Health Risk Assessment and Statistical Analysis of Natural Radioactivity in the Uranium Exploration Area of Zabili, Chad

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Health Risk Assessment and Statistical Analysis of Natural Radioactivity in the Uranium Exploration Area of Zabili, Chad

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Abstract

The activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in nineteen soil samples from Zabili uranium exploration area were measured using a low-background digital gamma-ray spectrometer equipped with broad energy germanium detector. The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K range from 16.5 to 1110.9 Bq.kg^{-1}, 19.45 to 76.97 Bq.kg^{-1}, and 3.28 to 839.5Bq.kg^{-1}, with their mean values of 478.0, 58.9, and 562.5, respectively. In addition, radiological hazard parameters from the activity concentrations obtained were assessed by estimating radium equivalent activity, external and internal hazard index, indoor and outdoor absorbed gamma dose rate and the corresponding annual effective dose, effective dose rate to different body organs and tissues, and excess lifetime cancer risk. Derived radiological hazard indices have been revised in accordance with relevant national and international legislation and guidelines. The radiological hazard parameter values were found to be above the relevant limit values for soils. In order to investigate the distribution of radionuclides and associated health hazard parameters in the study area, a statistical study was performed.

Key-words: Natural radionuclides, gamma-ray spectrometry, activity concentrations, radiological hazards parameters, soils, statistical analysis.

1. Introduction

The richness and endemism of the mining resources of the Mayo-Kebbi West region of Chad are known for their originality. For more than a decade, mining has become a major activity in this region and constitutes a source of income for a significant number of populations while raising many hopes for development. Unfortunately, the exploitation of these mining resources generally remains without any significant impact on the local population and constitutes moreover a source of environmental degradation (IAEA, 2006; UNSCEAR, 2000). Therefore, behind the exploitation of these mining resources, sites remain potentially contaminated by various radioactive substances. People who frequent and feed on these resources are exposed to the chemical and radiological toxicity of the radionuclides contained in the residues generated by the exploitation of these ore deposits.

Several previous investigations have revealed the existence of radioactive elements and toxic heavy metals in this region (Oyamta, 2013; Penabei et al., 2018; Ajani et al., 2020; Ajani et al., 2022, Olivier et al., 2021). A prior work of ours (Penabei et al., 2018) reported on the assessment of natural radioactivity levels and associated radiological risks in building materials in the vicinity of mining and exploration sites in the Mayo-Kebbi region. The findings of this assessment show a very high level of ^{238}U activity in the soil brick samples from Zabili. This high level of radioactivity demonstrates that soil pollution resulting from environmental conditions generated by mining activities remains a very serious problem in the Zabili localities, in view of the specificity of these highly toxic, radioactive ores and the very harmful effects they produce on the environment and health in particular.

According to many sources consulted, there have been exploration activities for radioactive minerals for radium and uranium production, which were started in the 1977s by the UNDP (Oyamta, 2013). In 2007, airborne research was undertaken by the South African company Global Blue Marine, first in the Léré region, specifically in Madajang - Zabili over an area of 193 km², then in Gamboké in the Pala region over an area of 333 km² (Claudia Frank & Lena Guesnet). This exploration was continued in 2008 in the same areas and over a larger area (841 km²) by Signet Mining Services Ltd (SMS), a European-based mining company known in Chad as Chad Mining Services. The company's exploration activities included airborne geophysical surveys, geological surveys, surface radiometric surveys, and most importantly, it completed over 170 vertical (18,541 m) and core (2,676 m) percussion drill holes, 22 trenches, and a dozen inclined drill holes (Oyamta, 2013; Claudia Frank & Lena Guesnet). Of note, piles of uranium mining products have been left on the site and exposed to the elements for many years. Yet, after rainfall, surface runoff as well as acid mine drainage can lead to dispersion of waste and contamination of surrounding areas (IAEA, 2002; IAEA, 2006; Pereira et al., 2014; Carvalho et al., 2016).

Dispersal of these substances and the non-restoration of soils after extraction processes generally lead to the spread of naturally occurring radioactive material (NORM) contaminating

the environment, resulting in potential exposure to radioactivity of the public (Innocent, Onimisi, & Jonah, 2013). Exposure to ionizing radiation from sources of radioactivity in the environment represents one of the major health risks to humans. As a result, DNA damage can occur and be the cause of various cancers in the long term. The main radionuclides responsible for human exposure are uranium (^{238}U), uranium (^{235}U), thorium (^{232}Th), their decay products and potassium (^{40}K). These four radionuclides each have a very long half-life and have been present on Earth since its formation. In the uranium (^{238}U) decay chain, radium 226Ra is one of the most radiologically important radionuclides and, therefore, ^{226}Ra is often referred to instead of ^{238}U. Radium (^{226}Ra) with a half-life of 1600 years produces radon (^{222}Rn) with a half-life of 3.82 days when it decays to an alpha ionizing particle. Radon (^{222}Rn) is a radioactive gas that is ubiquitous on the surface of the Earth because its half-life (3.82 days) is long enough to allow it to migrate in the soil, from the rock that gave it birth, to the atmosphere. Inhalation of radon and its progeny (example, Polonium-218 with a half-life of 3.05 minutes) is the leading cause of exposure among natural sources of ionizing radiation (Ademola et al., 2014). When radon is inhaled, the alpha particle dose is delivered directly to bronchial tissue, creating the potential for radiogenic lung cancer. Due to the health risks associated with exposure to naturally occurring radioactive materials and inhalation of radon's short-lived decay products, international bodies and governmental organizations such as the International Commission on Radiological Protection (ICRP, 1991) and the Environmental Protection Agency (EPA, 2007) have adopted optimization measures to minimize this exposure.

In order to find exhaustive and quality information on this high level of uranium, thorium, and potassium concentration in soils and on the environmental pollution resulting from uranium prospecting activities in the Zabili and Madajang localities, a more thorough investigation was carried out. The study focused on the determination and distribution of natural radionuclides of the ^{238}U/^{232}Th decay series and ^{40}K in soil samples from these localities. For this purpose, soil samples were collected from randomly selected points, followed by laboratory analysis using the HPGe gamma detector. This detection method based on gamma spectrometry allows a qualitative and quantitative investigation of the different radionuclides present in the soil. With the aim of describing the statistical characteristics of primordial radionuclides concentrations and the associated health risk parameters, statistical studies (skewness, kurtosis and correlations) were performed. At the end of this study, the data on the levels of natural radioactivity and radiological exposure in the study area will be communicated to the competent authorities and subsequently, an awareness on the issue of NORM in the study area.

