Scientific African 13 (2021) e00902

Contents lists available at ScienceDirect

Scientific African

journal homepage: www.elsevier.com/locate/sciaf

Study of water radioactivity transfer from telluric origin in the Amber Mountain, Antsiranana, Madagascar ☆

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ARTICLE INFO

Article history:
Received 10 June 2020
Revised 19 July 2021
Accepted 26 July 2021

Editor: DR B Gyampoh

Keywords:
Madagascar
Amber Mountain
Radioactivity
Water
Soil
Transfer factors

ABSTRACT

Water supply to the extreme north of Madagascar comes from the Amber Mountain catchment. The objectives of this work are to study the presence of natural radionuclides in the waters from the mountain catchment and investigate their origin by determining the radioactivity levels in water and soil, and their transfer factors. Forty samples were collected from various locations at the study site including 13 of water and 27 of soils. The results for the water samples show that ^{40}K, ^{238}U series and ^{232}Th series activities vary, respectively from 2.7 to 19.8 $Bq.l^{-1}$, 1.7 to 8.2 $Bq.l^{-1}$ and 0.4 to 3.3 $Bq.l^{-1}$. For soil samples, the activities are in the range of 126 to 327 $Bq.kg^{-1}$, 14 to 73 $Bq.kg^{-1}$ and 10 to 402 $Bq.kg^{-1}$, respectively for the ^{40}K, the ^{238}U series and the ^{232}Th series. Regarding the radionuclide soil-water transfer factors, the mean values for the whole study site are 0.04 for ^{40}K, 0.10 for ^{238}U series and 0.03 for ^{232}Th series. These results show that the Amber Mountain catchment soil radionuclides migrate weakly to the water system and their activities vary widely from one place to another. This low level of migration is explained by the geochemical behavior and the physico-chemical properties of the radionuclides present in the area.

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Introduction

Madagascar is renowned for the existence of high level radioactive background in many parts of the country, due to its U/Th borne minerals richness. Many studies conducted, especially in the central, western and southern part of the country, have confirmed this finding. However, in spite of the fact that the northern region is inhabited by large population, studies related to this part of the country are scarce. The current study will contribute to fill the gap, making it possible to assess the exposure hazards in order to take preventive measures against the harmful consequences of ionizing radiation on human

☆ Edited By: Dr. B Gyampoh

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https://doi.org/10.1016/j.sciaf.2021.e00902
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health. As a first of its kind, it will also produce baseline data for future investigations. Data obtained during this study will help to initiate the environmental radioactivity mapping of the region.

The Amber Mountain, located in the northern part of Madagascar provides the main sources of water supply in the extreme north, especially Antsiranana City. In the central part of the mountain, at altitudes between 1000 m and 1475 m, is located the Andranomitehitehy spring and the Mahasarika Crater-Lake. In the eastern catchment is located the Besokatra river, which is one of the most important streams of the Amber Mountain catchment [1,3]. Natural radionuclides are ubiquitous in the earth crust, which infiltrate into groundwater and penetrate the water cycle. Natural radioactivity in the environment depends on geological properties and local geographical conditions and appears at different levels around the world [4]. In hydrological systems, water passes through various geological formations that contain essentially natural radioactive elements belonging to the ^{238}U series, the ^{232}Th series and ^{40}K [5]. The resulting drinking water contributes to the radiation dose of consumers, due to the aqueous radioactivity. Studies on the drinking water radioactivity have been carried out in the central region [6–8] and north-eastern region of Madagascar [9], asserting the presence of these radionuclides. The present work aims to study the presence of natural radionuclides in the waters from the Amber Mountain catchment and to explain their origin, by determining the radioactivity levels in water and soil, as well as their transfer factors.

