



Assessment of natural radioactivity (^{40}K , ^{238}U and ^{232}Th) and radiological risk in building construction materials: the case of Benin hill granites

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Abstract: Continuous exposure to ionizing radiation can have harmful effects on human health. In this respect, a study was carried out in the Communes of Dassa-zoume and Glazoue to determine the levels of ^{40}K , ^{238}U and ^{232}Th in granites and sand. The specific activities obtained made it possible to assess the possible radiological risks associated with the resident population. During sampling work in November 2022, twelve granite samples were taken from twelve quarries. In addition, a sand sample was collected in Cotonou for comparison. All samples were then sent to the INSTN-Madagascar laboratory for gamma-ray spectrometry analysis using a NaI(Tl) detector. The results show that the average specific activities of ^{40}K , ^{238}U and ^{232}Th in the granites are $(1329 \pm 128) \text{ Bq.kg}^{-1}$, $(44 \pm 11) \text{ Bq.kg}^{-1}$ and $(129 \pm 44) \text{ Bq.kg}^{-1}$ respectively. Those in sand are $(144 \pm 8) \text{ Bq.kg}^{-1}$, $(30 \pm 2) \text{ Bq.kg}^{-1}$ and $(56 \pm 7) \text{ Bq.kg}^{-1}$ respectively. The average equivalent radium activity is $(330 \pm 74) \text{ Bq.kg}^{-1}$ versus $(122 \pm 10) \text{ Bq.kg}^{-1}$ for sand. The mean gamma index for granite was (1.2 ± 0.3) , compared with (0.43 ± 0.03) for sand. The average absorbed dose rate in air is $(153.4 \pm 9.4) \text{ nGy.h}^{-1}$ versus $(53.7 \pm 5.1) \text{ nGy.h}^{-1}$ for sand. For adults, for 80% building occupancy factor, the mean annual indoors and outdoors effective dose (E) inside a building are $(0.75 \pm 0.05) \text{ mSv.y}^{-1}$ and $(0.19 \pm 0.01) \text{ mSv.y}^{-1}$ versus $(0.26 \pm 0.05) \text{ mSv.y}^{-1}$ and $(0.07 \pm 0.01) \text{ mSv.y}^{-1}$ for sand. For 60% factor, the means are $(0.56 \pm 0.03) \text{ mSv.y}^{-1}$ and $(0.38 \pm 0.02) \text{ mSv.y}^{-1}$ versus (0.20 ± 0.01) and $(0.13 \pm 0.01) \text{ mSv.y}^{-1}$ for sand. For the children, for a building occupancy factor of 80%, the average E are $(0.86 \pm 0.05) \text{ mSv.y}^{-1}$ and $(0.21 \pm 0.01) \text{ mSv.y}^{-1}$ versus $(0.30 \pm 0.01) \text{ mSv.y}^{-1}$ and $(0.08 \pm 0.01) \text{ mSv.y}^{-1}$ for sand respectively for indoors and outdoors. For infants, the E are $(1.00 \pm 0.06) \text{ mSv.y}^{-1}$ and $(0.25 \pm 0.01) \text{ mSv.y}^{-1}$ versus $(0.35 \pm 0.01) \text{ mSv.y}^{-1}$ and $(0.09 \pm 0.01) \text{ mSv.y}^{-1}$ for sand respectively for indoors and outdoors. For children, for a building occupancy factor of 60%, the average E are $(0.64 \pm 0.03) \text{ mSv.y}^{-1}$ and $(0.43 \pm 0.02) \text{ mSv.y}^{-1}$ versus $(0.23 \pm 0.01) \text{ mSv.y}^{-1}$ and $(0.15 \pm 0.01) \text{ mSv.y}^{-1}$ for sand respectively indoors and outdoors. For infants, for a building occupancy factor of 60%, the E are $(0.75 \pm 0.05) \text{ mSv.y}^{-1}$ and $(0.50 \pm 0.02) \text{ mSv.y}^{-1}$ versus $(0.26 \pm 0.01) \text{ mSv.y}^{-1}$ and $(0.18 \pm 0.01) \text{ mSv.y}^{-1}$ for sand respectively



indoors and outdoors. For adults, the excess lifetime cancer risk at age 66 for a total annual effective dose induced by granite is $(3.1 \pm 0.01) \cdot 10^{-3}$ versus $(1.1 \pm 0.01) \cdot 10^{-3}$ for sand. For the children and infants, the average ELCR is $(3.5 \pm 0.01)E-3$ and $(4.1 \pm 0.01)E-3$ versus $(1.2 \pm 0.01)E-3$ and $(1.4 \pm 0.01)E-3$ for sand respectively. Statistical analysis of the data was performed with Python 3.11 and R 4.3.2 on the Spyder and studio interface. The p-value is < 0.001 compared with the UNSCEAR reference value for absorbed dose rate in air, effective dose and excess risk, which are $84 \text{ nGy} \cdot \text{h}^{-1}$, $0.48 \text{ mSv} \cdot \text{y}^{-1}$ and $0.29 \cdot 10^{-3}$. This shows that the granites in the quarries studied present a radiological risk when used as building materials and need specific radiation protection measures for his users.

Keywords: radionuclide, effective dose, specific activity, absorbed dose rate, excess lifetime cancer risk.



Évaluation de la radioactivité naturelle (^{40}K , ^{238}U et ^{232}Th) et du risque radiologique dans les matériaux de construction des bâtiments : cas des granites des collines du Bénin

Résumé: L'exposition en permanence aux rayonnements ionisants peut entraîner des effets néfastes à la santé humaine. A cet égard, une étude a été menée dans les Communes de Dassa-zoumé et de Glazoué afin de déterminer les niveaux de la radioactivité ^{40}K , ^{238}U et ^{232}Th dans les granites et le sable. Les concentrations d'activités obtenues ont permis d'évaluer les éventuels risques radiologiques associées à la population qui y réside. Pendant les travaux de prélèvement au mois de novembre 2022, douze échantillons de granites ont été prélevés dans douze carrières. De plus, un échantillon de sable a été collecté à Cotonou pour comparaison. Tous les échantillons ont été ensuite envoyés au laboratoire de l'INSTN-Madagascar aux fins d'analyser par spectrométrie gamma muni d'un détecteur NaI(II). Les résultats obtenus montrent que les activités spécifiques moyennes de ^{40}K , ^{238}U et ^{232}Th dans les granites sont de $(1329 \pm 128) \text{ Bq.kg}^{-1}$, de $(44 \pm 11) \text{ Bq.kg}^{-1}$ et de $(129 \pm 44) \text{ Bq.kg}^{-1}$ respectivement. Celle du sable est de $(144 \pm 8) \text{ Bq.kg}^{-1}$, de $(30 \pm 2) \text{ Bq.kg}^{-1}$ et de $(56 \pm 7) \text{ Bq.kg}^{-1}$ respectivement. L'activité moyenne du radium équivalent est de $(330 \pm 74) \text{ Bq.kg}^{-1}$ contre $(122 \pm 10) \text{ Bq.kg}^{-1}$ pour le sable. L'indice gamma moyen du granite est de $(1,2 \pm 0,3)$ contre $(0,43 \pm 0,03)$ pour le sable. Le débit de dose absorbée dans l'air moyen est de $(153,4 \pm 9,4) \text{ nGy.h}^{-1}$ contre $(53,7 \pm 5,1) \text{ nGy.h}^{-1}$ pour le sable. Pour les adultes, pour un facteur d'occupation de 80%, les doses efficaces annuelles moyennes (E) à l'intérieur et à l'extérieur d'un bâtiment sont de $(0,75 \pm 0,05) \text{ mSv.an}^{-1}$ et $(0,19 \pm 0,01) \text{ mSv.an}^{-1}$ contre $(0,26 \pm 0,05) \text{ mSv.an}^{-1}$ et $(0,07 \pm 0,01) \text{ mSv.an}^{-1}$ pour le sable. Pour un facteur de 60%, les doses efficaces sont $(0,56 \pm 0,03) \text{ mSv.an}^{-1}$ et $(0,38 \pm 0,02) \text{ mSv.an}^{-1}$ contre $(0,20 \pm 0,01)$ et $(0,13 \pm 0,01) \text{ mSv.an}^{-1}$ pour le sable. Pour les enfants, pour un taux d'occupation du bâtiment de 80%, les E moyens sont de $(0,86 \pm 0,05) \text{ mSv.an}^{-1}$ et $(0,21 \pm 0,01) \text{ mSv.an}^{-1}$ contre $(0,30 \pm 0,01) \text{ mSv.an}^{-1}$ et $(0,08 \pm 0,01) \text{ mSv.an}^{-1}$ pour le sable respectivement pour l'intérieur et l'extérieur. Pour les nourrissons, les E sont de $(1,00 \pm 0,06) \text{ mSv.an}^{-1}$ et $(0,25 \pm 0,01) \text{ mSv.an}^{-1}$ contre $(0,35 \pm 0,01) \text{ mSv.an}^{-1}$ et $(0,09 \pm 0,01) \text{ mSv.an}^{-1}$ pour le sable respectivement pour l'intérieur et l'extérieur. Pour les enfants, pour un facteur d'occupation du bâtiment de 60 %, les E moyens sont de $(0,64 \pm 0,03) \text{ mSv.an}^{-1}$ et $(0,43 \pm 0,02) \text{ mSv.an}^{-1}$ contre $(0,23 \pm 0,01) \text{ mSv.an}^{-1}$ et $(0,15 \pm 0,01) \text{ mSv.an}^{-1}$ pour le sable respectivement à l'intérieur et à l'extérieur. Pour les nourrissons, pour un facteur d'occupation du bâtiment de 60 %, les E sont de $(0,75 \pm 0,05) \text{ mSv.an}^{-1}$ et $(0,50 \pm 0,02) \text{ mSv.an}^{-1}$ contre $(0,26 \pm 0,01) \text{ mSv.an}^{-1}$ et $(0,18 \pm 0,01) \text{ mSv.an}^{-1}$ pour le sable respectivement à l'intérieur et à l'extérieur. Pour les adultes, l'excès de risque du cancer au cours de la vie à 66 ans pour la dose efficace totale induite par le granite est de $(3,1 \pm 0,01).10^{-3}$ contre $(1,1 \pm 0,01).10^{-3}$ pour le sable. Pour les enfants et les nourrissons, les ELCR moyens est de $(3,5 \pm 0,01)E-3$ et $(4,1 \pm 0,01)E-3$ contre $(1,2 \pm 0,01)E-3$ et $(1,4 \pm 0,01)E-3$ pour le sable respectivement. L'analyse statistique des données



a été faite avec Python 3.11 et R 4.3.2 sur l'interface Spyder et studio. La p-value est $< 0,001$ par rapport aux valeurs de référence de l'UNSCEAR pour le débit de dose absorbé ambiant, la dose efficace totale et l'excès de risque qui sont respectivement 84 nGy.h^{-1} , $0,48 \text{ mSv.an}^{-1}$ et $0,29.10^{-3}$. Il ressort que les granites des carrières étudiées présentent des risques radiologiques en cas de leurs utilisations comme matériaux de construction et requièrent des mesures spécifiques de radioprotection.

Mots clés: radionucléide, dose efficace, activité spécifique, dose absorbée, excès de risque du cancer.

1. INTRODUCTION

The exposure of man and his environment to ionizing radiation of natural terrestrial origin is based both on the physical phenomenon of the radioactive daughter of heavy atoms (mother nuclide) and on the radioactive decay of light atoms present in the earth's crust. The earth's crust is composed of radionuclides in varying quantities, depending on the geology of the soil. Radiation from the earth is the result of nuclear energy emanations produced by the radioactive decay of natural radionuclides composed of uranium-238 (^{238}U), uranium-235 (^{235}U) and thorium-232 (^{232}Th) [1] and the radioactive decay of potassium-40 (^{40}K) and is one of the main sources of existing exposure [2]. These radionuclides are found in building materials such as granite [3, 4] and sand [5].