2. Materials and methods

2.1 Study area

The area under investigation is located in Léré (9°43'0" N, 14°6'0" E), one of the three regions the western Mayo-Kebbi region of Chad. The western Mayo-Kebbi region is rich in the diversity of its natural environments, where lakes, parks, forests and massifs alternate. Agriculture, livestock, fishing and handicrafts are the dominant livelihood strategies in the region. A more detailed description of the study areas was presented by (Oyamta, 2013; Penabei et al., 2018, Ajani et al., 2020). The first exploratory research on uranium was undertaken in 1977 by the UNDP in the Madajang and Léré regions (Claudia Frank & Lena Guesnet). In 2007, research by aerial photographs was undertaken by the South African company Global Blue Marine, first in the Léré region, more precisely in Madajang - Zabili

over an area of 193 km², then in Gamboké in the Pala region over an area of 333 km² (Claudia Frank & Lena Guesnet). More than 86 drillings with depths ranging from 36 to 130 m have been carried out in these areas. This research was continued in 2008 in the same areas and over a larger area (841 km^2) by Signet Mining Services Ltd (SMS), a European-based mining company known in Chad as Chad Mining Services. Its exploration activities included airborne geophysical surveys, geological surveys, surface radiometric surveys, and most importantly, it completed over 170 vertical (18,541 m) and core (2,676 m) percussion drill holes, 22 trenches, and a dozen inclined drill holes (Oyamta, 2013; Claudia Frank & Lena Guesnet). Mining activity was again observed in Madajang and Zabili (Léré) recently in 2021 by our research groups.

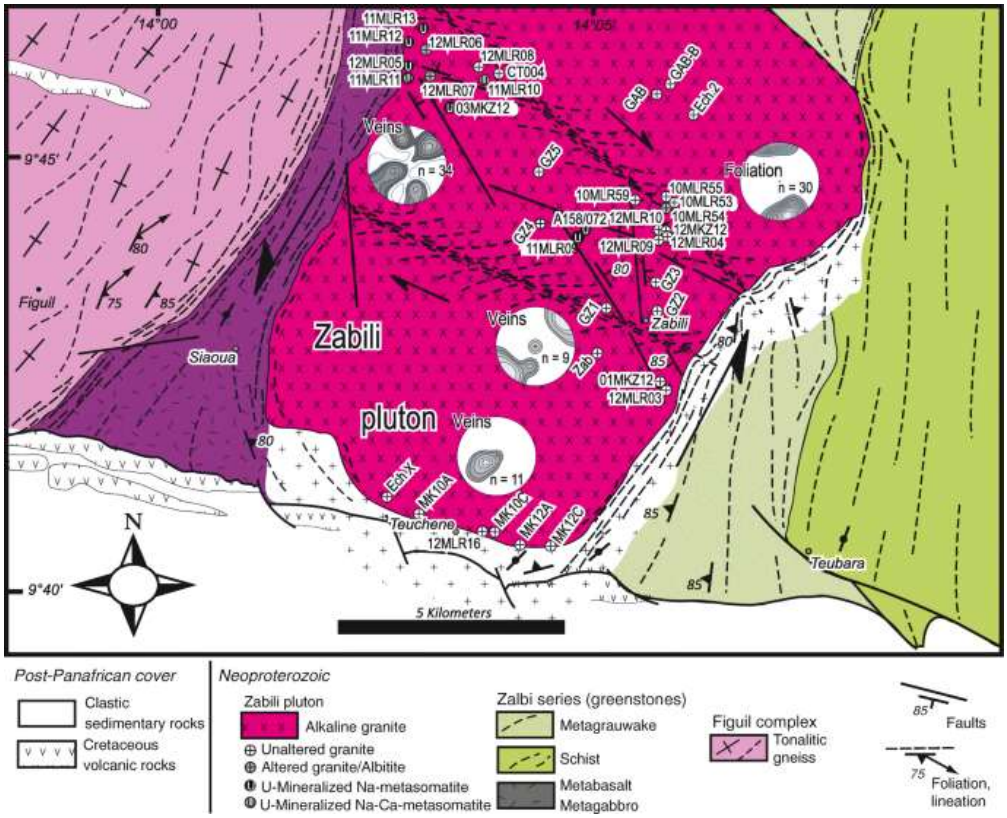

Figure: Geological map of Zabili (extracted from Olivier et al., 2021)

2.2 Sampling and preparation

To assess natural radioactivity levels, nineteen soil samples were randomly collected from the Zabili uranium exploration area to a depth of 2-10 cm from the top surface layer using a steel shovel. Each composite sample consisted of a mixture of five samples taken from a 5 m^2 area and separated from each other. Four samples were collected at the edges and one in the center. The spacing between composite samples was randomized to cover the study site and to observe significant local spatial variation in terrestrial radioactivity. Samples were packaged on-site in polyethylene bags and properly cataloged, labeled, and coded according to sample type and sample site location and transferred to the laboratory for preparation and analysis. In the laboratory, prior to analysis, the samples were ground to a powder, with a particle size of less

than 1 mm. Once this step was completed, the samples were oven dried at a temperature of 105°C for 48 hours to ensure that all significant moisture was removed from the samples. This step is necessary to accurately analyze the samples qualitatively and quantitatively. Dried samples were then packaged and hermetically sealed in 100 ml polyethylene plastic cylindrical containers to prevent the escape of ^{222}Rn gas and ^{220}Rn. All samples were weighed and stored for at least 4 weeks to reach secular equilibrium between the ^{238}U and ^{232}Th series and their progeny.

2.3 Radiometric analysis

Gamma-ray spectrometry technique was applied for the measurement of the radioactivity concentration of the samples under investigation. The spectrometry system consists of a high purity germanium (HPGe) detector (Canberra GC4520 coaxial p-type, 45% relative efficiency at 1332 keV, energy resolution of 2.2 keV FWHM at 1332 keV). The detector is maintained at liquid nitrogen temperature and surrounded by a 10 cm thick lead castle to reduce the background due to ambient natural radioactivity. Internal interface of the main lead shield is covered with 2 mm thick Cu plates to absorb X-rays coming from the main shield. The pulses delivered by the detector are amplified, adjusted and processed by a Digital Spectrum Analyzer (DSA) of Canberra brand. Data acquisition was performed on a PC equipped with GENIE 2000 software covering the energy range of gamma rays emitted between 50 and 3000 keV. Energy calibration was performed using a standard IAEA/RGTh reference source (3252 Bq for ^{232}Th) prepared in a 1-liter filled Marinelli beaker. Three reference materials IAEA/RGU-1 (4938 Bq for ^{238}U), IAEA/RGTh (3252 Bq for ^{232}Th), and KCl (13910 Bq for ^{40}K), were used for relative efficiency calibration of the spectrometer. Reference materials and samples were prepared in 100 ml pill bottles to maintain a similar counting geometry. Background count was determined by counting an empty Marinelli beaker for the same samples counting time and subtracting from the gross count. Each sample was counted for 43200 s (12 hours) to reduce statistical uncertainty.

2.3 Calculation of Activities

2.3.1 Activity concentrations of soil samples

Activity concentration (Bq.kg^{-1}) of the investigated radionuclides were calculated using the following equation (Penabei et al., 2018):

$$A_i = \frac{N_i}{\varepsilon(E)\cdot\gamma\cdot t\cdot m} \qquad (1)$$

Where, N_i is the net gamma count in a photo-peak (background corrected), $\varepsilon(E)$ the detector efficiency as function of gamma-ray energy, γ the number of gammas per disintegration of the given nuclide at energy E (the absolute transition probability of gamma-decay), m the sample mass (kg) and t the counting time (s).

