

Study of natural radioactivity to Assess of radiation hazards from soil samples collected from Mounana in south-east of Gabon

S.Y.L Mouandza^{1,2,3*}, A.B. Moubissi¹, P.E. Abiama^{2,3,4}, T.B. Ekogo¹,
G.H. Ben-Bolie^{2,3}

¹Departement de Physique, Faculté des Sciences, Université des Sciences et Techniques de Masuku

²Laboratory of Nuclear Physics, Department of Physics, Faculty of Science, University of Yaounde I
P.O. Box 812, Yaounde, Cameroon

³Centre d'Excellence Africain en Technologies de l'Information et de la Communication, Université de Yaoundé I
P.O. Box 812, Yaoundé, Cameroun

⁴Nuclear Technology Section, Energy Research Laboratory, Institute of Geological and
Mining Research, P.O. Box 4110 Yaoundé, Cameroon

ABSTRACT

► Original article

*Corresponding authors:

Dr. S.Y. Loemba Mouandza,

E-mail:

yannicksky43@gmail.com

Revised: January 2018

Accepted: April 2018

Int. J. Radiat. Res., October 2018;
16(4): 443-453

DOI: 10.18869/acadpub.ijrr.16.4.443

Background: The present work aims to determine the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples collected in Mounana in the Southeastern region of Gabon, to evaluate the contribution of the natural. **Materials and Methods:** An HPGe p-type detector coupled with a multichannel analyzer DSA-1000 was used to perform measurements and process data. Data acquisition and analysis were made possible using Genie 2000 software version 2.1 and detector was calibrated using the IAEA certified soil reference standard material multi-gamma (No71863/3). **Results:** The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K varied respectively from $12 \pm 03 \text{ Bq kg}^{-1}$ to $23022 \pm 2572 \text{ Bq kg}^{-1}$, $12 \pm 04 \text{ Bq kg}^{-1}$ to $265 \pm \text{Bq kg}^{-1}$ and $64 \pm 13 \text{ Bq kg}^{-1}$ to $811 \pm 74 \text{ Bq kg}^{-1}$ with means values of $2811 \pm 198 \text{ Bq kg}^{-1}$, $63 \pm 14 \text{ Bq kg}^{-1}$ and of $355 \pm 93 \text{ Bq kg}^{-1}$ wet weight. In order to assess the radiological risks due to primordial radionuclides in Mounana, the absorbed dose rate, effective dose, radium equivalent and external hazard index were estimated. **Conclusion:** Absorbed dose rates which have been calculated for each site varied from 26.12 nGy h^{-1} to $10817.99 \text{ nGy h}^{-1}$ with a mean value of $1352.76 \text{ nGy h}^{-1}$. The mean value of annual effective dose was estimated to be 10.96 mSv y^{-1} from the studied area. The areas investigated in this study showed a strong radioactive contamination of sites inhabited by populations.

Keywords: Activity concentration, ^{226}Ra , ^{232}Th , ^{40}K , HPGe, high background radiation, Mounana, Gabon.

INTRODUCTION

The region of Mounana in the south east of Gabon is a uranium-rich area. It was during many years (1961-1999) the center of industrial uranium exploitation (uranium mining); with its famous quarry of Oklo (natural reactor of fission). This mining expanded in several areas including Mounana quarry from 1961 to 1975

with a total quantity of about 5759 tons U (0.49%); Okelobondo quarry between 1965 and 1971 with a total quantity of about 3500 tons U (0.33%); Oklo quarry from 1968 to 1993 and from 1993 to 1999 with a quantity of approximately 15342 tons U (0.45%, 0.32%) and Boyindzi quarry between 1980 and 1991 with 2691 tons or so U (0.31%) as shown in figure 1 and this mining ended definitively in

June 1999^(1,2). The primordial radionuclides that are uranium 238, thorium 232, potassium 40 and other related ones like (^{226}Ra etc.) are always present in soils, rocks and in many building materials⁽³⁻⁸⁾. After the exploitation, the sites of extractions and processing were dismantled and restored. The restoration program was managed exclusively by the Gabonese government, but financed by the European Union. This program had two main phases including: (1) the decommissioning mining bodies between August 2003 and January 2004 and (2) the environmental remediation and restoration of contaminated sites from June to October 2004⁽⁹⁾.

However, scientific reports from the work done by the laboratories of the French NGO named "Commission de Recherche d'Information Indépendantes sur la Radioactivité" (CRIIRAD), approved by the French nuclear safety authority, has shown that radioactive wastes are still present in the areas accessible to natives. In the forest, the soil is contaminated by the extraction of the uranium tailings having a significant radioactivity

concentration⁽²⁾. It should be noted that, there is not enough scientific published data on the radiological impact of uranium mining or the radioactivity concentration in this study area.

It is significant to measure the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in building materials in various places for assessing the radiological dangers for residents⁽¹⁰⁾.

This paper deals with the measurement of decay products of uranium (^{226}Ra), thorium (^{232}Th) series and primordial radionuclide ^{40}K in the soil samples collected in the Mounana region in the south east of Gabon. In this work, radiological parameters that include absorbed dose rate, annual effective dose, radium equivalent and *external hazard index* have been estimated from the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K . The aim of this work is to assess the contribution of the natural radioactivity and radiation hazards received by the populations living in the area of Mounana. The obtained results were compared with the recommended values and the similar studies carried out in other areas.