Granite is a plutonic rock with a grainy texture. Rich in quartz, it contains more alkali feldspar than plagioclase. It is generally made up of the following minerals: quartz, potassium feldspars, plagioclases and micas. According to Breda in 1989, the rocks of the Benin hills are very varied in terms of their lithology [6]. They are crushed by hand by the population of the Dassa-zoume and Glazoue communes, particularly women and children, to meet the high demand from construction companies for community buildings and roads.

The use of these granites contributes to public exposure to low doses of ionizing radiation, and thus to stochastic effects. These effects are due to biological changes in exposed cells, and in particular in cellular deoxyribonucleic acid (DNA), induced by exposure to ionizing radiation. These are late effects, appearing several years or even decades after exposure. This category includes cancers [7]. Zinsou's work in 2014 showed that artisanal granite stone crushers are exposed to granite dust

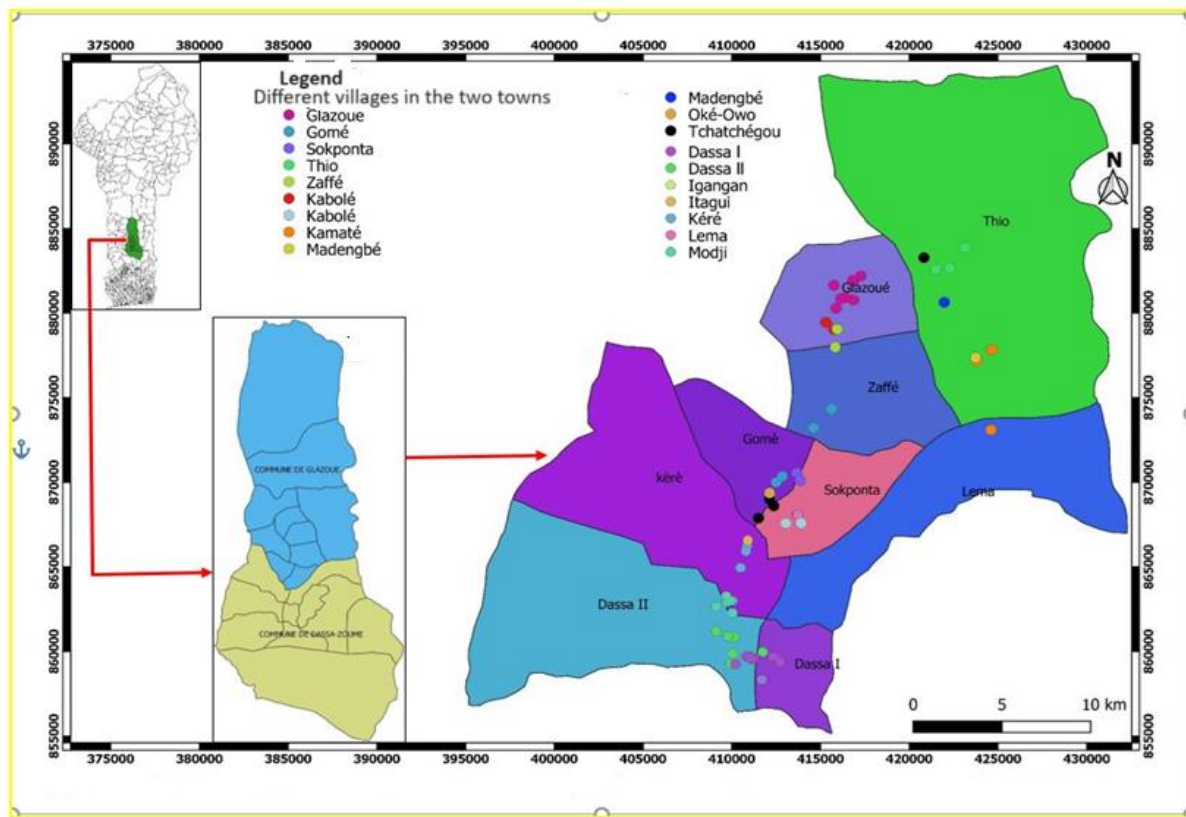
on a daily basis. This work showed the presence of chromosomal aberrations in the peripheral blood lymphocyte cells of these crushers [8].

According to the International Atomic Energy Agency (IAEA), the limit for specific activity concentrations of these radionuclides is set at 1 Bq.g⁻¹ for uranium and thorium-232. The limit for ⁴⁰K is 10 Bq.g⁻¹ [2]. As part of a preventive strategy, it is therefore important to assess the radiochemical composition of granites in the Dassa-zoume and Glazoue communes. This study will determine the radiological risk factors relating to the presence of natural radionuclides and will help predict the probability of cancerous diseases occurring in populations living in dwellings built from these granites.

1.1. Study area

Dassa-zoume and Glazoue are two communes in the Collines department of Benin, about 203 km and 234 km from Cotonou [9]. The department lies between 7° and 8° north latitude and meridians 1°60' and 2°80' east longitude. There are rocky outcrops that encourage the development of raw mineral soils unsuitable for agriculture [10]. These rocky detachments constitute artisanal crushing sites for the local population. They work there all day long with their offspring. Our work was carried out in five (05) arrondissements of each of the two communes, namely: Dassa I, Dassa II, Kere, Tre, Lema et Gome, Sonkpota, Thio, Zaffe and Glazoue. Figure 1 shows the geolocalized map of the sites explored in the study area.

Figure 1: Geolocation map of the study area



2. MATERIALS AND METHODS

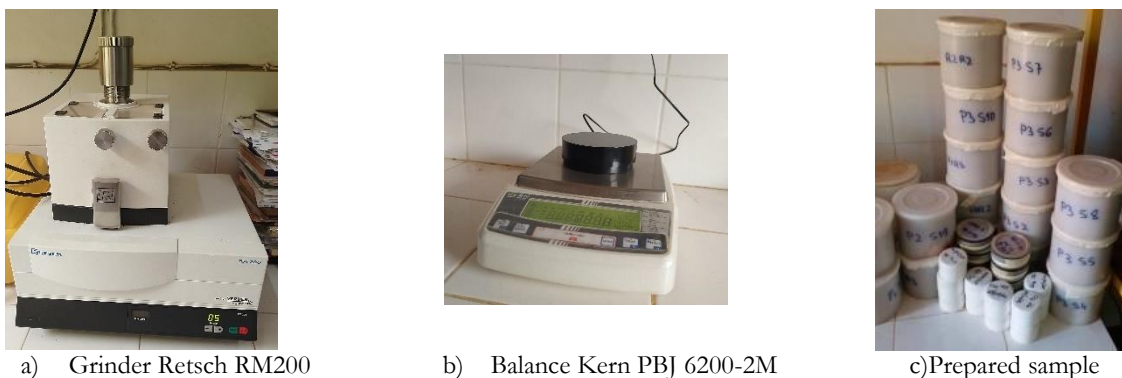
2.1. Sample collection and preparation

Sampling sites were selected according to ambient dose rate levels in the twelve granite crushing quarries in the two (02) communes. Granite samples weighing 250 g each were collected and placed in sterile, well-labelled vials. No preservatives were added to the samples. Laboratory work was carried out at the Analyses and nuclear technical department of the Institut National des Sciences et Techniques Nucléaires de Madagascar (INSTN-Madagascar).

Prior to analysis, the samples were ground to homogeneity using the Retsch RM 200 mill. These powdered samples were then placed in hermetically sealed 100

cm³ cylindrical sample holders for one month, to achieve secular equilibrium between ²²⁶Ra and its progeny. The masses of the samples analyzed were measured using a Kern PBJ 6200-2M precision electronic balance and used to calculate the mass activities (Bq.kg⁻¹) of the three radionuclides ⁴⁰K, ²³⁸U and ²³²Th. Figure 2 shows the equipment and materials used during sample preparation.

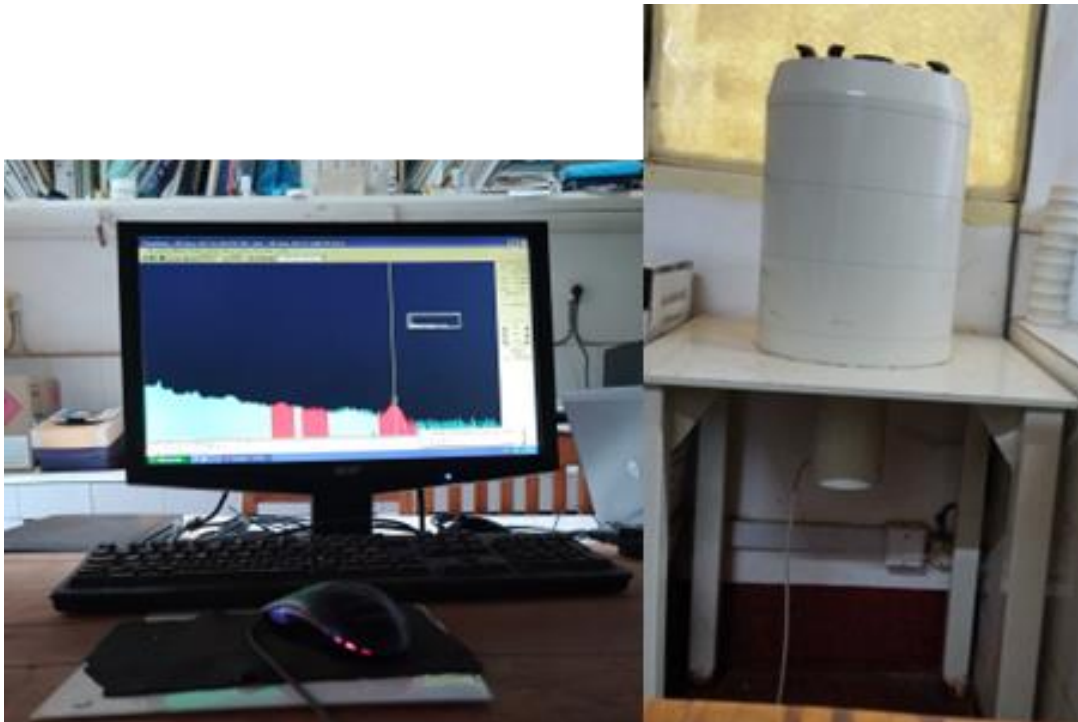
Figure 2: Sample preparation equipment, INSTN-Madagascar laboratory



2.2. Sample analysis

Analyses were carried out using gamma-rays spectrometry with an Ortec NaI(Tl) "3x3" detector (Figure 3). This type of scintillation detector is based on the interaction of radiation emitted by the radionuclides present with the detection medium [5,11]. As natural radionuclides are ubiquitous in the environment, this detector is housed in a lead castle with a thickness of 5 cm of lead and 2 mm of copper, to minimize the contribution of ambient radioactivity. The detector is connected to a preamplifier, a multi-channel analyzer and a microcomputer. The latter houses the ScintiVision software for adjusting measurement chain parameters, starting acquisition, storing spectra and processing. The various parameters were determined according to the methodology described in the reference document "Génie TM 2000" [12].

Figure 3: Gamma-ray spectrometry with NaI(Tl) "3x3" detector, INSTN-Madagascar laboratory



2.2.1. Energy calibration

Energy calibration is the process of determining the relationship between energy E and the channel number of a given radionuclide. To do this, we note the numbers of the channels of the peaks of total absorption and its corresponding energies. The data obtained make it possible to plot the energy calibration curve (Figure 4). The smoothing of the curve by a linear function therefore makes it possible to establish this calibration equation.

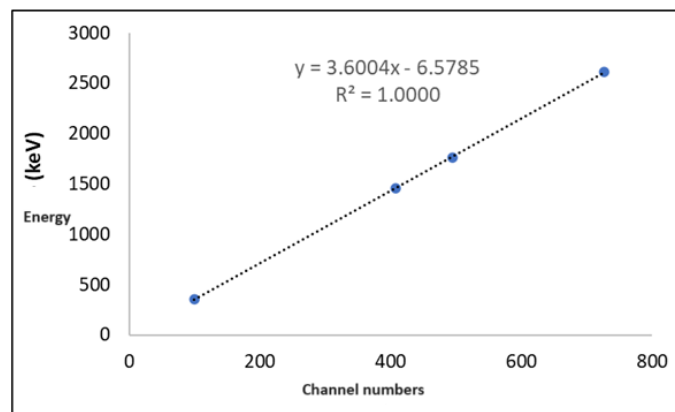
To maximize the resolution of the NaI(Tl) detector, four photoelectric peaks were exploited (Table 1).