2.3.2 Radiation hazards Indices and Dose Parameters

Consideration of the distribution of naturally occurring radionuclides provides an understanding of the radiological implications of these elements on the exposure of the human body to gamma radiation and the irradiation of lung tissues by inhalation of radon and its progeny. In an effort to estimate the radiological impact of the studied soil radiation, several parameters were calculated in this work. Potential radiological hazards associated with soil samples were evaluated by calculating radium equivalent activity (Raeq), indoor absorbed dose rates (D_{in}) and outdoor absorbed dose rates (D_{out}), external hazard index (Hex), internal hazard index (H_{in}), annual effective dose equivalent (E), effective dose rate to different body organs and tissues, and excess lifetime cancer risk (ELCR). This method has been used in radiation studies to reach a better and safer conclusion for the environment and health status of an exposed individual.

2.3.2.1 Radium Equivalent Activity

Natural radionuclides in the investigated soil samples are not uniformly distributed. Therefore, a common radiological index has been introduced to assess the actual activity level of ^{226}Ra, ^{238}Th and ^{40}K in the samples and the radiation hazards associated with these radionuclides. This index is generally known as the radium equivalent activity, and has been calculated by the following equation (Beretka et al., 1985; Belivermis et al., 2010).

$$Ra_{eq} = AT_{Ra} + (1.43 \times AT_{Th}) + (0.077 \times AT_{K}) \quad (2)$$

where AT_{Ra} , AT_{Th} and AT_{K} are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The permissible maximum value of the radium equivalent activity is 370 Bq/kg (Krisiuk et al., 1971) which corresponds to an effective dose of 1 mSv for the general public (Ndour et al., 2020).

2.3.2.2 External and Internal Hazard Index (H_{ex} and H_{in})

Internal and external hazard indexes were calculated according to (Ajayi et al., 2009; Zubair et al., 2020):

$$H_{ex} = \frac{AT_{Ra}}{370} + \frac{AT_{Th}}{259} + \frac{AT_{K}}{4810} \leq 1 \quad (3)$$

$$H_{in} = \frac{AT_{Ra}}{185} + \frac{AT_{Th}}{259} + \frac{AT_{K}}{4810} \leq 1 \quad (4)$$

where AT_{Ra}, AT_{Th} and AT_{K} are the activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively. Internal and external hazard index value must be less than unity in order to keep the radiation hazard to be insignificant.

2.3.2.3 Absorbed Dose Rates

In order to measure any radiological hazard, radiation exposure from radionuclides in the soil can be determined based on many parameters. A connection between radioactivity concentrations of natural radionuclides and its exposure is known as the absorbed dose rate in the air at 1 metre above the ground surface. Average activity concentrations of ^{226}Ra (U), ^{232}Th, and ^{40}K (Bq.kg-1) in soil samples are used to calculate absorbed dose rate according to the following formula (Beck et al., 1972, Belivermis et al., 2010):

$$D_{out}\ (nGy.h^{-1}) = 0.043A_{Ra} + 0.666\ A_{Th} + 0.047\ A_K \qquad (5)$$

Where, D_{out} is the absorbed dose rate in the air and A values are the measured activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. According to (Ajani et al., 2020, Penabei et al., 2018) we calculate the Indoor absorbed dose rate based on the worldwide average gamma dose rate indoors of 1.4 times higher than outdoors:

$$D_{in}\ (nGy.h^{-1}) = 1.4\ D_{out} \qquad (6)$$

2.3.2.4 Annual Effective Dose rates

Absorbed dose rate in air at 1 meter above the ground surface does not directly provide the radiological risk to which an individual is exposed (Jibiri et al., 2027). Absorbed dose can be considered in terms of an annual effective dose rate of terrestrial outdoor gamma radiation that is converted from absorbed dose by taking into account two factors, namely the coefficient for converting absorbed dose in air to effective dose and the outdoor occupancy factor. Annual effective dose equivalent can be estimated using the following formula (UNSCEAR, 2000, Turhan et al., 2008; Al-Kharouf et al., 2008, Nada et al., 2009;):

$$E_D(nSv.y^{-1}) = (D_{out} \times OF_{out} + D_{in} \times OF_{in}) \times 8760\ h \times 0.7\ Sv/Gy \times 10^{-6} \qquad (7)$$

According to report given by UNSCEAR 2000 report (UNSCEAR, 2000), conversion coefficient from absorbed dose in air to effective dose received by adults is 0.7 Sv/Gy and the occupancy factor for indoor and outdoor is 0.8 and 0.2, respectively, i.e. the fraction of time spent indoors and outdoors is 0.8 and 0.2, respectively.

2.3.2.5 Excess Lifetime Cancer Risk (ELCR)

It is known that low doses of ionizing radiation can increase the risk of cancer. This risk becomes evident at doses above 100 mSv, but to a very small amount. Risk of cancer increases as the radiation dose increases. Excess lifetime cancer risk was calculated using the following equation (Ajani et al., 2020; Penabei et al., 2018):

$$ELCR = E_D \times DL \times RF \qquad (8)$$

where, E_D, DL, and RF are annual effective dose rate, duration of life (70 years) and risk factor (0.05 Sv^{-1}), respectively. For stochastic effects, ICRP 60 uses values of 0.05 for the public (Taskin et al., 2009).

2.3.2.6 Effective dose rate to different organs and tissues

Effective dose rate to different organs and tissues (E_{org}) was estimated using the following formula (Darwish et al., 2015; Ndour et al., 2020):

$$E_{org}(mSv.y^{-1}) = E \times CF$$

Table 1: Conversion coefficient CF for different organs or tissues (Ndour et al., 2020)

Tissue or organ	Averages values of CF
Bone marrow	0.69
Whole-body	0.68
Lung	0.64
Ovaries	0.58
Teste	0.82

3. Results and Discussion

3.1 Activity concentrations

Activity concentration results of natural radionuclides measured in the studied soil samples are presented in Figure 1. Radionuclides found in soil samples are radioisotopes from the ^{238}U, ^{232}Th decay chains and a non-series radionuclide, ^{40}K. Activity concentration results are determined using the gamma-ray line of ^{226}Ra, ^{214}Pb and ^{214}Bi for ^{238}U, while transitions from ^{212}Pb, ^{212}Bi, ^{228}Ac and ^{208}Tl are used for ^{232}Th. Activity values of ^{238}U, ^{232}Th, and ^{40}K range from 16.5 to 1110.9 Bq.kg^{-1}, 19.45 to 76.97 Bq.kg^{-1}, and 3.28 to 839.5Bq.kg^{-1}, with their mean values of 478.0, 58.9, and 562.5, respectively.