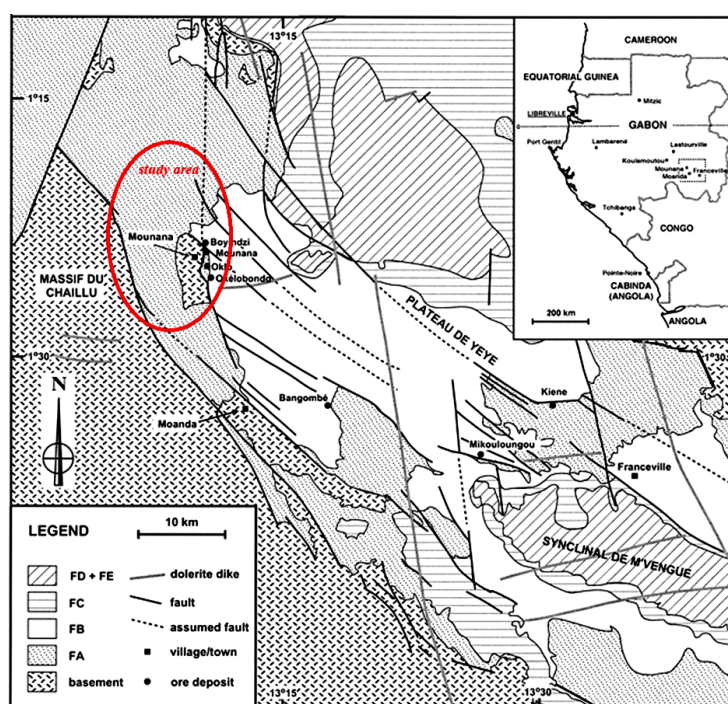


Figure 1. Geology of Franceville basin and the study area (Mounana).

MATERIALS AND METHODS

Study area

The study area is the region of Mounana in Gabon which is located within 1°25' S, and 13° 06'E in the Haut-Ogooue province, with about 11,443 inhabitants ⁽³⁾. The geology of Mounana is based on the bedrock of the Franceville Precambrian as shown in figure 1. The sedimentary series consists of two successive sandstone-pelitic cycles from 1000 to 1500 m thick and fills a basin characterized by a regular subsidence. The uraniferous mineralization is contained in the upper part of the basic sandstones. The origin of uranium deposits is the result of the leaching of fertile rocks (basement or volcanic layers) and to the percolation of these solutions into tectonic-sedimentary traps ⁽¹¹⁾. The average altitude of Mounana is about 459 m. It has a savannah climate with a dry winter according to the classification of Köppen-Geiger. The variation of average rainfalls per month ranges from 20 mm to 171 mm with an average humidity of 83%. The average temperature ranges from 22°C in July to 25°C in March ⁽¹²⁾.

Sampling and preparation

Fifteen samples of soil surfaces (about 0-5 cm) were collected in Mounana according to the two sampling collection methods (systematic and probabilistic). The probabilistic method is made randomly without prior knowledge of the studied area while the systematic method is based on a priori knowledge of the spatial distribution of radioactivity or answering a specific research objective. At each sampling location, an area of about 1.0 m² was marked out. Soil samples were dug up to a depth of about 5 cm with an auger at each corner of the identified 1.0 m² area within its center. The five soil subsamples obtained were then mixed thoroughly to make a composite sample and extraneous materials such as plant roots were removed from the mixture. Each sample was marked using a global positioning system (GPS) as shown in table 1. The sample of about 500 g was then packed in a plastic bag ⁽¹³⁾.

In the laboratory, all samples were dried in a

drying oven at 105°C for 24 h. Each soil sample was ground into thin powder. Finally all samples were sieved using a 2-millimeter mesh to obtain a homogenous sample for measurement ⁽¹⁴⁾. Each sieved sample was sealed in a 50-milliliter cylindrical plastic container (SG 50, standardized by the CEA). The sealed samples were stored for about 30 days before carrying out gamma analysis to allow ²²⁶Ra and its short-lived progenies to reach equilibrium.

Radioactivity measurements

The activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in the soil samples were determined using a p-type high-purity germanium HPGe detector (Canberra) shielded with a 5-centimeter thick lead. It was coupled with a multichannel analyzer of the Digital Spectrum Analyzer (DSA-1000). The energy resolution of the 1332 keV line from ⁶⁰Co was found to be 1.8 keV at full width of half maximum (FWHM), with a relative efficiency of 30%. The data acquisition and the analysis were made possible by using Genie 2000 software version 2.1. The detector was calibrated using the IAEA certified soil reference standard material multi-gamma (N°71863/3), with density similar to the pulverized samples to be measured. A documentation of radionuclides containing the energy of the characteristic gamma peak for each analyzed radionuclide and its corresponding emission probabilities was built from the data supplied by the software. A spectral interactive deconvolution proposed by the Genie 2000 software was performed to examine the multiplets lines.

The ²²⁶Ra activity concentration was calculated using the 186.1 keV peak following its own decay, the 609.3 keV (44.8%), 1120.3 keV (14.9%), 1238.0 keV (5.8%) and 1764.0 keV (15.3%) peak from the ²¹⁴Bi decay and the 351.9 keV (35.8%) peak from the ²¹⁴Pb peak. Then the ²³²Th was measured using the peaks at 338.3 keV (11.3%), 911.2 keV (26.6%), 968.3 keV (17%) of ²²⁸Ac, 583.2 keV (85%) (²⁰⁸Tl). Activity concentration of ⁴⁰K was estimated from its gamma peak 1460.0 keV (10.67%). The gamma spectrum of each sample was measured for 43200 s. The activity concentrations in each sample were evaluated following Equation 1.

$$A_i(^AX) = \frac{S_i}{\varepsilon \times t \times m_i \times P_\gamma} \quad (1)$$

where, $A_i(^AX)$ is the activity concentration of the radionuclide AX (Bq kg⁻¹) in the sample, S_i the count rate obtained under the corresponding peak, m_i is the sample mass (kg), ε is the detection efficiency at a specific energy, P_γ is the emission probability (%) and, t is the active time of acquisition of the peak.