The phenomenon of self-absorption was corrected by using the high energies of the photoelectric peaks.

Table 1: Photoelectric peak energies and its corresponding channel numbers

Channel numbers	Energy (keV)
110	351.5
414	1461.0
497	1764.5
730	2614.5

Figure 4: Energy calibration curve



2.2.2. Efficiency calibration

As illustrated in Figure 5, the method used exploits three gamma energy regions of interest 1461 keV, 1764 keV and 2614 keV to determine the specific activities ^{40}K , ^{214}B and ^{208}Tl respectively. As ^{238}U and ^{232}Th are not gamma emitters, they are determined from their progeny ^{214}Bi and ^{208}Tl respectively. The width of the regions of interest (ROI) is set at 10% of the maximum energy peak of a characteristic nuclide [13]. The measurement time for each sample is set at 24 hours to obtain good counting statistics.

The ROI is determined at the base of the photopeak. By setting the width to 10%, this region is slightly increased by increasing the energy. The ROI is different than the other parameter such as energy resolution of the NaI(Tl) detector. This energy resolution of a detector system is obtained from the peak full width at one-half of the maximum height (FWHM) which is decreased by increasing gamma ray energy.

Quality control of the equipment was carried out using IAEA-certified reference materials RGK-1, RGU-1 and RGTh-1. These are potassium, uranium and thorium ores. Thus, these are the geological standards. In view of the characteristic energy of the photoelectric peaks used ($E > 200$ keV), the phenomenon of self-absorption is insignificant.

It should also be noted that the calibration constant (ϵ) is determined from these reference materials RGK-1, RGU-1 and RGTh-1 according to the following relationship:

$$\epsilon = \frac{\tau_{net}}{A \cdot P_{\gamma} \cdot m} \tag{1}$$

- With τ_{net} : Net photoelectric peak rate (cps),
- A : activity of the reference material at the measurement date ($\text{Bq} \cdot \text{kg}^{-1}$),
- P_{γ} : Probability of emission or gamma intensity,
- m : mass of the reference material analyzed (kg).

The net rate of the photoelectric peak (τ_{net}) is given by the following relationship:

$$\tau_{net} = \frac{N_{rf}}{tc_{rf}} - \frac{N_{bg}}{tc_{bg}} \tag{2}$$

Where N_{rf} et N_{bg} : Count number of photoelectric peaks measured from the reference material and the background respectively.

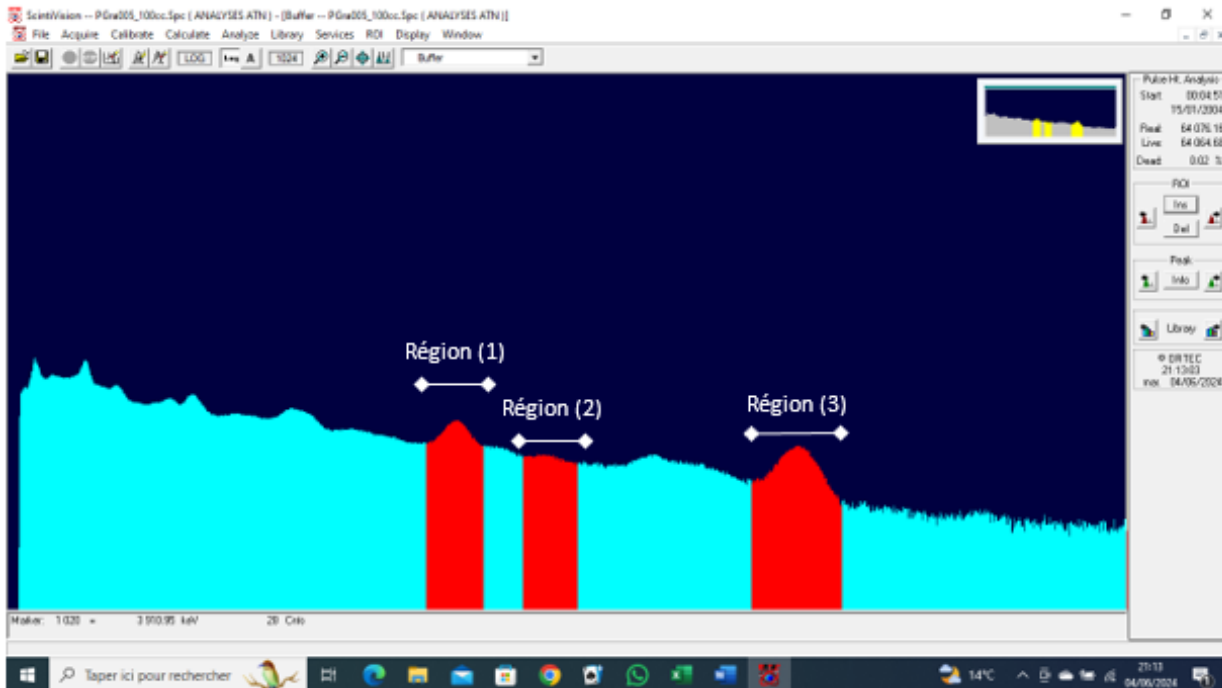
tc_{rf} et tc_{bg} : Counting time of the reference material and background respectively.

After analysis, the values obtained for the calibration constants are presented in Table 2.

Table 2: Calibration Constants

Radionuclides	Energy (keV)	Efficiencies constants		
		Geometry 25 cm ³	Geometry 100 cm ³	Geometry 200 cm ³
⁴⁰ K	1461.0	0.159635	0.020092	0.017346
²¹⁴ Bi	1764.5	0.123267	0.016654	0.016124
²⁰⁸ Tl	2614.5	0.082641	0.0099211	0.009869

Figure 5: Gamma spectrum of a granite sample illustrating the three regions of interest



This principle is also used to assess indirectly, through mathematical formulae, the atmospheric radon concentration in dwellings built using selected and analyzed building materials. According to the literature, Kall *et al.* (2014) used this technique in Madagascar to assess the mass concentration of ^{238}U , ^{235}U , ^{232}Th and ^{40}K in the sands of the Ramena beaches [5]. Kurnaz, Kovacs, Onjefu and Sidique *et al.* respectively in 2021 in Turkey and Iran and in 2022 in Namibia and Egypt used this technique to assess the mass concentration of ^{238}U or ^{226}Ra , ^{232}Th and ^{40}K in granites, building materials, ceramic tiles and marble [14-16].

Thus, we used the measured activity concentrations to calculate the absorbed dose rates in air at 1 m height at the ground surface (granite/sand) through equation 5 described below.

2.3. Radiological risk assessment

Equations 1, 2, 3, 4 and 5 were used to calculate equivalent radium activity, gamma index, absorbed dose rate in air, external effective dose estimated induced by a granite or sand habitat and excess cancer risk at age 66, respectively.

2.3.1. Calculation of radium equivalent activity (Ra_{eq})

Ra_{eq} is one of the most effective radiological indices for determining gamma radiation exposure risks due to the radionuclides ^{40}K , ^{238}U and ^{232}Th in building materials, given the non-uniform distribution of these radionuclides in the material. According to Sidique et al (2022), Ra_{eq} is described by equation 1 in the UNSCEAR 2000 report on the sources and effects of ionizing radiation [17,18].

$$Ra_{eq} [Bq/kg] = \left[\frac{AC_U}{370} + \frac{AC_{Th}}{259} + \frac{AC_K}{4810} \right] \times 370 \quad (3)$$

where AC_U , AC_{Th} and AC_K are the specific activities of the radionuclides ^{238}U (^{226}Ra), ^{232}Th and ^{40}K , respectively. This is a weighting of the concentrations of these three radionuclides in building materials, based on the principle of equality of the gamma dose rates of 4,810 Bq.kg^{-1} of ^{40}K , 259 Bq.kg^{-1} of ^{232}Th and 370 Bq.kg^{-1} of ^{238}U (^{226}Ra).

2.3.2. Calculation of the Gamma Index (I_γ)

The Gamma Index (I_γ) is considered a control tool for assessing whether or not building materials are safe to use. Assuming that external exposure to gamma radiation from cladding materials (surface or roof) is limited to 1 mSv.y^{-1} , the gamma index adopted by the European Commission [19] is estimated using equation 2.

$$I_\gamma = \frac{AC_{Ra}}{A_{Ra}} + \frac{AC_{Th}}{A_{Th}} + \frac{AC_K}{A_K} \leq 1 \quad (4)$$

Where AC = activity concentration measured by gamma spectrometry and A_x is the reference value for the activity concentration of radionuclide X .

For this work, the reference values of the European Commission have been used, i.e. $A_{Ra}=300\text{Bq.kg}^{-1}$, $A_{Th}=200\text{ Bq.kg}^{-1}$ and $A_K=3000\text{Bq.kg}^{-1}$.

According to the European Commission, for roofing materials such as granites that have a gamma index $I_\gamma \leq 2$, lead to an increase in the annual dose rate of 0.3 mSv.y^{-1} from these materials. Furthermore, if the materials meet the criteria $2 < I_\gamma \leq 6$, they will contribute 1 mSv.y^{-1} to the annual gamma dose rate and will be within the recommended limit for the public. Finally, materials with $I_\gamma > 6$ are not suitable for safe use in buildings [19].

Furthermore, according to the UNSCEAR report in 2000, the value of the gamma index I_γ must be less than 1 for the radiation risk to remain insignificant [17]. This means that the maximum value of I_γ is equal to 1 and corresponds to the upper limit of the equivalent activity of ^{226}Ra which is 370 Bq.kg^{-1} .

Calculation of absorbed dose rate in air and annual external effective dose for adults, children or infants inside a home is one of the important parameters for determining external exposure caused by terrestrial radionuclides (^{232}Th , ^{238}U and ^{40}K). According to the UNSCEAR reports 1993 and 2008, equations 5 and 6 can be used to estimate D_{in} and E_{in} in air inside buildings constructed from granite and ceramic tiles [19,20]. The concept of effective dose was developed by the International Commission on Radiological Protection (ICRP) in its report 147 as a risk-adjusted dosimetry quantity for the management of protection against stochastic effects, mainly cancer, enabling comparison of estimated doses with dose limits, dose constraints and reference levels expressed in the same quantity [22]. Bearing in mind the uncertainties associated with risk projection to low doses or low dose rates, ICRP had concluded that effective dose may be considered as an approximate indicator of

possible ionizing risk, recognizing also that lifetime cancer risks (ELCR) vary with age at exposure, sex and population group [22].

$$\dot{D}[nGy \cdot h^{-1}] = 0.462AC_{U-238} + 0.604AC_{Th-232} + 0.0417AC_{K-40} \quad (5)$$

Where AC_U , AC_{Th} and AC_K are respectively the activity concentrations measured in granite samples of natural radionuclides ^{238}U , ^{232}Th and ^{40}K . According to UNSCEAR 2008 report, the constants 0.462, 0.604 and 0.0417 are dose conversion factors (DCF) to absorbed dose rate in air for ^{238}U , ^{232}Th and ^{40}K respectively in granite or sand sample [21]. In order to assess the effects of ionizing radiation on the human body, an effective dose from ionizing radiation is calculated. The effective dose is an estimate of the whole-body dose of radiation received that takes into account the sensitivity of the internal organs to ionizing radiation.

$$E[mSv \cdot y^{-1}] = \dot{D}[nGy \cdot h^{-1}] \times F_1 \times F_2 \times F_3 \times 10^{-6} \quad (6)$$

where F_1 is the conversion factor from absorbed dose rate in air induced by granite or sand to annual effective dose received by adults, children and infants i.e. $0.7 Sv.Gy^{-1}$, $0.8 Sv.Gy^{-1}$ and $0.93 Sv.Gy^{-1}$ respectively [18,20]. F_2 is the indoor occupancy factor, i.e. 0.8 [18]. F_3 is the annual duration, equal to 8760 hours. This means that, according to UNSCEAR, people spend 80% of their time inside buildings and 20% outside. This hypothesis is based on temperate countries (winter). Some authors indicate that, in Sub-Saharan Africa climatic conditions, populations spend about 60% of their time inside buildings and 40% outdoors [23].