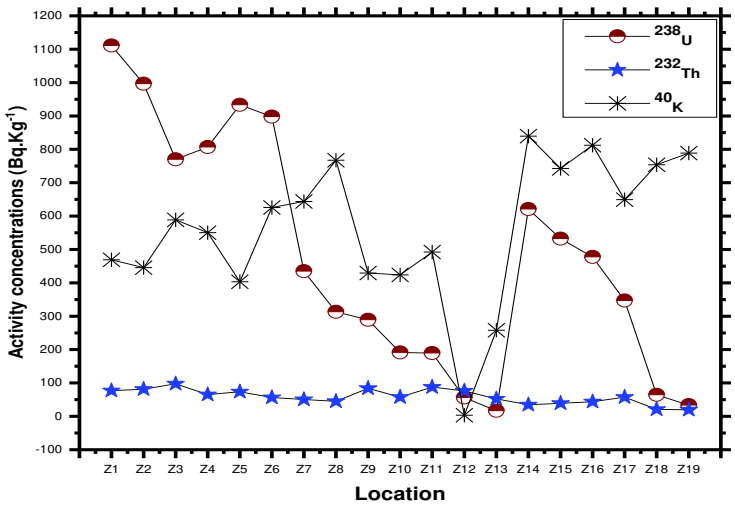

Figure 1: Activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soil samples.

Figure 1 shows that ^{238}U, ^{232Th} and ^{40}K concentration values vary from one sampling point to another, but the highest activities of the radionuclides ^{238}U, ^{232}Th were found in the Z1 sample while that of ^{40}K was found in the Z14 sample. It can also be seen that the ^{238}U concentrations in all samples are higher than the ^{232}Th concentrations, except for samples Z12 and Z13. This variation can be explained as being due to the different behavior of these radionuclides in soils. One can also observe that the activities of ^{40}K are higher in most cases than those of ^{238}U and ^{232}Th in the sampling sites. This is due to the presence of clay, which contains a relatively high concentration of potassium. In Figure 2 are shown the average activity concentration of ^{238}U, ^{232}Th and ^{40}K in the studied soil samples. From these results, it can be seen that the concentrations of ^{238}U, ^{232}Th and ^{40}K are very high with a negative trend of the sampling sites. When comparing our values to the world values, it is clear that the average activity concentration of ^{238}U, ^{232}Th, and ^{40}K in the analyzed soils is significantly higher than the world allowable values of 33, 30, and 400 $Bq.kg^{-1}$, respectively (UNSCEAR, 2000). These high values of radionuclide concentration reflect some influence of intensive exploration or mining activities in this area, confirming the work conducted by Penabei et al., 2018. This distribution indicates that the study sites are affected by releases to soil from exploration and mining activities and therefore could pose a significant risk to people living near the sites, farming in the area, or using the soils for construction materials.

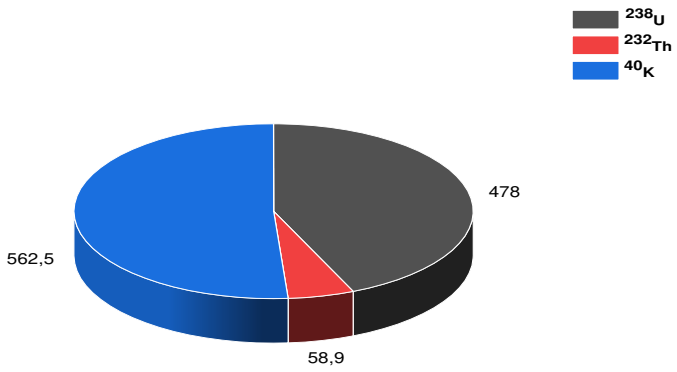

Figure 2: Average activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soil samples

3.2 Comparison of the average concentration of ^{238}U, ^{232}Th and ^{40}K measured in the samples with those obtained from other countries around the world

Tables 2, compares the reported values of the activity concentrations of radionuclides in this study with those obtained in other countries. As shown from this table, the measured average of ^{238}U in this work is well above the highest values reported in other countries. The ^{232}Th and ^{40}K values are comparable to these values. Observed discrepancies could be attributed to the difference in radionuclide distribution, probably related to the content of radioactive minerals and the geological, geochemical, and geographical origins of the raw materials, among other factors.

Table 2: Comparison of the average concentration of ^{238}U, ^{232}Th and ^{40}K measured in the samples with those obtained from other countries around the world

Country	Activity concentrations ($Bq.kg^{-1}$)			**References**
	^{226}Ra	**^{232}Th**	**^{40}K**	
Chad	477,99	58,93	562,46	This work
Kenya	33	45	420	(Osoro et al., 2011)
Nigeria	55,3	26,4	505,1	(Ademola et al., 2014)
Thailand	105,25	37,38	532,39	(Kessaratikoon et al., 2014)
India	48,07	230,77	807,08	(Raghavendra et al., 2019)
Sudan	7,54	20,74	111,87	(Idriss et al., 2016)
Iraq	77,33	9,36	426,31	(Al-Gazaly et al.,2014)
	35	30	400	**(UNSCEAR, 2000)**
Worldwide values				

3.3 Radiological hazard assessment

The calculated values of radium equivalent activity (Ra_{eq}), indoor absorbed dose rates (D_{in}) and outdoor absorbed dose rates (D_{out}) of all analyzed samples are given in **Figure 3**, and their average values are presented in columns 1- 3 of **Table 3**.

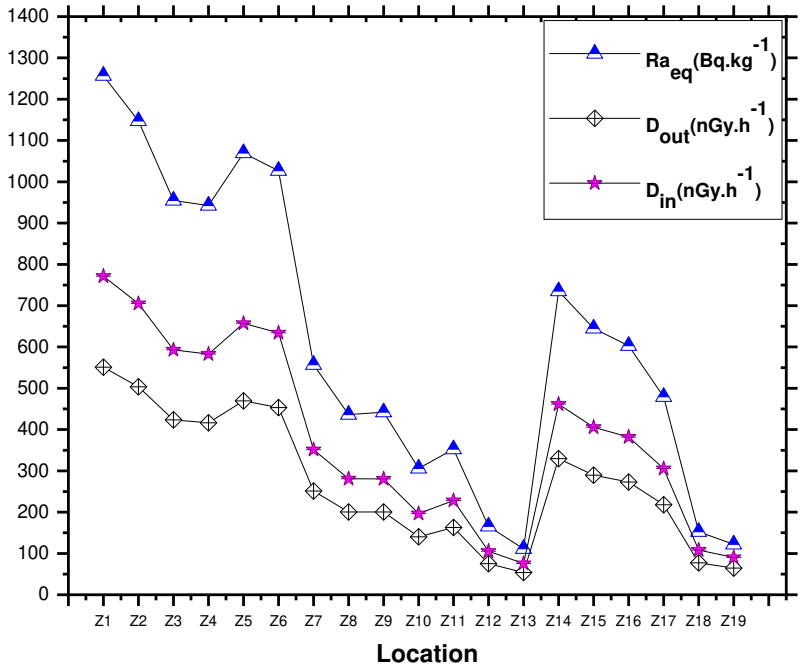

Figure 3: Concentration of radium equivalent activity (Ra_{eq}), absorbed dose rate outside (D_{out}) and inside (D_{in}) of the soil samples.