Absorbed dose rate in air

The absorbed gamma dose rate in air at 1.0 m (average gonadal height) above the surface (D in nGy h⁻¹) for the uniform distribution of radionuclides in the soil was calculated following Eq.2 (14).

$$D = 0.462 \times A_{Ra} + 0.621 \times A_{Th} + 0.0417 \times A_K \quad (2)$$

Where A_{Ra} , A_{Th} and A_K are activity concentrations in-situ respectively of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹.

Annual effective dose

The annual effective dose was found from values of annual outdoor and indoor effective doses. The annual outdoor effective dose equivalent was calculated for each sampling point using the conversion factor of 0.7 Sv/Gy. It translates the absorbed dose rate in the air into effective dose and that includes an outdoor occupancy factor of 0.2 as showed by Eq.3(15).

$$E_{out}(mSv) = D_{out}(nGy \text{ h}^{-1}) \times 8760 \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (3)$$

The annual indoor effective dose equivalent was calculated for each sample using the conversion factor of 0.7 Sv/Gy. It translates the absorbed dose rate (nGy h⁻¹) in air into effective dose (mSv y⁻¹) and that includes an indoor occupancy factor of 0.8 as showed by Eq.4 (15).

$$E_{in}(mSv) = D_{in}(nGy \text{ h}^{-1}) \times 8760 \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (4)$$

Where D_{out} is the outdoor dose rate in air, D_{in} the indoor dose rate where the ratio of indoor to outdoor exposure rates is 1.4 (16).

Radium equivalent

In the calculation of absorbed dose rates in the air, a uniform distribution of natural radionuclides in the soils was made. In reality, the distribution is not uniform. Therefore, a common radiological index that can be used to assess the level of real activity of ²²⁶Ra, ²³²Th and ⁴⁰K in the soil samples is required. This common radiological index, called the radium equivalent activity Ra_{eq} (17), provides a useful guide in the regulation of radiation protection safety standards for human population. This index is based on the estimation that 370 Bq kg⁻¹ of ²²⁶Ra, 259 Bq kg⁻¹ of ²³²Th and 4180 Bq kg⁻¹ of ⁴⁰K produce the same gamma-ray dose rate and can be expressed as:

$$Ra_{eq} = A_{Ra} + \frac{10}{7} A_{Th} + \frac{10}{130} A_K \quad (5)$$

where, A_{Ra} , A_{Th} and A_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹ respectively.

External hazard index

The recommendations of the upper limit of the radiation dose from building materials is 1.5 mSv y⁻¹ (18). Security requirements are issued by different countries to limit the radiation exposure caused by the use of raw construction materials containing high concentrations of natural radionuclides. Limiting the radiation dose to that value, is designed on the conservative model based on infinitely thick walls without windows and doors to serve as criteria for calculating the external hazards index, defined according to Krieger (19) in the following Equation 6.

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

where, A_{Ra} , A_{Th} and A_K are respectively activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹.

Hewamanna (20) corrected this model after considering a finite thickness of walls and the existence of windows and doors. Taking these considerations into account, the equation used for the calculation of external hazard index becomes.

$$H_{ex} = \frac{A_{Ra}}{740} + \frac{A_{Th}}{520} + \frac{A_K}{9620} \leq 1 \quad (7)$$

In addition to the external irradiation, radon and its related short-lived products are also hazardous to the respiratory organs. The internal hazard index (H_{in}) is used to control the

internal exposure to ^{222}Rn and its radioactive progeny ⁽²¹⁾. Taking these considerations into account, the equation used for the calculation of internal hazard index becomes:

$$H_{in} = \frac{A_{Ra}}{285} + \frac{A_{Th}}{520} + \frac{A_K}{4810} \leq 1$$

Table 1. samplings points' location and soil samples activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , per unit dry weight.

SampleID.	Latitude (S)	Longitude (E)	Altitude (m)	^{226}Ra (Bq kg ⁻¹)	^{232}Th (Bq kg ⁻¹)	^{40}K (Bq kg ⁻¹)	$1/^{226}\text{Ra}$ (kg ⁻¹ Bq)
Moun1	01°20'54''	13°07'37''	334±09	17±04	44±08	368±40	6.10^{-2}
Moun2	01°22'37''	13°09'19''	433±09	44±08	37±09	317±80	2.10^{-2}
Moun3	01°23'31''	13°09'52''	420±08	22±05	12±03	204±66	4.10^{-2}
Moun4	01°24'60''	13°09'37''	415±09	23022±2572	265±49	414±148	4.10^{-5}
Moun5	01°25'46''	13°10'13''	401±10	12±02	35±06	223±50	8.10^{-2}
Moun6	01°25'08''	13°09'38''	382±10	142±15	56±19	169±45	7.10^{-3}
Moun7	01°23'41''	13°09'35''	423±08	2660±249	50±09	552±64	4.10^{-4}
Moun8	01°23'48''	13°09'42''	411±07	3572±326	145±23	598±90	3.10^{-4}
Moun9	01°24'29''	13°09'15''	426±12	59±10	37±17	64±13	2.10^{-2}
Moun10	01°25'24''	13°09'57''	380±09	158±21	50±05	153±40	6.10^{-3}
Moun11	01°24'16''	13°09'47''	530±12	30±04	64±11	254±67	3.10^{-3}
Moun12	01°21'42''	13°07'50''	405±09	49±09	32±05	520±90	2.10^{-2}
Moun13	01°24'43''	13°09'28''	432±12	39±09	17±03	669±92	3.10^{-2}
Moun14	01°25'35''	13°09'50''	415±09	3659±341	56±10	811±74	3.10^{-4}
Moun15	01°23'56''	13°09'42''	425±10	8683±786	41±02	14±02	10^{-4}
Mean			416±09	2811±198	63±12	355±93	3.10^{-4}
Range			334-530	12-23022	12-265	64-811	4.10^{-5} - 8.10^{-2}