The excess lifetime cancer risk (ELCR) is an important parameter for calculating the incidence of cancer for an individual exposed to a low dose of gamma radiation over a lifetime (66 years). The ELCR can be estimated using equation 7, which is a function of the effective dose experienced by individuals due to low doses of radionuclides present in ceramic and granite tiles [17,24, 25].

$$ELCR = E[Sv \cdot y^{-1}] \times C_1 \times C_2 \quad (7)$$

where C_1 is the life span. As life expectancy is not yet calculated in our study area, we estimated the ELCR with an average lifespan of the population equal to 66 years. This mean age was considered within the mean range 60 to 69 years of Table 2.4 of the ICRP Report 147 in 2019 for liver cancer in both men and women [22]. According to the conclusion of the report 147 of ICRP, C_2 is a nominal lifetime fatal cancer risk of approximately 5.10^{-2} per Sv applies at low doses or low dose rates (i.e. $< 10^{-4}$ per mSv) [24].

3. RESULTS AND DISCUSSIONS

3.1. RESULTS

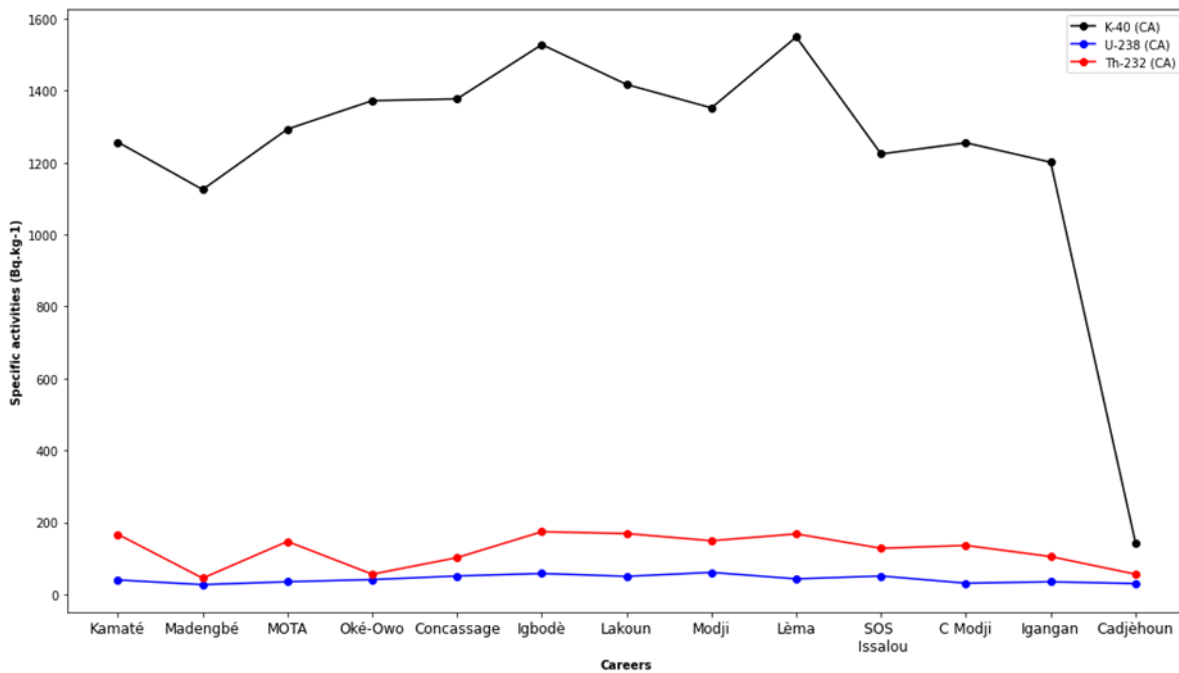
3.1.1. Specific activity

Figure 6 shows the specific activities found in twelve (12) granite samples taken from granite quarries in the Communes of Dassa-zoume and Glazoue, and one (01) sand sample taken from the Cadjehoun district of Cotonou. The concentration of specific activity of the three radionuclides in the granite sample taken from the artisanal crushing quarry of Kamate is (1257 ± 38) Bq.kg⁻¹ for ⁴⁰K, (40 ± 2) Bq.kg⁻¹ for ²³⁸U and (167 ± 8) Bq.kg⁻¹ for ²³²Th. For the sample taken at Society Mota Quarry 2, the specific activity concentration is (1377 ± 41) Bq.kg⁻¹ for ⁴⁰K, (51 ± 2) Bq.kg⁻¹ for ²³⁸U and (102 ± 7) Bq.kg⁻¹ for ²³²Th. These uncertainties were obtained with the technique of gamma spectrometry with NaI(Tl) detectors.

For the granites, the specific activities of ⁴⁰K, ²³⁸U and ²³²Th ranged respectively from (1125 ± 34) Bq.kg⁻¹ to (1549 ± 48) Bq.kg⁻¹, from (27 ± 2) Bq.kg⁻¹ to (61 ± 4) Bq.kg⁻¹ and from (45 ± 6) Bq.kg⁻¹ to (174 ± 11) Bq.kg⁻¹. The mean specific activities found were (1329 ± 128) Bq.kg⁻¹, (44 ± 11) Bq.kg⁻¹ and (129 ± 44) Bq.kg⁻¹. The uncertainties of these mean values are represented by the standard deviations.

For sand, the specific activities of three radionuclides are $(144 \pm 8) \text{ Bq.kg}^{-1}$, $(30 \pm 2) \text{ Bq.kg}^{-1}$ and $(56 \pm 7) \text{ Bq.kg}^{-1}$ respectively. These specific activities are well below the average values found in granites.

Figure 6: Radiochemical composition of granites from different quarries



3.1.2. Analysis of results by principal component

Figure 7 shows the results of statistical principal component analysis (PCA) on granite samples taken from the twelve (12) artisanal crushing quarries in the two above-mentioned Communes. This analysis describes the relationships between the radiochemical compositions of the granites and the quarries of the production communes, such as the correlation circle of the radiochemical compositions (Figure 7a) and the projection of the quarries of the production communes in the first factorial plane formed by axes 1 and 2 defined by the radiochemical compositions (Figure 7b).

Statistical principal component analysis showed that all concentrations were positively correlated with the first axis. Concentrations of ²³²Th are positively correlated with the second axis, while those of the other two radionuclides are negatively correlated with this axis. ²³²Th is much more concentrated in the Lema

and Lakoun quarries, while ^{40}K and ^{238}U are more concentrated at Tchatchegou and Modji. The C quarries at Modji, Mota, Igangan, Issalou, Madengbe and Oke Owo are all less rich in the radiochemical composition of these three radionuclides.

Figure 7: Principal component analysis of results.

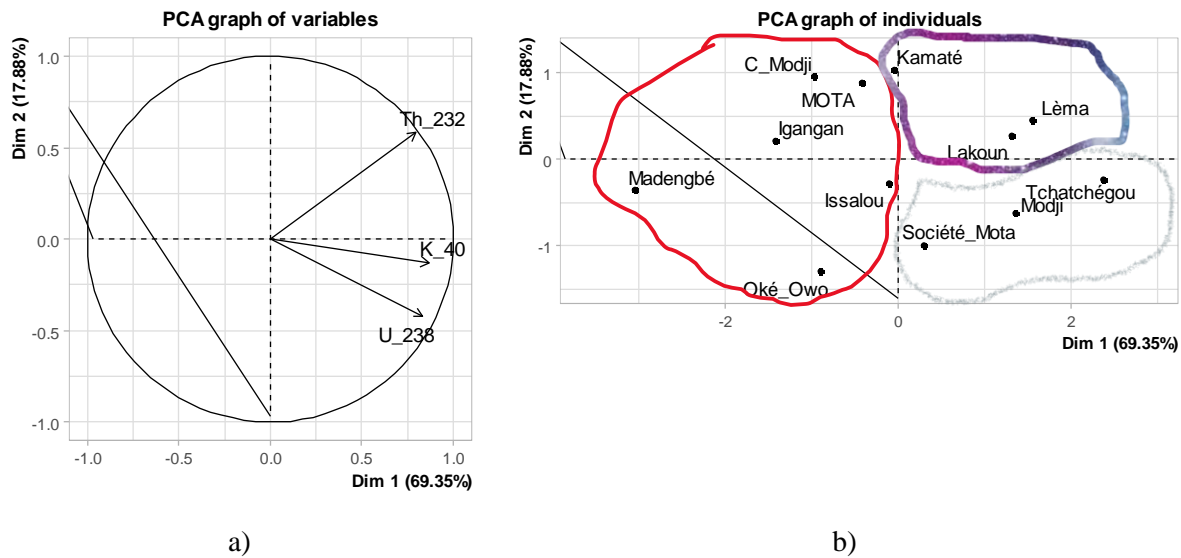


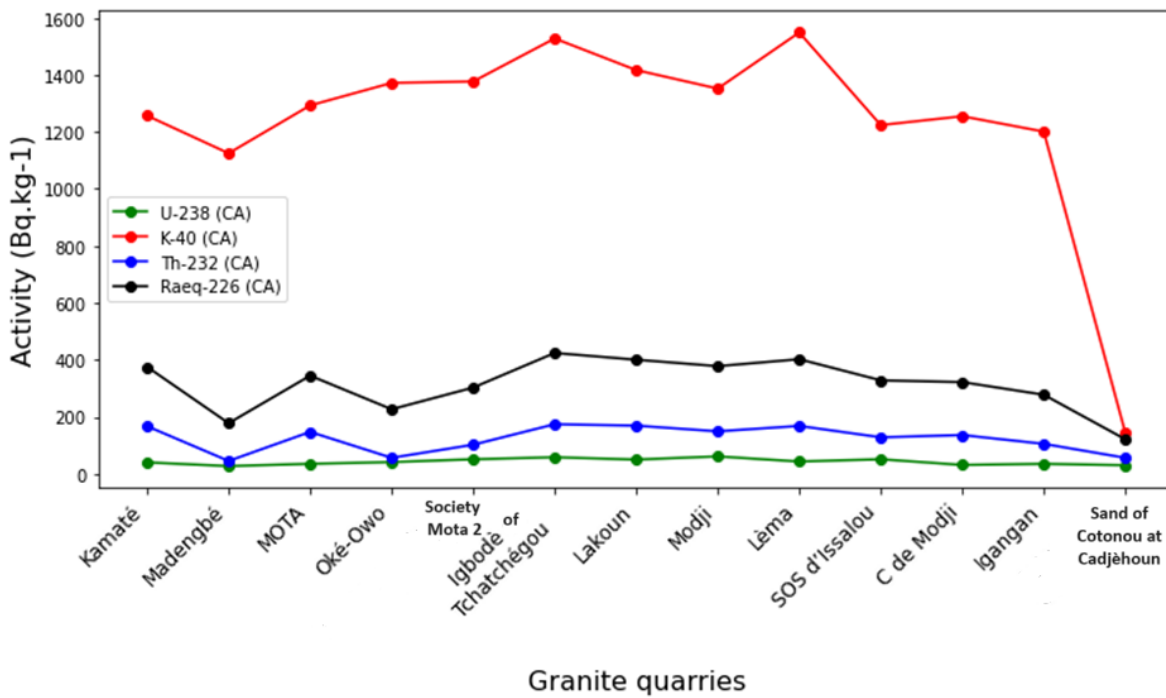
Figure 7 shows that the first component explains 69.35% of the information, and that the first two components explain 87.23% of the information sought. The correlation circle for granite radiochemical composition groups (Figure 7a) shows that all radiochemical compositions are positively correlated with the first axis.

The ^{232}Th radiochemical composition is positively correlated with the second axis, while the ^{40}K and ^{238}U compositions are negatively correlated with the same axis (Figure 7a). Projection of the quarries of the two Communes in the system of the first two axes (Figure 6b) shows that radiochemical composition ^{232}Th is much more concentrated in the Lema and Lakoun quarries, and less concentrated at Kamate. The ^{40}K and ^{238}U compositions are more concentrated at Tchatchegou and Modji, and less concentrated at Society Mota 2's crushing quarry 2. The C quarries at Modji, Mota, Igangan, Issalou, Madengbe and Oke-Owo are all less rich in radiochemical compositions (Figure 7b).