Radium equivalent concentration ranges from 111.16 to 1257.09 Bq.kg^{-1}, with mean value of 605.6 Bq kg^{-1}. From **Figure 3**, it can be seen that all soil samples except Z10 - Z13 and Z18 - Z19 have Ra_{eq} values above the maximum allowable value of 370 Bq.kg^{-1} (UNSCEAR 2000), which is equivalent to an annual external effective dose of 1 mSv.year^{-1} (OECD, 1979; EC, 1999). Comparatively to all investigated samples, Z1 sample has a highest Ra_{eq} value, yielding 3.4 times the world average value. **Figure 3**'s black curve shows that the outdoor absorbed dose rates range from 54.05 nGy.h^{-1} in the Z13 sample to 551.00 nGy.h^{-1} in the Z1 sample from Zabili, with a mean value of 271.2 nGy.h^{-1}. The world average value is 60 nGy.h^{-1}, which means that the average outdoor absorbed dose rates in the samples is 4.5 times the world average value. The purple curve in **Figure 3** illustrates that the indoor absorbed dose rates range from 75.68 nGy.h^{-1} in Z13 soil, to 771.40 nGy.h^{-1} in Z1 soil, with the average value of 379.7 nGy.h^{-1}. The population-weighted global average is 84 nGy.h^{-1} (UNSCEAR, 2000), implying that the average indoor absorbed dose rates in the Zabili soil samples is 4.5 times the world average value.

Figure 4 presents the calculated external hazard index (H_{ex}), internal hazard index (H_{in}), annual effective dose (E) and the excess lifetime cancer risk (ELCR) to the residents of the study area due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, and their average are presented in column 4 - 7 of **Table 3**.

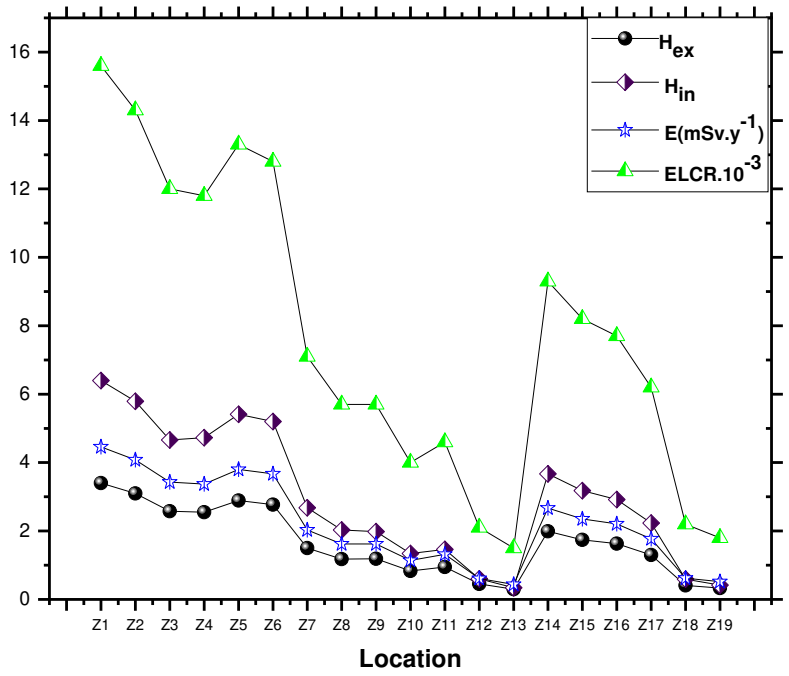

Figure 4: External hazard index (H_{ex}), internal hazard index (H_{in}), Annual effective dose equivalent (E) and Excess Lifetime Cancer Risk (ELCR) of the soil samples.

As results show, the external hazard index (H_{ex}) and internal hazard index (H_{in}) range from 0.30 to 3.40 and 0.34 to 6.40, with average values of 1.64 and 2.93, respectively. These average

values are higher than the world average value, indicating that the hazardous effects of radiation are not neglectful. From **Figure 4** and **Table 3**, the annual effective dose (E) ranges from 0.44 to 4.46 mSv y^{-1}, with an average value of 2.20 mSv y^{-1}, which is higher than the maximum value of 1 mSv y-1 recommended by (EC, 1999 and ICRP, 1990). The estimated excess lifetime cancer risk (ELCR) values range from 1.5 ×10-3 to 15.6 ×10-3 mSv y^{-1}, with a mean value of 7.7×10-3, which is approximately 27 times greater than the recommended value of 0.29× 10^{-3} (Ndour et al., 2020; Daulta et al., 2019). With such elevated ELCR values, there is a potential for radiation hazards, and the risk of developing cancer in people living in this environment is very high, so they should not be used in housing construction. Consequently, further studies are required to mitigate the exposure and, subsequently, the risk of developing cancer. Furthermore, attention should be paid by the government, the regulator, the mining companies in the study area and other stakeholders in order to provide a safe and radiologically clean environment for the population.

Shown in **Figure 5** are the annual effective dose rate results for different organs and tissues such as bone marrow, whole-body, lung, and reproductive organs such as ovaries and teste of the study area residents due to the specific activities of ^{238}U, ^{232}Th, and ^{40}K, and their average are presented in **Table 4**.

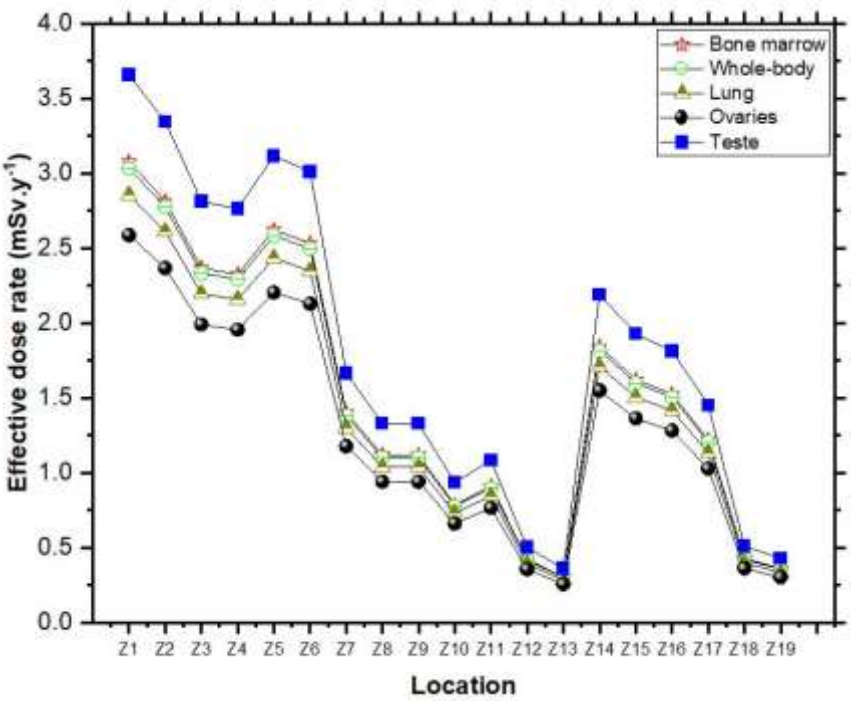

Figure 5: Effective dose rate to different organs and tissues

As illustrated in **Figure 5** and **Table 4**, the E_{org} values in bone marrow, whole-body, lung, ovaries, and testes ranged from 0.30 to 3.07, 0.29 to 3.03, 0.28 to 2.85, 0.26 to 2.59, and 0.36 to 3.66 mSv.y^{-1}, with mean values of 1.52, 1.50, 1.41, 1.28 and 1.80 mSv.y^{-1}, respectively. The estimated average E_{org} values in different types of organs and tissues were all more than twice

the set limit. The calculated average E_{org} values revealed that the testes were more sensitive to radiation compared with the ovaries.

