/yr [21].

Conclusion

The natural radioactivity and related radiation hazards in rocks from Southern Cameroon were assessed by gamma-ray spectrometry.

1. The concentration values for ^{226}Ra, ^{232}Th and ^{40}K in rocks have been found to lie within the range from 61±4 to 7256 ±364 Bq/kg, 90±6 to 5498±390 Bq/kg and 542±37 to 2505±191 Bq/kg respectively.

2. The highest ^{226}Ra activity was found in rocks from Ngombas area, while the highest ^{232}Th activity was found in rocks from Awanda area. ^{40}K contributes significantly to increase the radioactivity in rocks from the three locations studied in this work.

3. The radium equivalent activities ranging from 427.18 to 9462.36 Bq/kg are higher than the maximum admissible value 370 Bq/kg set in the UNSCEAR report.

4. The mean value of adsorbed dose rate in air was found to be 1427.52 Gy/h and the corresponding annual external effective dose rate was found to be 1.75mSv/yr which is about 4 times greater than the world average value 0.46mSv/yr[2] due to external exposure of terrestrial radionuclides.

In regard to the above results, localities of Ngombas, Awanda, Bikoué in the South Western of Cameroon can be considered as high natural background radiation areas (HBRA). The use of syenites from Ngombas, Awanda, Bikoué in buildings would therefore not be recommended since the average value of external hazard index is higher than unity. It would be recommended to evaluate the total natural radiation exposures of populations living in these high natural background radiation studied areas.

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References

1. Bruzzi L., Baroni M., Mele R.and Nanni E., Proposal for a method of certification of natural radioactivity in building materials, *J. Radiol. Prot.* **(17),** 85 **(1997)**

2. UNSCEAR, Sources and effects of ionizing radiation; Report to General Assembly with Scientific Annexes, United Nations Scientific Committee on the Effects of Atomic Radiation **(1993)**

3. Joshua E.O. et al, Natural radionuclides and hazards of rock samples collected from Southeastern Nigeria, *Radiat. Meas.*, **44, 1 (2009)**

4. Florou H. and Kritidis P., Gamma radiation measurements and dose rate in the coastal areas of a Volcanic Island, Aegean Sea, Greece, *Radiat. Prot. Dosimetry*, **(45),** 277 **(1992)**

5. Anjos R.M. et al, Natural radionuclide distribution in Brazilian commercial granites, *Radiat. Meas.* **(39),** 245 **(2005)**

6. Arafa W., Specific activity and hazard of granite samples collected from the Easterndesert of Egypt, *J. Environ. Radioact.* **(75),** 315 **(2004)**

7. Abd El-Naby H.H. and Saleh G.M., Radioelement distribution in the Proterozoic granites and associated pegmatites of Gabal El Fereyid area, Southeastern Desert, Egypt, *Appl. Radiat. Isot.* **(59),** 289 **(2003)**

8. Maurizot P. et al, Etude et prospection minière du Sud-Ouest Cameroun ; Synthèse des travaux du BGRM, de 1978à **(1986)**

9. Foucault A. and Raoult J.F., Dictionnaire de géologie, 4th edition, Masson **(1997)**

10. Mollah A.S., Ahmed G.U., Husain, S.R. and Rahman M.M., The natural radioactivity of some building materials used in Bangladesh, *Health Physics*, **50 (6)**, 849 **(1986)**

11. Farai I.P. and Sanni O.A., Year long variability of radon in ground water system in Nigeria, *J. Africa Sci.*, **15 (1992)**

12. Ngachin M. et al, Assessment of natural radioactivity and associated radiation hazards in some Cameroonian building materials, *Radiat. Meas.*, **(42),** 61 **(2007)**

13. El-Shershaby A., Study of radioactivity levels in granite of Gable Gattar II in the north eastern desert of Egypt, *Appl. Radiat. Isot.*, **(57),** 131 **(2002)**

14. NEA-OECD, Nuclear Energy Agency. Exposure to radiation from the natural radioactivity in building materials, Report by NEA group of Experts, OECD, Paris **(1979)**