3.1.3. Equivalent activities of radium (Ra_{eq}) in granites

Based on equation 3, the activities of radium in granites are shown in Figure 8.

Figure 8: Radium equivalent activity in granites



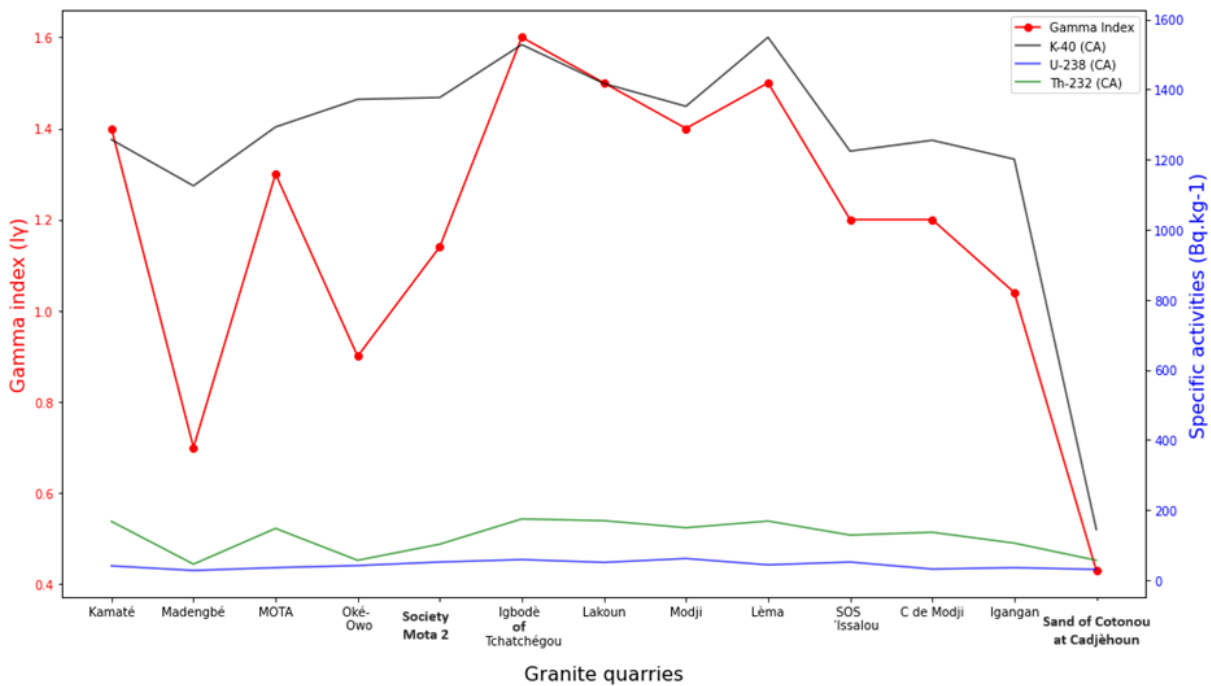
According to the results, the activity of radium equivalent in granites varies from (178 ± 10) Bq.kg⁻¹ to (424 ± 16) Bq.kg⁻¹ with a mean of (330 ± 74) Bq.kg⁻¹. The amount obtained in the sand collected in Cotonou is equal to (122 ± 10) Bq.kg⁻¹.

Figure 8 shows that the variation in the activity of equivalent radium follows that of ²³²Th and ²³⁸U. The high values are found in the quarries of Igbođe, Lema and Lakoun, which are respectively (424 ± 16) Bq.kg⁻¹, (402 ± 16) Bq.kg⁻¹ and (400 ± 12) Bq.kg⁻¹.

3.1.4. Gamma Index (I_γ)

Based on equation 4, figure 9 presents the distributions of the gamma index of the populations living in buildings built from granite from twelve (12) quarries in the Communes of Glazoue and Dassa-zoume.

Figure 9: Gamma indices of populations living in dwellings built from granite



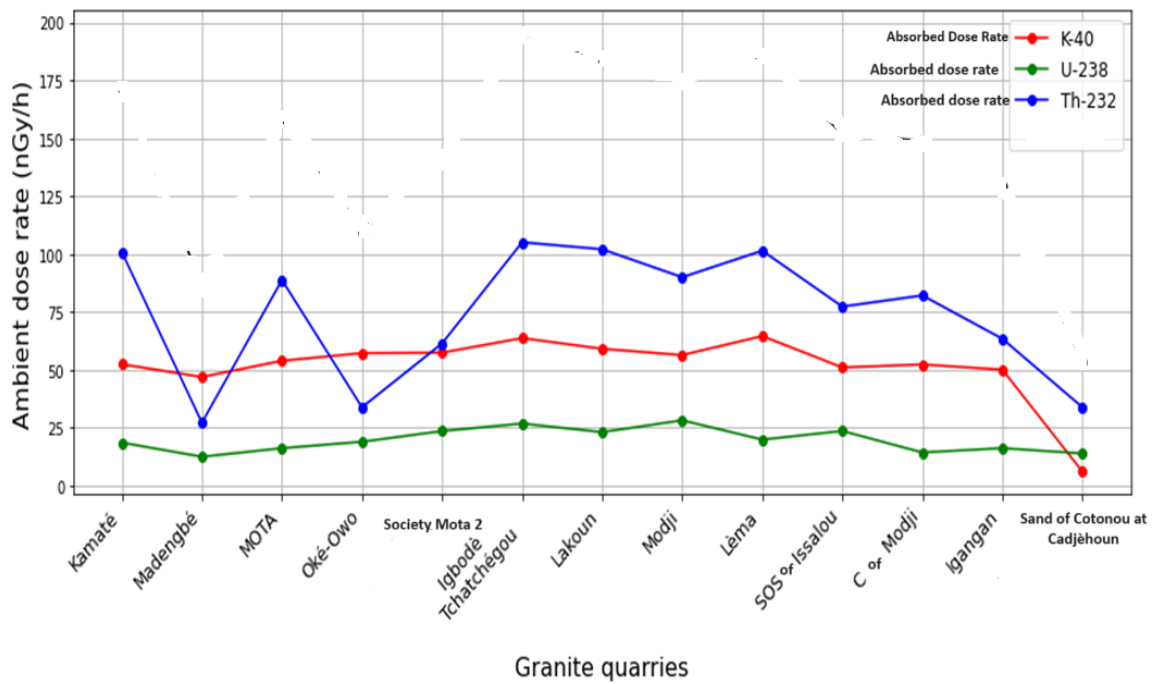
The values of gamma index obtained vary from 0.7 to 1.6 with a mean of 1.2. The maximum value is at the Igbode quarry in Tchatchegou. While the one in Madengbe has the minimum value. For the sand sample taken in Cotonou, the gamma index obtained is equal to 0.43.

3.1.5. Absorbed dose rate in air (\dot{D}) induced by granite and annual effective doses (E)

Based on equations 5 and 6, the results of absorbed dose rates in air (\dot{D}) due to granites from the Commune of Glazoue and Dassa-zoume, used as building materials (social housing and/or housing) are presented in Figures 10 and 11.

Based on Equation 5, Figure 10 presents the values of the absorbed dose rate in air induced by each radionuclide in the granite sample of each quarry.

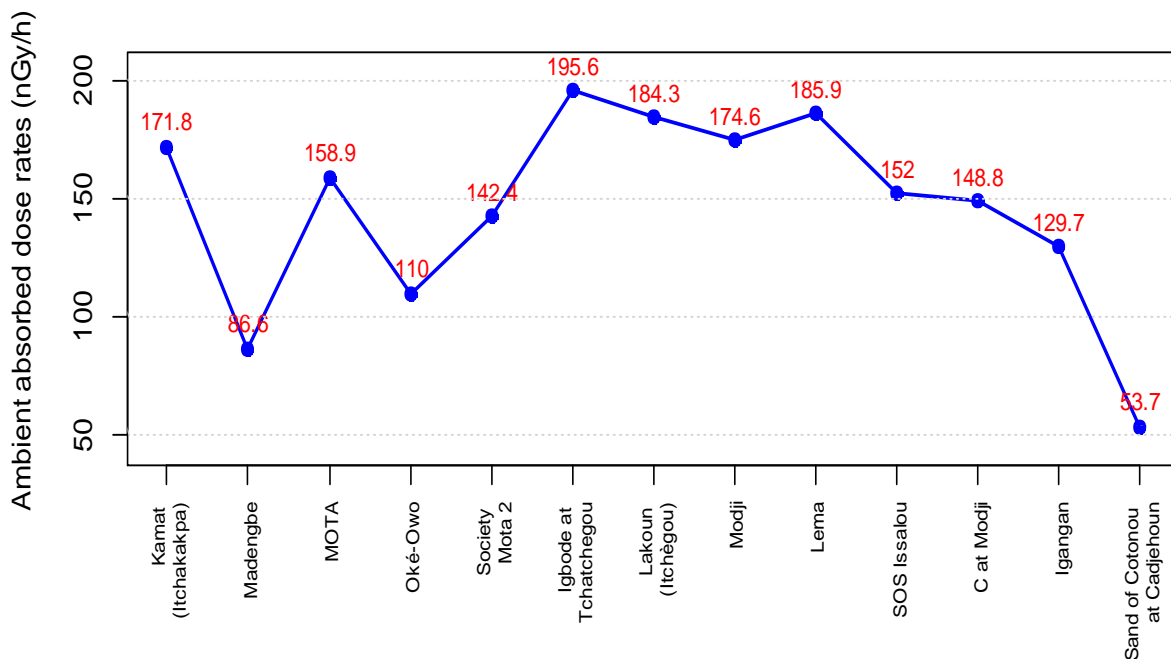
Figure 10: Variation of absorbed dose rate in air induced by granites as a function the quarries.



As seen in the figure 10 the values of the absorbed dose rate in air induced by the radionuclide ⁴⁰K in granites collected from artisanal crushing quarries in the communes of Glazoue and Dassa-zoume varied from 46.9 to 64.6 nGy.h⁻¹ with an average of (55.4 ± 5.1) nGy.h⁻¹. Those of radionuclide ²³⁸U ranged from 12.5 to 28.2 nGy.h⁻¹ with a mean of (20.1 ± 4.7) nGy.h⁻¹. Those of the radionuclide ²³²Th ranged from 27.2 to 105.1 nGy.h⁻¹ with a mean of (77.8 ± 31) nGy.h⁻¹. These averages are higher than the absorbed dose rate in air value induced by ⁴⁰K (6.0 nGy.h⁻¹), ²³⁸U (13.9 nGy.h⁻¹) and ²³²Th (33.8 nGy.h⁻¹) in the sands collected in Cotonou.

Based on Equation 5, Figure 11 presents the values of the total absorbed dose rate in air induced by three radionuclides in the granite sample of each quarry.