Table 3: Average values of the calculated radiological parameters.

Location	Ra_{eq} (Bq.kg^{-1})	D_{out} (nGyh^{-1})	Din (nGyh^{-1})	Hex	Hin	E (mSv.y^{-1})	ELCR (mSv.y^{-1})
AVR ± STD	605.6 ± 360.5	271.2 ± 155.7	379.7 ± 218.0	1.64 ± 0.97	2.93 ± 1.91	2.20 ± 1.26	0.0077 ± 0.0044
Worldwide values (UNSCEAR, 2000)	**370**	**60**	**84**	**1**	**1**	**1**	**0.00029**

Table 4: Average effective dose rate to different organs and tissues

	Effective dose rate (mSv.y^{-1})				
	Bone marrow	**Whole-body**	**Lung**	**Ovaries**	**Teste**
Average	1.52	1.50	1.41	1.28	1.80
Worldwide values	**0.69**	**0.68**	**0.64**	**0.58**	**0.82**

3.4 Statistical analysis

Statistical studies were performed to describe and also to have a comprehensive understanding of the statistical characteristic of the detected primordial radionuclides concentrations levels and the associated health risk parameters. For these purposes, skewness, kurtosis and correlations coefficients was used in this study.

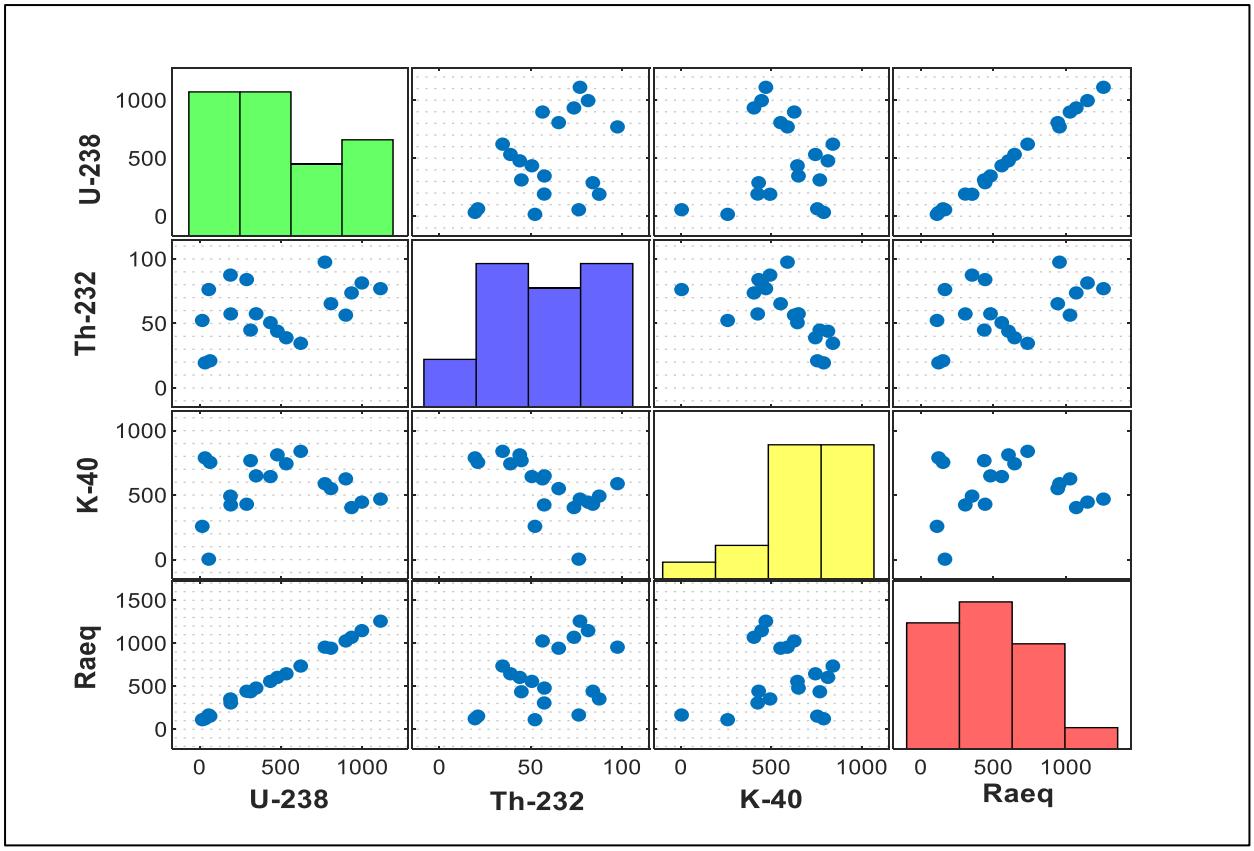

Figure 6: 2D frequency distribution of ^{238}U, ^{232}Th and ^{40}K and Radium equivalent activity concentrations.

The concept of skewness was related to the symmetrical distribution of radioactivity in soil samples. Once the skewness value is null, data related to activity concentrations can be assumed to be perfectly symmetrical. Skewness values varied from -0.85 to 0.29 (**Table 5**), which means that data are closed to be assumed as closely symmetrical distributed. Uranium and all health risk parameters depicted positive value of skewness. This means that uranium and health risk parameters are right skewed. Therefore, the right tail of the distribution is longer than the left one. Thorium and potassium activity concentrations display negative values of skewness, meaning that they are skewed left and the left tail is longer. These observations could be associated to sampling and samples preparation uncertainties (Glavič-Cindro et al., 2020). Despite the non-zero values of skewness, it can be assumed that data follows normal univariate distribution because they do not exceed the confidence normal distribution interval which varied from -2 to +2 (Westfall, 2014; Hazou et al., 2021).

Kurtosis in statistics is a measure of the combined weight of a particular distribution's tails regarding the center of the distribution. All data have a positive value of kurtosis which was higher than the unity. This observation indicates that radioactivity levels and health risk parameters distributions are too peaked. Health risk parameters measure the same value of kurtosis like that of activity concentration of uranium. Therefore, level of radiological exposure could be strongly correlated to that of uranium. Regarding kurtosis values which varied from 1.79 to 3.56, we can assume that all data displays normal distribution which was known in terms of interval to be -7 up to +7 (Westfall, 2014; Hazou et al., 2021).

Table 5: Skewness and Kurtosis of natural radionuclides and health risk parameters of soil samples from Zabili, Chad.

Natural radionuclides and health risk parameters																			
^{238}U		^{232}Th		^{40}K		Ra_{eq}		D_{out}		Din		Hex		Hin		E		ELCR	
S	K	S	K	S	K	S	K	S	K	S	K	S	K	S	K	S	K	S	K
0.29	1.79	-0.12	2.15	-0.85	3.56	0.25	1.81	0.23	1.8	0.23	1.8	0.26	1.81	0.27	1.8	0.23	1.8	0.23	1.8

S denote skewness and K denote kurtosis.