RESULTS AND DISCUSSION

Activity concentration

From these measurements, we can see that the activity concentrations of radium, thorium and potassium are not uniformly distributed in the studied soils. The results of activity concentrations measurements in the soil on a dry weight with their counting errors are reported in table 1. The activity concentrations of ^{226}Ra vary from $12 \pm 02 \text{ Bqkg}^{-1}$ to $23022 \pm 2572 \text{ Bqkg}^{-1}$ with a mean value of $2811 \pm 198 \text{ Bqkg}^{-1}$. The table 1 also showed that more than about 74% of soil samples have activity concentrations of ^{226}Ra higher than the world average value of 32 Bqkg^{-1} ⁽¹⁵⁾. The mean activity concentration is strongly influenced by the concentration of the Moun4 sample. Moun4 has an activity concentration of $23022 \pm 2572 \text{ Bqkg}^{-1}$ in ^{226}Ra , that is to say, more than 719 times the world average value. This high level of

radioactivity of ^{226}Ra could reflect the fact that this sample was collected on the Oklo rehabilitated site. Oklo is the uranium quarry where the sixteen (16) natural nuclear fission reactors were discovered. Since the fissions would not only be produced only with the ^{235}U ; but also with the ^{238}U by fast neutrons and slow neutron fission with plutonium formed in situ by neutron capture in the ^{238}U ⁽²²⁻²⁴⁾. These different nuclear reactions could be the cause of this high activity concentration of ^{226}Ra (descending from the ^{238}U) in this zone. Moun7, Moun8, Moun14 and Moun15, whose activity concentrations are ranged between 83 and 271 times the world average value, all these are from rehabilitated sites, except Moun8, which comes from the sawmill factory in Mounana. The Moun7 and Moun14 samples are from the sites where uranium radioactive wastes were buried. Maybe the containment of these wastes is no longer effective and would therefore be responsible for

the increase the activity concentration in these two sites. The high activity concentration of ^{226}Ra in the Moun15 sample would be explained by the fact that it originated from the restored site of the former processing plant. After the dismantling of the plant and the restoration of the site, some buried materials could have surfaced due to heavy rainfalls in the area; which would significantly increase the activity concentration of ^{226}Ra . Whereas the Moun8 sample comes from the bottom of the runoff water flowing from former uranium galleries; Which could explain the high concentration of ^{226}Ra in this sample Moun8. The Moun1 and Moun5 samples have the lowest activity concentrations of $17 \pm 04 \text{ Bq kg}^{-1}$ and $12 \pm 02 \text{ Bq kg}^{-1}$, respectively. These low activity concentrations could be explained by the fact that Moun1 and Moun5 were collected respectively at the exit and at the entrance of the town of Mounana far from the old deposits and houses.

The increase in activity concentration in the other five (5) samples collected around the dwellings would be due to the fact that the soil of Mounana is framed by the four (4) uranium mines and that ^{226}Ra would have been transported preferentially to uranium, because of its absorption on suspended matter ⁽²⁵⁾; what would increase the activity concentration of ^{226}Ra in the environment.

The use of the opposite of the activity concentration of ^{226}Ra could provide us with an explanation. Indeed radionuclides-rich soils present values of $1/^{226}\text{Ra}$ varying only one unit logarithmic (0.01 to 0.1 kg Bq^{-1}) contrary to the soil impacted by mining activity, for which variation ranges between 10^{-6} and $10^{-1} \text{ kg Bq}^{-1}$ ⁽²⁶⁾, we could say that the soil of Mounana has been contaminated by the uranium mining. The high activity concentrations of radium tend to be observed in the soils developed on storages of wastes, the sites of treatment and the sites of extractions. The high activity concentrations of ^{226}Ra observed in soil samples could be due uranium mining.

The activity concentrations of ^{232}Th vary from $12 \pm 03 \text{ Bqkg}^{-1}$ to $265 \pm 49 \text{ Bqkg}^{-1}$ with a mean value of $63 \pm 12 \text{ Bqkg}^{-1}$. This value is

relatively higher than the world average value of 45 Bqkg^{-1} ⁽¹⁵⁾. While the value of Moun4 and Moun8 are respectively about six (6) times and three (3) times higher than the world average value. The relative high concentrations of ^{232}Th analyzed in some soil samples indicated that thorium also contributed to the contamination of the environment. The isotopes of thorium (^{232}Th) tend to be less mobile than those of uranium ^(25, 27). Activity concentration of ^{232}Th is relatively high, but remains negligible compared with the contribution of ^{226}Ra .

^{40}K activity concentrations vary from $64 \pm 13 \text{ Bqkg}^{-1}$ to $811 \pm 74 \text{ Bqkg}^{-1}$ with a mean value of $355 \pm 93 \text{ Bqkg}^{-1}$. Only five soil samples representing about 33% of the samples showed ^{40}K activity concentrations higher than the world average value of 420 Bqkg^{-1} ⁽¹⁵⁾. ^{40}K contributes slightly to the total activity in soil samples from the study area. These various concentrations are also due to factors such as the presence and the nature of the organic matter in the Mounana soil, the relative mobility of radionuclides, the seasonal fluctuations of the level of the sheet (controlling the conditions of oxydo reduction) or the vegetation that contribute to the retention of the radionuclides in the studied zones (wet) ⁽²⁸⁻³²⁾.