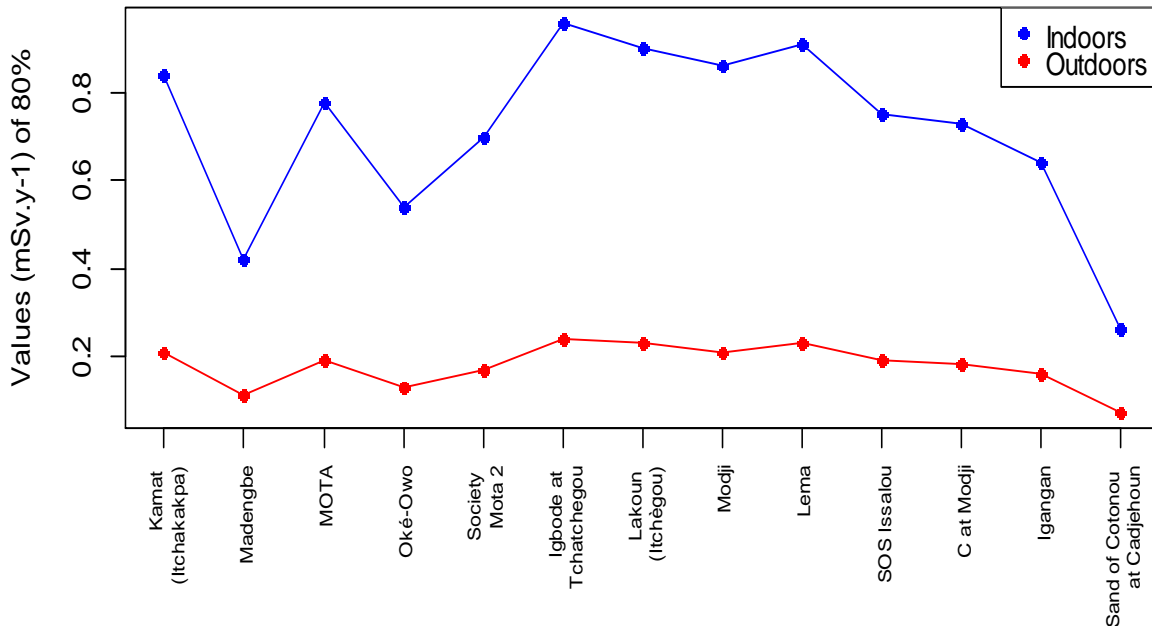
Figure 11: Total absorbed dose rate in air induced by granites as a function the quarries.



As seen in the figure 11 the overall absorbed dose rate in air induced by the three radionuclides in granites collected from the artisanal crushing quarries of the municipalities of Glazoue and Dassa-zoume varied from 86.6 to 195.6 nGy.h⁻¹ with a mean of (153.4 ± 9.4) nGy.h⁻¹. This average remains higher than the value of the global absorbed dose rate in air (53.7 nGy.h⁻¹) induced by the three radionuclides in the sands collected in Cotonou.

Based on Equation 6, Figure 12 and 13 presents the estimated values of the external effective dose inside and outside the building induced by the three (03) natural radionuclides for the building occupancy factor for 80% and 60%.

Figure 12: Variation of the annual effective dose for adults induced by granites for building occupancy factor for 80%.

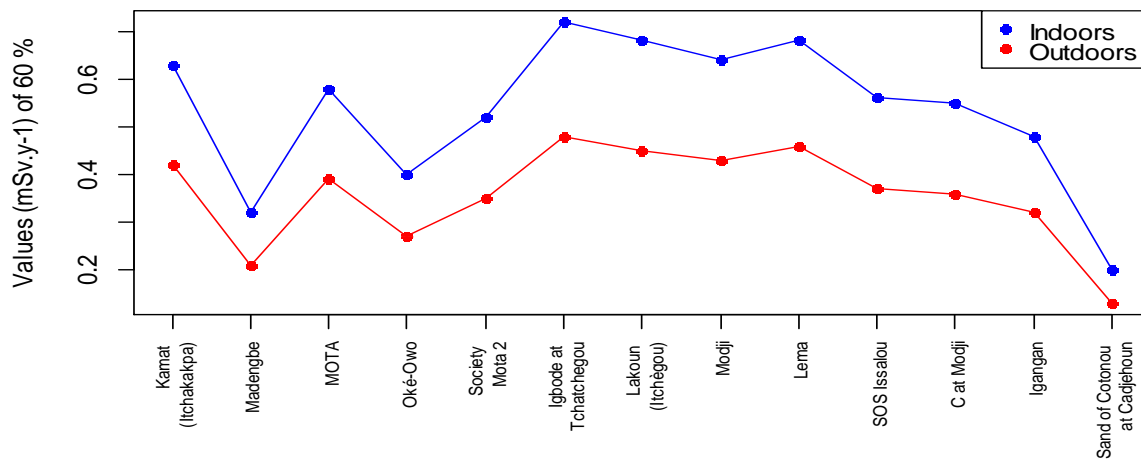


As seen in table 12, for a building occupancy factor of 80%, for adults, the induced annual external effective doses indoors range from (0.42 ± 0.05) mSv.y⁻¹ to (0.96 ± 0.1) mSv.y⁻¹ with a mean of (0.75 ± 0.05) mSv.y⁻¹. The annual external effective dose outdoors ranges from (0.11 ± 0.01) mSv.y⁻¹ to (0.24 ± 0.03) mSv.y⁻¹ with a mean of (0.19 ± 0.01) mSv.y⁻¹.

For children, for a building occupancy factor of 80%, the induced annual external effective dose indoors ranges from (0.49 ± 0.05) mSv.y⁻¹ to (1.10 ± 0.1) mSv.y⁻¹ with a mean of (0.86 ± 0.05) mSv.y⁻¹. The annual external effective dose outdoors ranges from (0.12 ± 0.01) mSv.y⁻¹ to (0.27 ± 0.03) mSv.y⁻¹ with a mean of (0.21 ± 0.01) mSv.y⁻¹.

For infants, for a building occupancy factor of 80%, the induced annual external effective dose indoors ranges from (0.56 ± 0.04) mSv.y⁻¹ to (1.27 ± 0.1) mSv.y⁻¹ with a mean of (1.00 ± 0.06) mSv.y⁻¹. The annual external effective dose outdoors ranges from (0.14 ± 0.01) mSv.y⁻¹ to (0.32 ± 0.03) mSv.y⁻¹ with a mean of (0.25 ± 0.01) mSv.y⁻¹.

Figure 13: Variation of the annual effective dose induced by granites for building occupancy factor of 60%.



As seen in table 13, for a building occupancy factor of 60%, for adults, the annual external effective dose indoors induced by granite to the representative public person range from (0.32 ± 0.02) mSv.y⁻¹ to (0.72 ± 0.05) mSv.y⁻¹ with a mean of (0.56 ± 0.03) mSv.y⁻¹. The external annual effective dose outdoors induced by granite to the representative public person range from (0.21 ± 0.03) mSv.y⁻¹ to (0.48 ± 0.05) mSv.y⁻¹ with a mean of (0.38 ± 0.02) mSv.y⁻¹.

For children, for a building occupancy factor of 60%, the annual external effective dose indoors induced by granite to the representative public person ranges from (0.36 ± 0.02) mSv.y⁻¹ to (0.82 ± 0.05) mSv.y⁻¹ with a mean of (0.64 ± 0.03) mSv.y⁻¹. The external annual effective dose outdoors induced by granite to the representative public person ranges from (0.24 ± 0.03) mSv.y⁻¹ to (0.55 ± 0.05) mSv.y⁻¹ with a mean of (0.43 ± 0.02) mSv.y⁻¹.

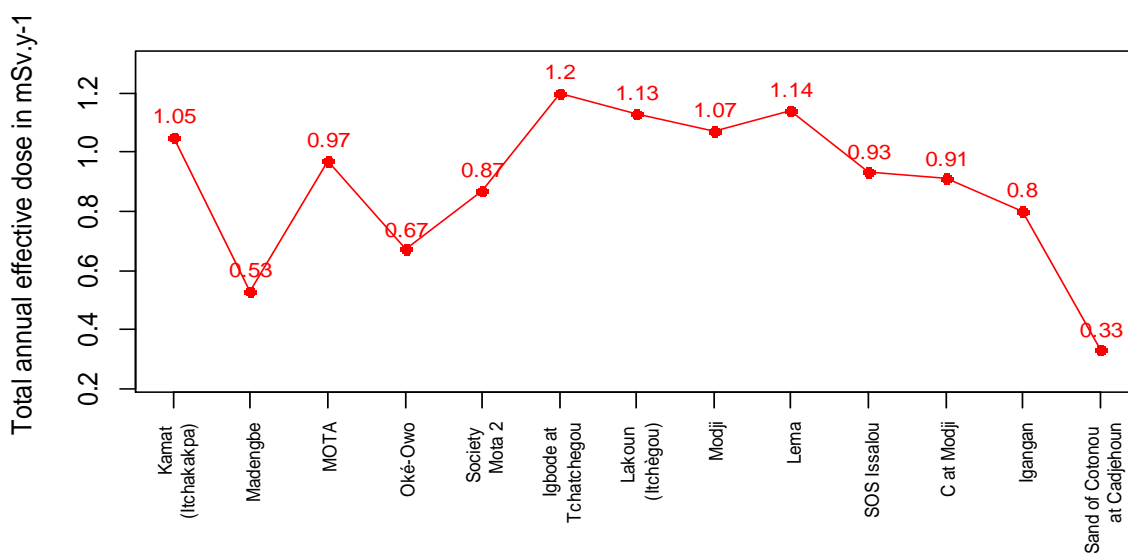
For infants, for a building occupancy factor of 60%, the annual external effective dose indoors induced by granite to the representative public person range from (0.42 ± 0.02) mSv.y⁻¹ to (0.96 ± 0.06) mSv.y⁻¹ with a mean of (0.75 ± 0.05) mSv.y⁻¹. The external annual effective dose outdoors induced by granite to the representative public person ranges from (0.28 ± 0.03) mSv.y⁻¹ to (0.64 ± 0.05) mSv.y⁻¹ with a mean of (0.50 ± 0.02) mSv.y⁻¹.

As seen in the figure 11, 12 and 13, the total absorbed dose rate obtained by the sand collected in Cotonou is equal to (53.7 ± 4.1) nGy.h⁻¹ with an estimated annual external effective dose induced by granite to the representative public person of (0.26 ± 0.01) mSv.y⁻¹ and (0.07 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors for a building occupancy factor of 80% for adults. For a building occupancy factor of 60%, the annual external effective dose are (0.20 ± 0.01) mSv.y⁻¹ and (0.13 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors with the sand of Cotonou for adults.

For children and infants, for a building occupancy factor of 80%, these annual external effective doses are induced by the sand of Cotonou are (0.30 ± 0.01) mSv.y⁻¹ and (0.08 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors, (0.35 ± 0.01) mSv.y⁻¹ and (0.09 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors.

For children and infants, for a building occupancy factor of 60%, these annual external effective doses are induced by the sand of Cotonou are (0.23 ± 0.01) mSv.y⁻¹ and (0.15 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors, (0.26 ± 0.01) mSv.y⁻¹ and (0.18 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors.

Figure 14: Variation of the total annual effective dose (indoors and outdoors) induced by granites for building.

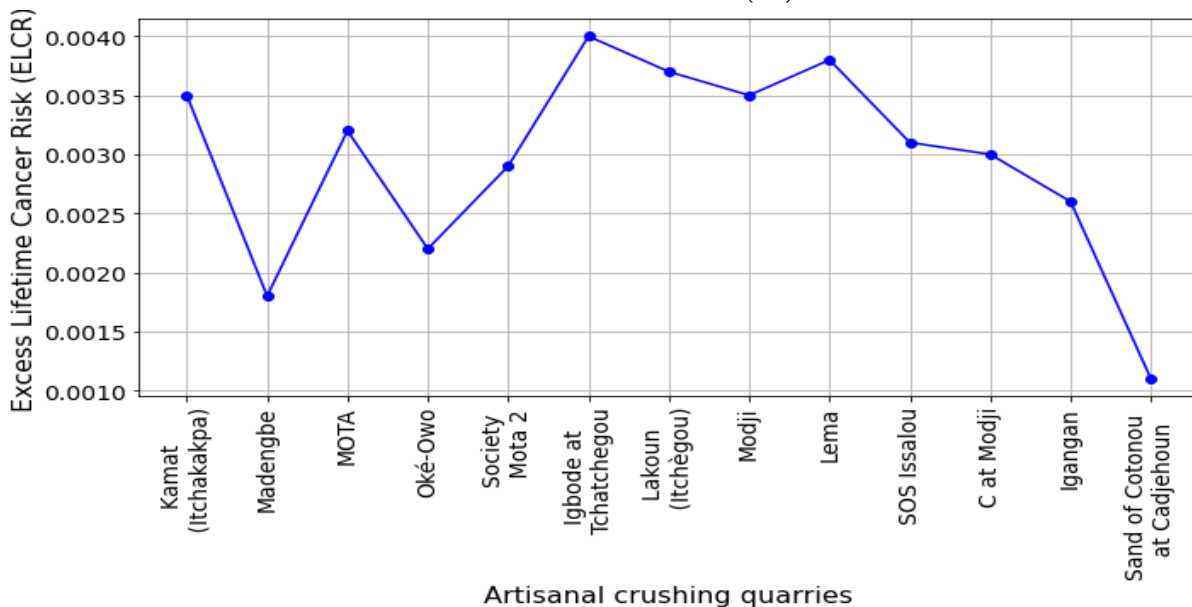


As seen in table 14, for adults, the total annual external effective dose induced by the granite to the representative public person (indoors and outdoors) ranges from 0.53 to 1.20 mSv.y⁻¹ with a mean of (0.94 ± 0.06) mSv.y⁻¹. For the children, the total annual external effective dose induced by the granite to the representative public person (indoors and outdoors) ranges from 0.61 to 1.37 mSv.y⁻¹ with a mean of (1.07 ± 0.06) mSv.y⁻¹. For the infants, the total annual external effective dose induced by the granite to the representative public person (indoors and outdoors) ranges from 0.71 to 1.59 mSv.y⁻¹ with a mean of (1.25 ± 0.1) mSv.y⁻¹. These values remain higher than the total annual effective dose obtained with the sand in Cotonou which are of (0.33 ± 0.03) mSv.y⁻¹, (0.38 ± 0.03) mSv.y⁻¹ and (0.44 ± 0.03) mSv.y⁻¹ respectively for adults, children and infants. The maximum values are at the Igbođe quarry in Tchatchégou.