The strength between variables is known as correlation coefficients and was used to describe the degree of dependence among studied parameters. Combining results observed for skewness and kurtosis values, data was considered to be normal distributed in all soil samples. Once all assessed radioactivity parameters follow normal distribution, the most dedicate type of correlation which could described well enough link between data is Pearson's correlation (Westfall, 2014). Therefore, linear correlation coefficients obtained and presented in matrix view in **Table 6** was Pearson's type. All coefficients were assessed with their associate p-values. Health risk parameters correlate positively and strongly with activity concentrations of uranium in soil samples. The radiological exposition in Zabili area was mainly due to the level of radioactivity of uranium 238 and its decay products. **Table 6** denote a poor correlation between thorium, potassium and dedicated heath risk parameters. The only one moderate negative correlation value (-0.620) between activity concentrations of potassium and thorium could be explained by the fact that these two radionuclides have different chemical processes (Hazou et al., 2019). The probability to develop cancer after amount period spend in this area was strongly related to the uranium level, poorly due to thorium level and extremely poorly due to the level of potassium (Hazou & Patchali, 2021). Despite the potassium level being above the recommended limit of 400 Bq kg^{-1} (UNSCEAR, 2000), it doesn't contribute significantly to radiological exposure in human life. The level of potassium can't be a problem for population from this area because potassium is homeostatically regulated in human body (Hazou et al., 2019, 2021).

Table 6: Correlation matrix between activity concentrations of natural radionuclides and health risk parameters.

	^{238}U	^{232}Th	^{40}K	Ra_{eq}	D_{out}	Din	Hex	Hin	E	ELCR
^{238}U	1.000	0.400[b]	0.089[b]	0.998[a]	0.998[a]	0.998[a]	0.998[a]	0.999[a]	0.998[a]	0.998[a]
^{232}Th		1	-0.620[a]	0.442[b]	0.436[b]	0.436[b]	0.442[b]	0.421[b]	0.436[b]	0.437[b]
^{40}K			1	0.076[b]	0.090[b]	0.090[b]	0.076[b]	0.083[b]	0.089[b]	0.090[b]
Ra_{eq}				1	0.999[a]	0.999[a]	1.000[a]	0.999[a]	0.999[a]	0.999[a]
D_{out}					1	1.000[a]	0.999[a]	0.999[a]	1.000[a]	1.000[a]
Din						1	0.999[a]	0.999[a]	1.000[a]	1.000[a]
Hex							1	0.999[a]	0.999[a]	0.999[a]
Hin								1	0.999[a]	0.999[a]
E									1	1.000[a]

[a] Correlation is significant at the 0.01 level (2-tailed) and [b] Correlation is significant at the level higher than 0.05.

4. Conclusion

The ^{238}U, ^{232}Th, and ^{40}K activity concentrations were assessed in soil samples from the Zabili uranium exploration area by a high-purity γ-ray spectrometry system. Average activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in samples from the mine sites were estimated to be 478.0, 58.9, and 562.5, respectively, and were all above worldwide average values. Average values of the estimated radiological parameters in the soil samples were also found greater than the worldwide value. Average annual effective dose (E) was 2.2 times higher than the recommended worldwide average value. The calculated average ELCR was 27 times higher than its worldwide average value of 0.29×10^{-3}, which could serve as an alert to the radiation protection authority. Special attention should be paid by the government, regulator, mining companies in the study area and other stakeholders to provide a safe and radiologically clean environment for the population. Baseline statistics on the investigated soil samples from Zabili, such as skewness and kurtosis, showed that the radioactivity generated by the primordial gamma-ray emitting radionuclides could be considered a normal distribution. Pearson's correlation applied to all investigated parameters display strong positive coefficients between uranium activity concentrations and health risk parameters. The high level of associated radiological exposure was primarily related to uranium and its decay products, and this observation allows us to focus on the regulation of the level of uranium 238. The high level of uranium-238 calls for further investigation of radon in this area, as radon-222 represents a potential risk parameter and is a decay product of uranium-238.

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Conflict of interest statement

The authors confirm that there are no known conflicts of interest associated with this manuscript.

References

Ademola, A. K., Bello, A. K., & Adejumobi, A. C. (2014). Determination of natural radioactivity and hazard in soil samples in and around gold mining area in Itagunmodi, south-western,

Nigeria. Journal of Radiation research and applied sciences, 7(3), 249-255.

Ajani, M.B., Maleka, P.P., Usman, I.T. and Penabei, S. (2022). Health risk assessment of heavy metals concentration from soil; a case study of the Mayo-Dallah in Southern area of Chad. Journal of Radiation Research and Applied Sciences.

Ajani, M.B., Maleka, P.P., Usman, I.T. and Penabei, S.(2020). Assessment of environmental radiation exposure from soil radioactivity around the Southern area of Chad. Radiation Protection and Environment, 43(2), p.70.

Ajayi, O. S. (2009), "Measurement of Activity Concnetrations of 40K, 226Ra and 232Th for Assessment of Radiation Hazards from Soils of the Southwestern Region of Nigeria", Radiation and Environmental Biophysics 48, 323-332.

Al-Gazaly, H. H., Al-Ulum, M. A. B., Al-Hamidawi, A. A., & Al-Abbasi, A. M. (2014). Natural radioactivity in soil at regions around the uranium mine in Abu-Skhair Najaf Province, Iraq. Advances in Applied Science Research, 5(1), 13-17.

Al-Kharouf, S. J., Al-Hamarneh, I. F. and Dababneh, M. (2008), "Natural Radioactivity, Dose Assessment and Uranium Uptake by Agricultural Crops at Khan Al-Zabeeb, Jordan", Journal of Environmental Radioactivity 99 (7), 1192-1199.

Beck, H.L. (1972), "The physics of environmental radiation fields. Natural radiation environment II, CONF-720805 P2", Proceedings of the Second International Symposium on the Natural Radiation Environment.

Belivermis, M., Kikic, O., Cotuk, Y. and Topcuoglu, S. (2010), "The Effect of Physicochemical Properties on Gamma Emitting Natural Radionuclide Levels in the Soil Profile of Istanbul", Environmental Monitoring and Assessment 163, 15-26.

Beretka, J., & Matthew, P. J. (1985). Natural radioactivity of Australian building materials, industrial wastes and by-products. Health physics, 48(1), 87-95.

Carvalho, F. P., Oliveira, J. M., & Malta, M. (2016). Preliminary assessment of uranium mining legacy and environmental radioactivity levels in Sabugal region, Portugal. International Journal of Energy and Environmental Engineering, 7(4), 399-408.

Claudia Frank and Lena Guesnet. The Influence of Petroleum on Conflict Dynamics in Chad. Available from: https://www.files.ethz.ch/isn/112372/brief41.pdf

Darwish, D. A. E., Abul-Nasr, K. T. M., & El-Khayatt, A. M. (2015). The assessment of natural radioactivity and its associated radiological hazards and dose parameters in granite samples from South Sinai, Egypt. Journal of Radiation Research and Applied Sciences, 8(1), 17-25.

Daulta, R., Garg, V. K., & Singh, B. (2019). Natural radioactivity in soil, associated radiation exposure and cancer risk to population of Eastern Haryana, India. Journal of the geological society of India, 94(5), 525-532.