This research is important as drinking water potentially affects human health, vitality and diseases. The natural radioactivity contents in drinking water used by the population can generate a potential radiological risk from ingestion. Prior to the current studies, natural radioactivity data in the area are scarce, if not unavailable. This will be the first time that radioactivity measurement to assess drinking water quality has been undertaken, using standard and accepted radioanalytical technique, i.e. gamma spectrometry. This research is also important as it is the first to be undertaken using this approach in the northern region of Madagascar.

Materials and methods

Description of the study sites

The study area, i.e. the Amber Mountain, is important as six lakes and several rivers and streams drain the mountain watershed, which supplies 50 million cubic meters of drinking water every year to the 130,000 inhabitants of Antsiranana City. Apart from the radioactivity level in soil and water, the soil-water transfer and migration information related to the geological radionuclides present in the area will allow the ionizing radiation exposure risk assessment through drinking water ingestion. Such results will support the decision-makers to mitigate the related health risks.

The Amber Mountain is located in Joffreville municipality, 45 km south-west of Antsiranana City, with geographical coordinates ranging from 12° 31′ S to 12° 44′ S (latitude) and from 49° 03′ E to 49° 13′ E (longitude). The geological constitution of the Amber massif is as follows:

- The top is formed by basalt ash in which the edge is comprised of basalts with basanitoids and limburgites. Some other rocks such as phonolites and tinguaites are associated with them. There are a large number of well-preserved craters, some of which are now lakes such as Mahery Lake, Mahasarika Lake, Grand Lake, etc. These are of the Hawaiian volcanic type associated with Strombolian projections which are the main origin of the massif.
- This volcanic region was achieved in four phases: (i) acid phase (Rhyolites and Trachytes, found in pebbles in the Coniacian and of probable Turonian age); (ii) anti-Aquitaine phase (eruptions in the Amber mountain); (iii) recent phase (erection of the Amber mountain) and (iv) very recent phase (creaction of cones, craters and flows).
- The volcanic rocks, mainly basaltic rocks, cover more than two-thirds of the region. The sedimentary rocks, mainly limestone, sandstone and marl, surround the entire periphery of the massif as far as Antsiranana City [2,10].

The soils vary from a ferruginous type of recent basalt to a ferrallitic type of old basalt. The soil surface is strongly acidic (pH = 4.5); the acidity decreases gradually with depth to a pH = 5.2 [2,3,10]. The soil profile in this area varies in depth. Between 0–10 cm, the soil is dark brown, very soft to touch, many roots, quite fine, intertwined, some plant breakage on the surface, silty and lumpy. Between 10–50 cm, it is dark brown, clay-silty, lumpy, with some fine and porous roots. Between 50–100 cm, the soil is dark yellow-brown, clay-silty, lumpy, with rare and porous roots. The clay contents vary between 30 to 40%. The proportion which is silt is between 15 and 30%. The fine fraction gradually decreases with depth and the sand fraction increases as one approaches the bedrock. In forests, the organic matter contents are particularly high at around 20 to 30%. The mean chemical composition of soil derived from basalts in the Amber Montagne is as follows: 48.92% of SiO_2, 16.16% of Al_2O_3, 3.72% of Fe_2O_3, 5.14% of FeO, 7.66% of MgO, 9.64% of CaO, 2.74% of Na_2O, 1.34% of K_2O, 0.94% of TiO_2, and 0.49% of P_2O_5 [2].

The extreme north of Madagascar has a tropical climate with two seasons. From November to March, the rainfall is abundant and sometimes intense, with temperatures varying from 25 to 32°C. From April to October, the season is dry with lower temperatures, from 19 to 27°C. Annual rainfall varies from 1200 to 3000 mm, from low altitudes to the mountain top. Most rivers are supplied by mountain spring water. The rugged reliefs and steep slopes generally result in torrential streams (Fig. 1a). The region is covered by a rainforest rich in mosses, lichens and orchids. Thick vegetation cover protects the soil from erosion phenomena [1,2].