3.1.6. Excess lifetime cancer risk (ELCR)

Based on equation 7, the results of the excess cancer risk of the population due to the use of these granites as building materials are presented in Figure 15.

Figure 15: Change in the population's excess cancer risk over their lifetime at age 66 as a function of the annual effective dose (E_{in}) for adults.



According to Figure 15, for adults, the excess cancer risk of the population during their lifetime at age 66 varies from (1.8 ± 0.01)E-3 to (4.0 ± 0.01)E-3 with an

average equal to $(3.1 \pm 0.01)E-3$. On the other hand, the one obtained with the sand sample taken in Cotonou is equal to $(1.1 \pm 0.01)E-3$.

For the children and infants, the excess cancer risk of the population during their lifetime at age 66 range from $(2.0 \pm 0.01)E-3$ to $(4.5 \pm 0.01)E-3$ with a mean $(3.5 \pm 0.01)E-3$ and from $(2.3 \pm 0.01)E-3$ to $(5.3 \pm 0.01)E-3$ with a mean $(4.1 \pm 0.01)E-3$ respectively. The ELCR obtained with the sand of Cotonou are $(1.2 \pm 0.01)E-3$ and $(1.4 \pm 0.01)E-3$ respectively for children and infants.

3.1.7. Statistical analyses

Table 3 presents the statistical analyses for results obtained on the absorbed dose rate in air and the external effective doses for adults.

Table 3: Comparison of averages of absorbed dose rate, effective doses and the excess cancer risk to the UNSCEAR 2000 and 2008 reference value

Data	One Sample t-test					
	df	t	Sample estimates: mean of x	Alternative hypothesis	p-value	
Absorbed dose rate in air in nGy.h ⁻¹	-	11	7.3818	153.383	Alternative hypothesis: true mean is greater than 84	<0,001*
External annual effective doses in mSv.y ⁻¹ for a building occupancy factor of 80%	Indoors	11	7.4203	0.752	Alternative hypothesis: true mean is greater than 0,41	<0,001*
	Outdoors	11	10.111	0.187	Alternative hypothesis: true mean is greater than 0.07	< 0.001*
External annual effective doses in mSv.y ⁻¹ for a building occupancy factor of 60%	Indoors	11	7.3394	0.563	Alternative hypothesis: true mean is greater than 0.31	< 0.001*
	Outdoors	11	9.7175	0.375	Alternative hypothesis: true mean is greater than 0.15	< 0.001*
External annual effective doses in mSv.y ⁻¹	Indoors and outdoors	11	7.9522	0.939	Alternative hypothesis: true mean is greater than 0,48	< 0.001*
Excess Lifetime Cancer Risk (ELCR)	Relating to Indoors and outdoors	11	14.796	0.003	Alternative hypothesis: true mean is greater than 0.00029	< 0.001*

The results indicate all probability values $p < 0.001$. This means that the absorbed dose rate in air (\dot{D}), the annual effective dose induced (E) for all building occupancy factor and the excess Lifetime cancer risk (ELCR) inside homes by the granites of these two municipalities is statistically different from that of the UNSCEAR 2000 and 2008 reference, which is respectively equal to 84 nGy.h^{-1} , 0.48 mSv.y^{-1} and 0.00029 [18].

3.2. DISCUSSIONS

3.2.1. Specific activities

Concerning the report of UNSCEAR in 1993, the overall average activities (MCVs) of ^{40}K , ^{238}U and ^{232}Th in ceramics and granites are 500 Bq.kg^{-1} , 50 Bq.kg^{-1} and 50 Bq.kg^{-1} respectively [20]. Indeed, the specific activity limit is 100 Bq.g^{-1} , 10 Bq.g^{-1} and 10 Bq.g^{-1} respectively [2]. The average specific activities found in our study in granite are $(1329 \pm 128) \text{ Bq.kg}^{-1}$, $(44 \pm 11) \text{ Bq.kg}^{-1}$, and $(129 \pm 44) \text{ Bq.kg}^{-1}$ for ^{40}K , ^{238}U , and ^{232}Th respectively. They are significantly higher than the global averages published by UNSCEAR 1993 and 2000 in Appendix B [18, 20] for ^{40}K and ^{232}Th . However, they remain below the exemption levels set by the IAEA in the Basic Safety Standard GSR part 3.

Concerning the sand collected in Cotonou, the specific activities of these three radionuclides (^{40}K , ^{238}U and ^{232}Th) are $(144 \pm 8) \text{ Bq.kg}^{-1}$, $(30 \pm 2) \text{ Bq.kg}^{-1}$ and $(56 \pm 7) \text{ Bq.kg}^{-1}$ respectively. The values obtained in Cotonou are lower than the global averages for ^{40}K and ^{238}U . It is significantly higher than the world average for thorium-232, which is 30 Bq.kg^{-1} . As mentioned in the report of UNSCEAR in 2008, the global averages and standard error soil concentrations are ^{40}K ($400 \pm 24) \text{ Bq.kg}^{-1}$, ^{238}U ($37 \pm 4) \text{ Bq.kg}^{-1}$ and ^{232}Th ($33 \pm 3) \text{ Bq.kg}^{-1}$ [20]. Our values obtained in the Cotonou sand are much lower than the averages used by UNSCEAR in its 2008 report to estimate the external absorbed dose rate induced by the three natural

radionuclides in the soil. . On the other hand, the three radionuclides are still below the IAEA's exemption levels [2]. From the point of view of health impact, the natural radionuclides in the Cotonou sand have normal levels of radioactivity.

The specific activities obtained are then compared with the results found in different materials at the global level which are presented in Table 4.

For ^{40}K , the average specific activity in the granites studied is significantly higher than those found in some countries of the world. This value is also higher than the world average in ceramic, which is 500 Bq.kg^{-1} [18]. While the activity obtained from the Cotonou sand is much lower than the values determined in the soils of Mexico, Turkey and Greece [26-28]. Similarly, it is also below those found in various building materials such as Egyptian granites, Namibian and Chinese marbles, and other materials [16, 17, 35]. On the other hand, the average value of the Cotonou sand is much higher than the results found in the marbles of Nigeria, Egypt and Kuwait.

For ^{238}U , the average specific activity in the granites in this study is lower than the average values found in soil from Mexico and building materials from Turkey. It is also lower than those obtained in the marbles of Namibia, Egypt and China. Whereas this average value is relatively similar to the results found in the soils of Turkey and Greece. Similarly, it is almost identical to the results obtained in Egyptian granite, the local building materials of Nigeria. On the other hand, the average value of the granites studied is significantly higher than those found in the sand of Lebanon, the building materials of Iran and Ghana, and the marbles of Nigeria, Kuwait and Brazil. As for the sand collected in Cotonou, the specific activity of ^{238}U found is relatively similar to that obtained in the soils of Turkey and the building materials of Nigeria. This average value is also relatively similar to the world average in soil, which is 35 Bq.kg^{-1} [18].

For ^{232}Th , the average specific activity in granites is relatively similar to the results found in building materials from Turkey and Nigeria, and marbles from

Namibia and China. On the other hand, this average value is much higher than the specific activities obtained in the soils of Mexico, Turkey and Greece. Similarly, it is vastly superior to those found in the local building materials of Iran and Ghana, granite from Egypt, and marbles from Nigeria, Egypt, Kuwait, and Brazil. As for the Cotonou sand, the specific activity of thorium-232 is relatively similar to soil samples from Mexico and granite from Turkey. Nevertheless, this average value is significantly higher than that of the world average for soil, which is 30 Bq.kg⁻¹ [18].

Table 4 : Extracts from the specific activities of three naturally occurring radionuclides from selected countries of the world

Country	Sample Type	Specific activity (Bq.kg ⁻¹)			Références
		⁴⁰ K	²³⁸ U	²³² Th	
Aldama, State of Chihuahua, Mexico	Ground	1031	50	61	[26] Colmeneros <i>et al.</i> , 2004
Kastamonu, Turkey	Ground	431	33	27	[27] Kam <i>et al.</i> , 2007
Ptolemais, Greece	Ground	496	42	36	[28] Papaefthymiou <i>et al.</i> , 2007
South Lebanon	Sand	n.d.	[4 - 61]	n.d.	[29] Kobeissi <i>et al.</i> , 2008
Cankiri, Turkey	Ground	357	18	22	[30] Kam <i>et al.</i> , 2012
Samsun, Turkey	Ground	341	31	22	[31] Damla <i>et al.</i> , 2012
Egypt	Granite	702	46	52	[17] Sidiq <i>et al.</i> , 2022
Iran	Local building materials	322 [29 - 1085] *	27 [7 - 44] *	23 [6 - 60] *	[15] Kovacs <i>et al.</i> , 2021
Turkey	Clay brick, marble and granite	9 - 1044	3 - 146	1 - 154	[14] Kurnaz <i>et al.</i> , 2021
Namibia	Marble	3 - 929	0,4 - 340	0,2 - 210	[16] Onjefu <i>et al.</i> , 2022
Nigeria	Marble	7	2	1	[32] Ademola <i>et al.</i> , 2008
Egypt	Marble	1	57	6	[33] Fares <i>et al.</i> , 2011
Kuwait	Marble	4	4	0,2	[34] Bou-rabee <i>et al.</i> , 1996
China	Marble	44 - 1353	8 - 157	6 - 166	[35] Lu <i>et al.</i> , 2007
Espirito Santo, Brazil	Marble	18	3	2	[36] Reginaldo <i>et al.</i> , 2011
Zaria, Nigeria	Local building materials	390	30	133	[37] Arabi <i>et al.</i> , 2015
Accra, Ghana	Local building materials	63 - 1222	3 - 47	4 - 43	[38] Otoo <i>et al.</i> , 2018
Ankara, Turkey	Building materials	2.0 - 1792	0.5 - 145	0.6 - 170	[39] Turhan <i>et al.</i> , 2008
Ramsar, Iran	Building materials	LD - 1350	LD - 86	LD - 187	[40] Bavarnegin <i>et al.</i> , 2013

* Average value [minimum value – maximum value]

3.2.2. Activities of Radium Equivalent

The activity of radium equivalent in granites ranges from (178 ± 10) Bq.kg⁻¹ to (424 ± 16) Bq.kg⁻¹ with a mean of (330 ± 74) Bq.kg⁻¹.

That of the Cotonou sand is (122 ± 10) Bq.kg⁻¹. The mean values found are below the recommended limit value of 370 Bq.kg⁻¹. This value indicates that the granites and sand studied do not present radiological risks for their use as construction materials.