European Commission (EC) (1999) Report on Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials, Radiation Protection No. 112. Directorate-General Environment, Nuclear Safety and Civil Protection.

Glavič-Cindro D, Hazou E, Korun M, et al (2020) Measurement uncertainty arising from sampling of environmental samples. Appl Radiat Isot 156:. https://doi.org/10.1016/j.apradiso.2019.108978.

Hazou E, Guembou Shouop CJ, Nguelem Mekongtso EJ, et al (2019) Preliminary assessment of natural radioactivity and associated radiation hazards in a phosphate mining site in southern area of Togo. Radiat Detect Technol Methods 3:. https://doi.org/10.1007/s41605-018-0091-x

Hazou E, Patchali TE (2021) Assessment of Radiological Hazards in the Phosphate Mining Area of Kpogamé, Togo. Case Stud Chem Environ Eng 3:100077. https://doi.org/10.1016/j.cscee.2020.100077.

Hazou, E., Zorko, B., Dzagli, M. M., Haliba, E. M., Shouop, C. J. G., Moyo, M. N., & Tchakpele, P. K. (2021). Transfer from Soil to Grass and Statistical Analysis of Natural Occurring Radionuclides in Soil from Phosphate Mining and Processing Sites in Maritime Region of Togo.Available from: http://dx.doi.org/10.21203/rs.3.rs-160904/v1.

IAEA: Monitoring and surveillance of residues from the mining and milling of uranium and thorium. Safety Reports Series No 27. International Atomic Energy Agency, Vienna (2002). http:// www-pub.iaea.org/MTCD/publications/PDF/Pub1146_scr.pdf

IAEA: Release of sites from regulatory control on termination of practices. IAEA Safety Standards Series No. WS-G-5.1. International atomic Energy Agency, Vienna (2006). http://www-pub.iaea.org/MTCD/Publications/PDF/Pub1244_web.pdf

Idriss, H., Salih, I., Alaamer, A. S., Saleh, A., & Abdelgali, M. Y. (2016). Environmental-impact assessment of natural radioactivity around a traditional mining area in Al-Ibedia, Sudan. Archives of environmental contamination and toxicology, 70(4), 783-792.

Innocent, A. J., Onimisi, M. Y., & Jonah, S. A. (2013). Evaluation of naturally occurring radionuclide materials in soil samples collected from some mining sites in Zamfara State, Nigeria. British Journal of Applied Science & Technology, 3(4), 684-692.

Jibiri, N.N., Farai, I.P. and Alausa, S.K. (2007), "Estimation of Annual Effective Dose due to Natural Radioactive Elements in Ingestion of Foodstuffs in Tin Mining Area of Jos-Plateau, Nigeria", Journal of Environmental Radioactivity 94, 31-40.

Kessaratikoon, P., Jewawongsakul, J., Boonkrongcheep, R., & Pholthum, S. (2019, November). Radiological hazard assessment and excess lifetime cancer risk evaluation in surface soil samples collected from Ban Chang and Nikhom Phatthana districts in Rayong province, Thailand. In Journal of Physics: Conference Series (Vol. 1380, No. 1, p. 012104). IOP Publishing.

Krisiuk, E.M., S.I. Tarasov, V.P. Shamov, N.I. Shlak, E.P. Lisachenko and L.G. Gomslsky (1971). A Study of Radioactivity in Building Materials. Research Institute of Radiation Hygiene Leningrad.

Nada, A., Maksoud, T. M. A., Hosnia, M. A. El-Nagar, T. and Awad, S. (2009), "Distribution of Radionuclides in Soil Samples from a Petrified Wood Forest in El-Qattamia, Cairo, Egypt", Applied Radiation and Isotopes 67, 643-649.

Ndour, O., Thiandoume, C., Traore, A., Cagnat, X., Diouf, P. M., Ndeye, M., ... & Tidjani, A. (2020). Assessment of natural radioactivity and its radiological hazards in several types of cement used in Senegal. SN Applied Sciences, 2(12), 1-8.

NEA-OECD (1979) Exposure to radiation from natural radioactivity in building materials, Report by NEA Group of Experts, OECD, Paris.

Olivier, V., André-Mayer, A. S., Diondoh, M., Aurélien, E., Maryse, O., Moussa, I., ... & Marieke, V. L. (2021). Uranium mineralization associated with late magmatic ductile to brittle deformation and Na–Ca metasomatism of the Pan-African A-type Zabili syntectonic pluton (Mayo-Kebbi massif,

SW Chad). *Mineralium Deposita*, *56*(7), 1297-1319.

Osoro, M. K., Rathore, I. V., Mangala, M. J., & Mustapha, A. O. (2011). Radioactivity in surface soils around the proposed sites for titanium mining project in Kenya.

Oyamta B, Bayang D, Mianyo D. Study on Mineral and Petroleum Resources in Mayo Kebbi West Chad; 2013. Available from: https://www.peacere-sources.net/les/docs/publications/RapportEtudeRessourceminesMKO25-06-2013.pdf. [Last accessed on 2009 Jul 09].

Penabei S, Bongue D, Maleka P, Dlamini T, Sadou CJ, Guembou Shouop YI, et al (2018). Assessment of natural radioactivity levels and the associated radiological hazards in some building materials from Mayo-Kebbi region, Chad. Radioprotection.

Pereira, R., Barbosa, S., & Carvalho, F. P. (2014). Uranium mining in Portugal: a review of the environmental legacies of the largest mines and environmental and human health impacts. Environmental geochemistry and health, 36(2), 285-301.

Raghavendra, T., Vishwaprasad, K., Kalyani, G., Vijayalakshmi, T., Himabindu, V., Arunachalam, J., ... & Tripathi, R. M. (2019). Assessment of Natural Radioactivity in Soils around the Proposed Uranium Mining Site of Lambapur–Peddagattu and Seripally, India. Journal of the Geological Society of India, 93(2), 223-227.

Taskin, H., Karavus, M., Ay, P., Topuzoglu, A., Hidiroglu, S., & Karahan, G. (2009). Radionuclide concentrations in soil and lifetime cancer risk due to the gamma radioactivity in Kirklareli, Turkey. Journal of Environmental Radioactivity

Turhan, S. and Gundiz, L. (2008), "Determination of Specific Activity of ^{226}Ra, ^{232}Th and ^{40}K for Assessment of Radiation Hazards from Turkish Pumice Samples", Journal of Environmental Radioactivity 99, 332-342.

UNSCEAR (2000) Sources and Effects of Ionizing Radiation United Nations Scientific Committee on the Effects of Atomic Radiation. Report to the General Assembly with Scientific Annexes, New York, USA. p 111-125.

Westfall, Peter H. (2014) "Kurtosis as Peakedness, 1905–2014. R.I.P." The American Statistician 68(3): 191–195. Retrieved 15 May 2016 from http://www.ncbi.nlm.nih.gov/pmc/articles/PMC4321753/.

Zubair, M. (2020). Measurement of natural radioactivity in several sandy-loamy soil samples from Sijua, Dhanbad, India. Heliyon, 6(3), e03430.