Figures 2, 3 and 4 show the dependence between activity concentrations of radionuclides. The low correlations between ^{226}Ra and ^{40}K ($R^2 = 0.0048$) and between

^{232}Th and ^{40}K ($R^2 = 0.0343$) in figure 2 and figure 3 show that activity concentration of ^{40}K could not be linked to the presence of ^{226}Ra and of ^{232}Th in soil samples in the study area. However, the clear correlation between the reports $^{226}\text{Ra} / ^{40}\text{K}$ and $^{232}\text{Th} / ^{40}\text{K}$ in the figure 4 ($R^2 = 0.9543$) shows that the radioactivity in the samples of collected soil would be induced by the presence of radioactive heavy mineral enriched in ^{232}Th and ^{226}Ra . This correlation may suggest that the thorium mineralization and uranium were made during the same geological period in the study area.

Table 2 shows a comparison of the average activity concentrations values obtained in this study with values from other areas in the world. In this table the average activity concentration

values of ^{226}Ra obtained in the present study are fourteen (14) times higher than those obtained in the uranium mines in Portugal. ^{232}Th values obtained are relatively high compared to reference value from UNSCEAR but are still comparable to the values obtained in other

countries with the exception of Cameroon, where measurements were made in Thorium-rich a zone. The average value of ^{40}K is still below the average value proposed by UNSCEAR.

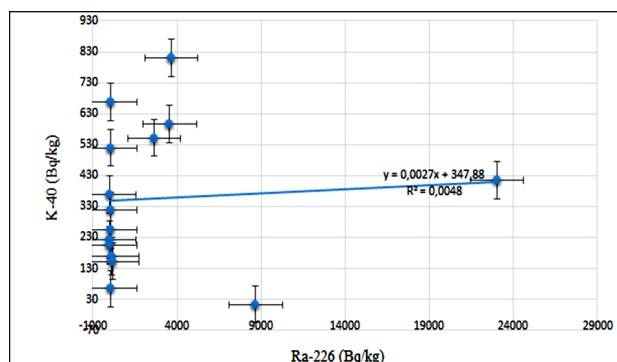


Figure 2. Correlations between ^{226}Ra and ^{40}K .

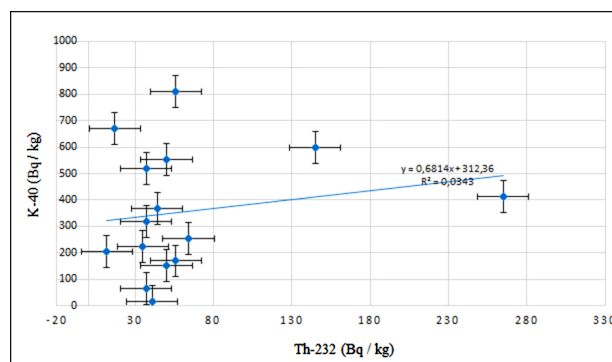


Figure 3. Correlations between ^{232}Th and ^{40}K .

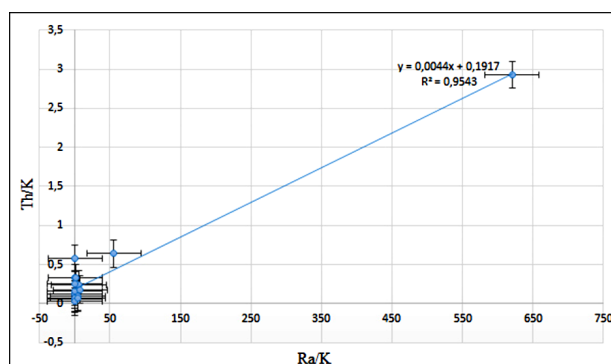


Figure 4. Correlations between $^{226}\text{Ra}/^{40}\text{K}$ and $^{232}\text{Th}/^{40}\text{K}$.

Table 2. Comparison of the activity concentrations of terrestrial radionuclides with other published values in the world.

Sample ID	D(nGy h^{-1})	E_{out} (mSv y^{-1})	E_{in} (mSv y^{-1})	Ra_{eq} (Bq kg^{-1})	H_{in}	H_{ex}
Moun1	50.52	0.062	0.35	108.26	0.34	0.29
Moun2	56.52	0.069	0.39	121.32	0.45	0.33
Moun3	26.12	0.032	0.18	54.87	0.21	0.15
Moun4	10817.99	13.267	74.30	23432.83	125.55	63.33
Moun5	36.58	0.045	0.25	79.22	0.25	0.21
Moun6	107.43	0.132	0.74	235.10	1.02	0.64
Moun7	1282.99	1.573	8.81	2774.00	14.69	7.50
Moun8	110.43	0.135	0.76	241.28	1.08	0.65
Moun9	52.90	0.065	0.36	116.84	0.48	0.32
Moun10	1765.25	2.165	12.12	3825.40	20.00	10.34
Moun11	64.20	0.079	0.44	141.08	0.46	0.38
Moun12	67.30	0.083	0.46	141.95	0.52	0.38
Moun13	56.47	0.069	0.39	114.83	0.42	0.31
Moun14	1759.05	2.157	12.08	3801.53	20.16	10.27
Moun15	4037.59	4.952	27.73	8742.71	47.10	23.63
Mean	1352.76	1.66	9.30	2928.75	15.51	7.92
Range	26.12-10817	0.032-13.267	0.18-74.30	54.87-23432.8	0.21-125.55	0.15-63.33