15. Beretka J. and Mathew P.J., Natural radioactivity of Australian building materials, industrial wastes and by-products, *Health Phys*., **(48),** 87 **(1985)**

16. Khan K. et al, Radiometric Analysis of Hazara Phosphate Rock and Fertilizers in Pakistan, *J. Environ. Radioact*., **(38),** 77 **(1997)**

Table 1
Specific activities of Ra, Th, and K in Bq/kg in rock samples collected from South West of Cameroon

Location	Sample I.D.	Specific activity (Bq/kg)		
		^{226}Ra	^{232}Th	^{40}K
Ngombas	NGO-08-R1	2719±190	886±64	2505±192
	NGO-08-R2	704±50	1287±89	2358±165
	NGO-08-R3	3772±189	633±43	542±37
	NGO-08-R4	5680±397	989±69	1765±13
	NGO-08-R5	7256±364	1423±99	2227±156
Awanda	AWA-08-R1	128±10	90±6	2214±160
	AWA-08-R2	836±58	605±44	1642±116
	AWA-08-R3	481±35	5498±390	1561±110
	AWA-08-R4	61±4	409±28	1470±100
	AWA-08-R5	81± 6	323±22	1736± 124
Bikoué	BIK-08-R1	70±6	1128±84	859±63
	BIK-08-R2	570±40	1182±85	1340±96
	BIK-08-R3	102±8	261±18	747±53
	BIK-08-R4	784±55	272±20	799±56
	BIK-08-R5	485±34	571±40	962± 67
	Mean values	1582±158	1037±116	1515±113

Table 2
Radium equivalent activity Ra_{eq}, external hazard index H_{ex}, absorbed dose rate D and annual effective dose E for the rock samples

Location	Sample I.D.	Ra_{eq} (Bq/kg)	H_{ex}	D ($nGyh^{-1}$)	E ($mSvyr^{-1}$)
Ngombas	NGO-08-R1	4178.86	11.29	1855.76	2.28
	NGO-08-R2	2725.98	7.36	1254.47	1.54
	NGO-08-R3	4718.92	12.75	2053.10	2.52
	NGO-08-R4	7230.17	19.54	3156.33	3.87
	NGO-08-R5	9462.36	25.57	4136.54	5.07
Awanda	AWA-08-R1	427.18	1.15	209.88	0.26
	AWA-08-R2	1827.58	4.94	828.41	1.02
	AWA-08-R3	8463.34	22.85	3912.50	4.80
	AWA-08-R4	759.06	2.05	360.31	0.44
	AWA-08-R5	676.56	1.83	323.41	0.39
Bikoue	BIK-08-R1	1749.18	4.72	813.73	0.99
	BIK-08-R2	2363.44	6.38	1083.76	1.33
	BIK-08-R3	532.75	1.44	248.61	0.30
	BIK-08-R4	1234.48	3.34	549.35	0.67
	BIK-08-R5	1375.60	3.72	626.66	0.77
	Mean values	3181.70	8.60	1427.52	1.75

17. Hayambu P. et al, Natural radioactivity in Zambian building materials collected from Lusaka, *J. Radioanal. Nucl. Chem.*, **199 (3)**, 229 **(1995)**
18. Xinwei L. et al, Specific activity and hazards of Archeozoic–Cambrian rock samples collected from the Weibei area of Shaanxi, China, *Radiat. Prot. Dosimetry,* **(118),** 352 **(2006)**

19. Beck, H.L., Decompo, J. and Gologak, J., In Situ Ge (Li) and NaI (Tl) gamma ray spectrometry. Health and Safety Laboratory AEC, Report HASL New York, 258 **(1972)**

20. UNSCEAR, Sources, effects and risks of ionizing radiation; United Nations Scientific Committee on the effects of atomic radiation; Report to the General Assembly on the Effects of Atomic Radiation, United Nations, New York **(1988)**

21. ICRP 60, International Commission on Radiological Protection, Recommendations of the International Commission on Radiological Protection, Pergamon Press, England **(1991)**

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