Similar studies carried out in local building materials (sand, cement, gravel, gypsum and paint) in southern Lebanon show that the activities of equivalent radium vary from (9 ± 1) Bq.kg⁻¹ to (74 ± 9) Bq.kg⁻¹ [29]. In Namibia, the average value of equivalent radium in marble is 227 Bq.kg⁻¹ [16]. In Ghana, the activities of equivalent radium found in building materials range from 33 Bq.kg⁻¹ to 174 Bq.kg⁻¹ [38]. However, the results obtained by some of the above-mentioned countries in various building materials are all inferior to those found in the granites of the Communes of Glazoue and Dassa-zoume. Nevertheless, the activity of the equivalent radium obtained in the Cotonou sand is much lower than that found in the Ghanaian building material.

3.2.3. Gamma Index (I_γ)

According to the results of this work, the gamma I_γ indices calculated from the granites of the Communes of Glazoue and Dassa-zoume vary from 0.69 ± 0.03 to 1.57 ± 0.06 with a mean of 1.23 ± 0.26 . The Cotonou sand has a gamma index of 0.43 ± 0.03 . These results are lower than the gamma index I_γ found in Namibian marble of 1.59 [16].

Therefore, in accordance with the European Directive (EC), granites from the studied quarries increase the annual effective dose of 0.3 mSv.y⁻¹ to the population that uses these granites as building materials. According to this directive, these granites fall under the exemption level for building materials and can be used without strict control of natural radioactivity [19].

3.2.4. Absorbed dose rate in air and annual effective dose

For external exposure doses, the absorbed dose rate in air (\dot{D}) estimated within a dwelling built with granites from the Communes of Glazoue and Dassa-zoume can vary from 86.6 to 195.6 nGy.h⁻¹ with a mean of (153.4 ± 9.4) nGy.h⁻¹ whose the indoors external dose rate is 122.72 nGy.h⁻¹ (80%) and the outdoors dose rate is 30.68 nGy.h⁻¹ (20%). These results obtained with granite are well high the global indoors value of 84 nGy.h⁻¹ and well below the global outdoors value which is 59 nGy.h⁻¹ [18]. This confirms that the granite studied presented a danger for its uses.

As for the sand sample taken in Cotonou, the dose rate can reach (53.7 nGy.h⁻¹). This external dose rate is well below all global values indicated in the report of UNSCEAR in 2000. This confirms that the sand studied does not present a danger for its uses. Compared to other global studies, these dose rates obtained with granite sample are also higher those found in Namibian marble of 103 nGy.h⁻¹ [16] and local Ghanaian building materials ranging from 9.5 to 76.3 nGy.h⁻¹ [38]. Regarding the annual external effective dose for 80% building occupancy factor, the estimated values are (0.26 ± 0.01) mSv.y⁻¹ and (0.07 ± 0.01) mSv.y⁻¹ for the sand of Cotonou respectively for indoors and outdoors. This outdoor external effective dose is similar to the outdoor external effective dose (0.07 mSv.y⁻¹) estimated by UNSCEAR in his report in 2000. This indoor external effective dose is well below the indoor effective dose (0.41 mSv.y⁻¹) for UNSCEAR in his report in 2000. For 60% building occupancy factor, the annual external effective doses induced by the sand of Cotonou are (0.20 ± 0.01) mSv.y⁻¹ and (0.13 ± 0.01) mSv.y⁻¹ respectively for indoors and outdoors. These results are well below the indoor and outdoor effective dose estimated by UNSCEAR (0.33 mSv.y⁻¹ < 0.48 mSv.y⁻¹) in his report in 2000. These results confirmed that the sand of Cotonou does not represent a danger to the users.

For a building occupancy factor of 80%, the annual indoors external effective doses induced by the granite of Dassa-zoume and Glazoue varies from (0.42 ± 0.05)

mSv.y⁻¹ to (0.96 ± 0.1) mSv.y⁻¹ with a mean of (0.75 ± 0.05) mSv.y⁻¹. The annual external effective dose outdoors ranges from (0.11 ± 0.01) mSv.y⁻¹ to (0.24 ± 0.03) mSv.y⁻¹ with a mean of (0.19 ± 0.01) mSv.y⁻¹. These results for indoors and outdoors annual effective dose are well high the dose determined in the soils of Cankiri in Turkey, which is 44.4 μ Sv.y⁻¹ [30]. The results of the present study are still high the average of the annual effective dose for external exposure due to terrestrial ionizing radiation inside the building, which is 0.41 mSv.y⁻¹ indoors and 0.07 mSv.y⁻¹ outdoors [18], for a building occupancy factor of 80%, in report of UNSCEAR 2000. Considering a building occupancy factor in Sub-Saharan Africa which is 60%, the annual external effective dose indoors ranges from (0.32 ± 0.02) mSv.y⁻¹ to (0.72 ± 0.21) mSv.y⁻¹ with a mean of (0.56 ± 0.03) mSv.y⁻¹. The external annual effective dose outdoors ranges from (0.21 ± 0.03) mSv.y⁻¹ to (0.48 ± 0.08) mSv.y⁻¹ with a mean of (0.38 ± 0.02) mSv.y⁻¹. These results are higher than the annual effective dose estimated by UNSCEAR in his report in 2000 for indoors effective dose (0.41 mSv.y⁻¹) and outdoors effective dose (0.07 mSv.y⁻¹) for a building occupancy factor of 80%. These results confirmed that the granite represented a danger for its users.

For the children and infants, the annual effective dose induced by the granite to the representative public person is also higher the annual effective dose induced by the granite to adults and the annual effective dose estimated by UNSCEAR in his report in 2000.

3.2.5. Excess lifetime cancer risk (ELCR)

According to the results, for adults, the excess lifetime risk of cancer is $(1.1 \pm 0.02)E^{-3}$ for sand of Cotonou and varies from $(1.8 \pm 0.02)E^{-3}$ to $(4.0 \pm 0.03)E^{-3}$ with a mean of $(3.1 \pm 0.12)E^{-3}$ for granites. The value found from the Cotonou sand is high that of the world average of 2.9E-4 reported by UNSCEAR 2000. While the values estimated from the granites of the Communes of Glazoue and Dassa-zoume present values significantly higher than that of the world average.

Similarly, they are higher than the results found for the various Malaysian building materials which are $(0.42 \pm 0.24)E-3$, $(3.22 \pm 1.83)E-3$ and $(3.65 \pm 1.85)E-3$ for the exterior and interior of buildings, and the total value of this risk [41]. Another similar study was conducted on local building materials in Ghana and showed that the excess lifetime risk of cancer ranged from $0.04E-3$ to $0.33E-3$ [38]. The author mentioned that only the gneissic rocks in the environment recorded lifetime excess cancer risk values of $0.32E-3$ and $0.33E-3$. Our results in the granite are well high that this values from Ghana in the gneissic rocks. Our results show that the granite presented a danger for its users.

Shashikumar et al evaluated indoor and outdoor effective dose and ELCR from gamma dose rates in and around Mandya District, Karnataka in 2022. This study had shown that the Effective doses indoors and outdoors range from 0.32 to 0.65 mSv per year, with a geometric mean value of 0.48 mSv per year, and from 0.06 to 0.10 mSv per year, with a geometric mean value of 0.07 mSv per year, which is slightly higher than the global average for effective doses. The lifetime excess cancer risk (ELCR) indoors and outdoors of residents along the various locations ranges from $1.14E-3$ to $2.26E-3$ with a geometric mean value of $1.68E-3$ and $0.20E-3$ to $0.35E-3$ with a geometric mean value of $0.25E-3$, which is similar to the global ELCR average [42]. Our results in the granite are well high that this values from Karnataka. But this estimate was made on the basis of the dose rates at 1 m from the ground using an ER-709 lightweight portable radiation dosimeter. On the other hand, in our work, our estimates were made on the basis of the activity concentrations of the radionuclides present in the granite samples. The difference observed can be justified by the two different techniques for estimating effective doses and the ELCR.

4. CONCLUSIONS

This work made it possible to study twelve (12) samples of crushed granite taken from the artisanal quarries of the Communes of Glazoue and Dassa-zoume, and one (01) sample of sand taken in the Commune of Cotonou. At the INSTN-Madagascar laboratory, these samples were analyzed by gamma spectrometry equipped with an Ortec 3"x3" NaI(Tl) detector.

The results obtained were then compared with the global average values (GMVs) provided in UNSCEAR 2000 publication, the limit values set by the IAEA and those available in other countries of the world. The associated radiological parameters and indices (R_{eq} , I_{γ} , \dot{D} and E) were calculated and also compared with the limits prescribed by the radiation protection commissions and organizations (WHO, ICRP, UNSCEAR, IAEA and EC) in order to ensure the safe use of the granites and sand studied.

The study revealed that the specific activities of natural radionuclides (^{40}K , ^{238}U and ^{232}Th) in the granites of the Communes of Dassa-zoume and Glazoue are well below the exemption levels set by the IAEA, but they are generally higher than the world average values reported by UNSCEAR 2000, which is contrary to the results obtained in the sands of Cotonou which has low levels of radioactivity. For the adults, the stochastic risk prediction parameters for granites (R_{eq} , I_{γ} , \dot{D} , E for 80% and 60% and ELCR) are $(330 \pm 74) \text{ Bq.kg}^{-1}$, (1.2 ± 0.3) , $(153.4 \pm 9.4) \text{ nGy.h}^{-1}$, $(0.75 \pm 0.05) \text{ mSv.y}^{-1}$ for indoors and $(0.19 \pm 0.01) \text{ mSv.y}^{-1}$ for outdoors for 80% building occupancy factor, $(0.56 \pm 0.03) \text{ mSv.y}^{-1}$ for indoors and $(0.38 \pm 0.02) \text{ mSv.y}^{-1}$ for outdoors for 60% building occupancy factor and $(3.1 \pm 0.01) \cdot 10^{-3}$ respectively. For the children, for a building occupancy factor of 80%, the average E are $(0.86 \pm 0.05) \text{ mSv.y}^{-1}$ and $(0.21 \pm 0.01) \text{ mSv.y}^{-1}$ respectively for indoors and outdoors. For infants, the E are $(1.00 \pm 0.06) \text{ mSv.y}^{-1}$ and $(0.25 \pm 0.01) \text{ mSv.y}^{-1}$ respectively for indoors and outdoors. For

children, for a building occupancy factor of 60%, the average E are (0.64 ± 0.03) mSv.y⁻¹ and (0.43 ± 0.02) mSv.y⁻¹ respectively indoors and outdoors. For infants, for a building occupancy factor of 60%, the E are (0.75 ± 0.05) mSv.y⁻¹ and (0.50 ± 0.02) mSv.y⁻¹ respectively indoors and outdoors. For the children and infants, the average ELCR $(3.5 \pm 0.001)E-3$ and $(4.1 \pm 0.01)E-3$ respectively.

For children and infants, for a building occupancy factor of 80%, these E are induced by the sand of Cotonou are (0.30 ± 0.01) mSv.y⁻¹ and (0.08 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors, (0.35 ± 0.01) mSv.y⁻¹ and (0.09 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors. For a building occupancy factor of 60%, these E induced by the sand of Cotonou are (0.23 ± 0.01) mSv.y⁻¹ and (0.15 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors, (0.26 ± 0.01) mSv.y⁻¹ and (0.18 ± 0.01) mSv.y⁻¹ respectively indoors and outdoors. The ELCR estimated is equal $(1.2 \pm 0.01)E-3$ and $(1.4 \pm 0.01)E-3$ respectively children and infants.

They are below the reference values 370 Bq.kg⁻¹, 2, 84 nGy.h⁻¹, 0.41 mSv.y⁻¹ for indoors, 0.07 mSv.y⁻¹ for outdoors for 80% building occupancy and 0.29E-3 respectively (UNSCEAR 2000 & 2008). Statistical analyses showed a p-value $p < 0.001$ for the external absorbed dose rate, the annual external effective dose and the excess Lifetime Cancer risk induced by granites. These results obtained indicate that granites in the Communes of Dassa-zoume and Glazoue are not safe for all age categories (adults, children and infants) and do require specific radiation protection measurements in order to use them as construction materials for buildings and social housing. But for the sand of Cotonou is safe and not require radiation protection measures.

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CONFLICT OF INTEREST

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