Absorbed dose rate in the air

The values of absorbed dose rates D in the air on wet weight basis are listed in table 3. The absorbed dose rates due to the presence of ^{226}Ra , ^{232}Th and ^{40}K , in soil samples in the studied area vary between 26.12 nGy h^{-1} and $10817.99 \text{ nGy h}^{-1}$ with a mean value of $1352.76 \text{ nGy h}^{-1}$, which is much higher than the world average value of 58 nGy h^{-1} ⁽¹⁵⁾. This mean value is more than twenty (20) times higher than the world average value. From these doses rates, the inhabitants of Mounana are exposed to public health problems. The source of the Moun4 (Oklo) sample is situated at about 70 meters from the main road and 150 meters from the houses. Despite warning signposts natives can easily go to these sites. Moun7 and Moun14 were both collected on restored sites and the areas are very frequently visited by the local

population, giving rise to the "Renovation City", the hospital, the sawmill factory and the main road enabling to enter the city. In these different sites, there are also some plantations where crops are grown around. The site from where Moun15 comes is not far from the "Ambié city" and this place is very frequented by the indigeneous population because it is a fi playing ground for young people (football stadium). Sawmill factory workers are also daily exposed to very high doses. Figure 5 displays the average absorbed dose rates of ^{226}Ra , ^{232}Th and ^{40}K from soil samples collected in Mounana. It is showed that radium highly contributes to the external exposure dose rate compared to thorium and potassium in the studied areas, and the radiological risk is present for people living in Mounana comparatively to people living in Ajaokuta North-central of Nigeria ⁽⁶⁾.

Table 3. Calculated radiological indices for the study area.

Location	^{226}Ra (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})	References
India (Bangalore)	26.20	53.10	635.10	Shiva Prasad ⁽²⁸⁾
Brazil (Rio Grande do Norte)	29.20	47.80	704	Malanca ⁽²⁹⁾
Eastern Germany (Ronneburg)	370	45	620	Winkelmann ⁽³⁰⁾
Portugal (Uranium mining)	200	91	-	Carvalho ⁽³¹⁾
Spain	46.0	49.0	650.0	Baeza ⁽³²⁾
UpperEgypt	31-40	52-61	3149-3210	Uosif ⁽³³⁾
Cameroon (Southwestern)	130	700	30	P. Ele ⁽¹³⁾
Nigeria (Southwesternregion)	54.50	91.10	286.50	Oladele Samuel ⁽³⁴⁾
Gabon (Mounana)	2811 ± 198	63 ± 12	355 ± 93	Presentstudy
World average	16–116 (33)	7–50 (45)	100–700 (420)	UNSCEAR 2008 ⁽¹⁶⁾

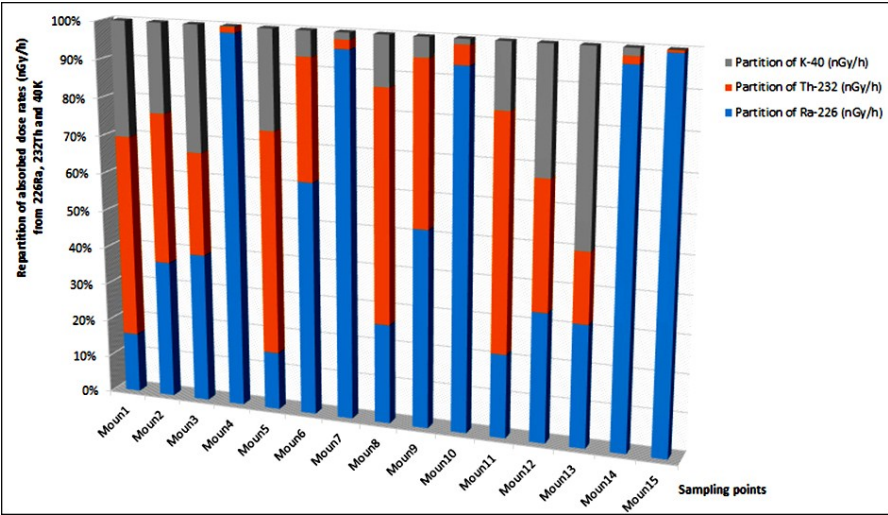


Figure 5. Absorbed dose rate from ^{226}Ra , ^{232}Th and ^{40}K .

Annual effective dose

As showed in the table 3, the annual outdoor effective dose equivalent (E_{out}) ranged between 0.03 mSv y^{-1} and 13.27 mSv y^{-1} , with a mean value of 1.66 mSv y^{-1} , which is much higher than the world average value of 0.07 mSv y^{-1} . The annual indoor effective dose (E_{in}) ranged between 0.18 mSv y^{-1} and 74.30 mSv y^{-1} , with a mean value of 9.30 mSv y^{-1} , which is more than twenty-two (22) times higher than the world average value of 0.41 mSv y^{-1} (4). The annual effective dose equivalent is a mean value of 10.96 mSv y^{-1} which is more than twenty-two (22) times higher than the world average value of 0.48 mSv y^{-1} (4). These effective doses could be due to the high concentrations measured in the samples taken in Mounana; particularly activity concentrations of ^{226}Ra .

Radium equivalent

From table.3, the Ra_{eq} ranged between 54.87 Bq kg^{-1} and $23432.82 \text{ Bq kg}^{-1}$, with a mean of $2928.75 \text{ Bq kg}^{-1}$ which is more than seven (7) times higher than the safety standards value of 370 Bq kg^{-1} (4). This implied that using the soils in the area as building materials might present significant radiological health risks.

External hazard index

From table 3, the values of external hazard index (H_{ex}) vary from 0.15 to 63.33, with a mean value of 7.92. However this value average is strongly impacted by values of samples Moun4 with 63.33, Moun7 with 7.50, Moun10 with 10.34, Moun14 with 10.27 and Moun15 with 23.63. So the sites where these samples were taken with an exposure index greater than 1, should not be used as building materials.

The values for internal hazard index (H_{in}) vary from 0.21 to 125.55, with a mean value of 15.51. Note that the soil samples having a value above the limit represents about 46% of all collected sample. Sampling points having values higher than the recommended limit which may cause harm to the people who live and carry out farming activities in these studied areas.

CONCLUSION

The activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in soil samples from the Mounana region in the south east of Gabon highly varies from one sample to another. The highest level of ^{226}Ra and ^{232}Th are observed in soil sample Moun4. The highest level of ^{40}K was found from soil sample Moun14. The main value of the absorbed dose rate and annual effective dose are estimated respectively at $1352.76 \text{ nGy h}^{-1}$, 10.96 mSv . We could say that Mounana's soil shows a high radioactive contamination despite the implementation of an ecology restoration program. The areas investigated in this study showed a strong radioactive contamination of the inhabited sites. This study could be used as a database for future investigations and this data might be useful for the natural radioactivity mapping. We recommend then, an epidemiological study in this area on radioactivity- related diseases, the birth and mortality rate in this area and even the geographical distribution of the population according to the distribution of the activity concentration.

ACKNOWLEDGMENTS

The authors express their gratitude to the Technical Cooperation of the International Atomic Energy Agency (IAEA) in Vienna for providing financial assistance via the AFRA regional project RAF/9/048 and the "Centre National de l'Energie des Sciences et des Techniques Nucléaires (CNESTEN)" in Morocco for providing technical assistance, that made this study possible. The first author is grateful to Miss Lily Esther Ndouna Despenaud NLO Gabon for her assistance.

Conflicts of interest: Declared none.

REFERENCES

1. NEA/IAEA Nuclear Energy Agency and International Atomic Energy Agency (2002) A Joint Report by the OECD; *Environmental Remediation of Uranium Production Facilities*, **2**: 217-220.
2. Commission de Recherche et d'Information Indépendantes sur la Radioactivité CRIIRAD (2009) Contamination radiologique relevée sur l'ancien site minier uranifère de COMUF-AREVA à Mounana au GABON. Note N°09-118. www.criirad.org/.
3. Manjunatha S, Jayasheelan A, Venkataranabauag P (2013) Study of distribution of ^{226}Ra , ^{232}Th and ^{40}K in different rock formations and their dose estimation in and around Chickmagalur, India. *Int J Radiat Res*, **11**: 183-187.
4. UNSCEAR-United Nations Scientific Committee on the Effects of Atomic Radiation (2000) Sources and effects of Ionizing radiation. Report to the General Assembly, with Scientific Annexes. New York: United Nations Publications.
5. Ravisankar R, Vanasundari K, Chandrasekaran A, Rajalakshmi A, Suganya M, Vijayagopal P, Meenakshisundaram V (2012) Measurement of natural radioactivity in building materials of Namakkal, Tamil Nadu, India using gamma-ray spectrometry. *Appl Radiat Isot*, **70**: 699-704.
6. Usikalu MR, Rabiou AB, Oyeyemi KD, Achuka JA, Maza M (2017) Radiation hazard in soil from Ajaokuta North-central Nigeria. *Int J Radiat Res*, **15(2)**: 219-224.
7. Njinga RL and Tshivhase VM (2017) Radio-epidemiological evaluation and remediation in water sources from two mines in South Africa. *Int J Radiat Res*, **15(3)**: 307-315.
8. Mvondo S, Ben-Bolie GH, Ema'aEma'a, Owono Ateba P, Ele Abiama P (2017) study of soil-fern transfert of naturally occurring alpha emitting radionuclides in the southern region of Cameroon. *Journal of Environmental Radioactivity*, **180**: 114-119.
9. Daoud S, Getti JP, Bourdon W (2007) Rapport d'enquête sur la situation des travailleurs de la COMUF, filiale gabonaise du groupe AREVA-COGEMA. www.asso-sherpa.org/uranium-afriq.
10. Yin N, Lu X, Li Y (2017) Radioactive analysis and radiological hazards of sand in Weifang, China. *Int Radiat Res*, **15(2)**: 225-228.
11. Bourrel J and Pfiffelmann JP (1972) La province uranifère du bassin de Franceville (République Gabonaise). *Mineral Deposita Bert*, **7**: 323-336.
12. Peel MC, Finlayson BL, Mac TA, McMahon TA (2007) World of the Köppen-Geiger climate classification. *Hydrology and Earth System Sciences*, **11**: 1633.
13. EleAbiama P, OwonoAteba P, Ben-Bolie GH, Ekobena FHP, El Khoukhi T (2010) High background radiation investigated by gamma spectrometry of the soil in the Southwestern region of Cameroon. *Journal of Environmental Radioactivity*, **101**: 739-743.
14. International Atomic Energy Agency (1989) Measurement of radionuclides in food and the environment. IAEA technical report series no. 295, Vienna.
15. UNSCEAR (2008) United Nations Scientific Committee on the Effect of Atomic Radiation report to the general assembly. Annex B: exposures of the public and workers from various sources of radiation.
16. Lidman, Fredrik, Henrik Ramebäck, ÅsaBengtsson, and HjalmarLaudon (2013) Distribution and Transport of Radionuclides in a Boreal Mire – Assessing Past, Present and Future Accumulation of Uranium, Thorium and Radium. *Journal of Environmental Radioactivity*, **12**: 87-97.
17. Beretka J and Mathew PJ (1985) Natural radioactivity of Australian building materials Industrial wastes and by-product. *Health Phys*, **48**:87-95.
18. ICRP, International Commission on Radiological Protection (1977) Recommendations of ICRP. Pergamon Press, Publication 26, Oxford.
19. Krieger VR (1981) Radioactivity of construction materials. *Betonwerk Fertigteil Tech*, **47**:468-473.
20. Hewamanna R, Sumithrachchi CS, Mahawatte P, Nanayakara HLC, Ratnayake HC (2001) Natural radioactivity and gamma dose from Sri Lankan clay bricks used in building construction. *Applied Radiation and Isotopes*, **54**: 365-369.
21. Al-Trabulsi HA, Khater AEM, Habbani FI (2010) Radioactivity levels and radiological hazard indices at the Saudi coastline of the Gulf of Aqaba. *Radiation Physics and Chemistry*, **80**: 343-348.
22. Bodu R (1972) Sur l'existence d'anomalies isotopiques rencontrées dans l'uranium du Gabon. *CR Academie des Sciences Paris*, **275p**:1731.
23. Neuilly M (1972) Sur l'existence dans un passé reculé d'une réaction en chaîne naturelle de fissions dans le gisement d'uranium d'Oklo (Gabon) *ibid*. p.1847.
24. Naudet R (1991) OKLO : Des reacteursnucleaires fossiles. Etude physique. Eyrolles, Paris.
25. Lozano JC, Blanco Rodríguez P, Vera Tomé F,(2002) Distribution of Long-LivedRadionuclides of the 238 U Series in the Sediments of a Small River in a Uranium Mineralized Region of Spain. *Journal of Environmental Radioactivity*, **63(2)**: 153-71.
26. Landström O, Sundblad B (1986) Migration of thorium, uranium, radium and Cs-137 in till soils and their uptake in organic matter and peat. SKB.
27. Blanco P, Vera Tomé F, Lozano JC (2005) Fractionation of natural radionuclides in soils from a uranium mineralized area in the South-West of Spain. *Journal of Environmental Radioactivity*, **79(3)**: 315-30.
28. Alicia Cuvier (2015) Accumulations et sources de l'uranium, de ses descendants et des éléments traces métalliques dans les zones humides autour des anciens sites miniers uranifères. thèse de doctorat de l'université de toulouse. *Institut National Polytechnique de Toulouse (INPT)*, P 58-63.
29. Kipp GG, Stone JJ, Stetler LD (2009) Arsenic and uranium transport in sediments near abandoned uranium mines in Harding County, South Dakota. *Applied Geochemistry*, **24(12)**: 2246-55.
30. O'Dea J and Mark D (1999) Spatial analysis of natural radionuclides in peat overlying a lithological contact in Co. Donegal, Ireland. *Journal of Environmental Radioactivity*, **44(1)**: 107-17.

31. Dowdall M and O'Dea J (2002) Ra-226/U-238 Disequilibrium in an upland organic soil exhibiting elevated natural radioactivity. *Journal of Environmental Radioactivity*, **59**: 91–104.
32. Dowdall M, Gwynn JP, Moran C, Davids c, O'Dea J, Lind B (2005) Organic Soil as a Radionuclide Sink in a High Arctic Environment *Journal of Radioanalytical and Nuclear Chemistry*, **266 (2)**: 217–23.
33. Krachler, Michael, William Shotyk (2004) Natural and Anthropogenic Enrichments of Molybdenum, Thorium, and Uranium in a Complete Peat Bog Profile, Jura Mountains, Switzerland Presented as Part of the Archives of Environmental Contamination at the 6th International Symposium on Environmental Geochemistry, Edinburgh, Scotland, 7/11 September 2003. *Journal of Environmental Monitoring*, **6 (5)**: 418.
34. Shiva Prasad NG, Nagaiah N, Ashok GV, Karanukara N (2008) Concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in the soils of Bangalore Region, India. *Health Phys*, **94(3)**:264–271.
35. Malanca A (1996) Distribution of ²²⁶Ra, ²³²Th, and ⁴⁰K in soils of Rio Grande do Norte (Brazil). *J Environ Radioact*, **30**: 55–67.
36. Winkelmann I, Thomas M, Vogl K (2001) Aerial measurements on uranium ore mining milling and processing areas in Germany. *J Environ Radioact*, **53**:301–311.
37. Carvalho FP, Madruga MJ, Reis MC, Alves JG, Oliveira JM, Gouveia JS (2007) Radioactivity in the environment around past radium and uranium mining sites of Portugal. *J Environ Radioact*, **96**: 39–46.
38. Baeza A, Del-Rio M, Miro C, Paniagua J (1992) Natural radioactivity in soils of the province of Caceres (Spain). *Radiat Protect Dosim*, **45(1/4)**: 261–263.
39. Uosif M (2011) Specific activity of ²²⁶Ra, ²³²Th and ⁴⁰K for assessment of radiation hazards from building materials commonly used in Upper Egypt. *SDU J Sci*, **6(2)**: 120–126.
40. Ajayi SO (2009) Measurement of activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th for assessment of radiation hazards from soils of the southwestern region of Nigeria. *Radiat Environ Biophys*, **48**: 323–332.