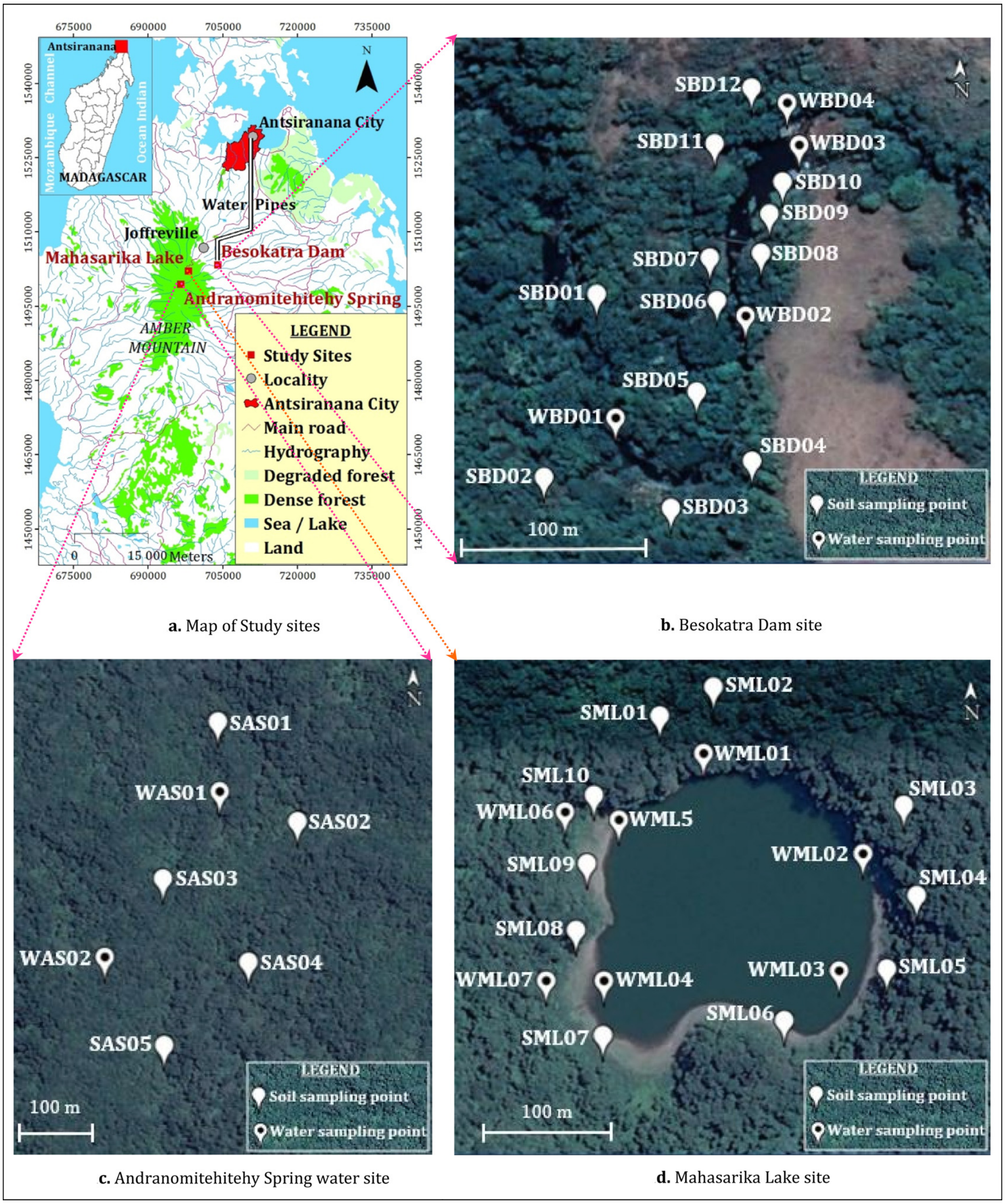

Fig. 1. Location of study sites and sampling points.

This region has been chosen for this study because of the particular sources of water supply used by the Antsiranana City population. Drinking water potentially affects human health, vitality and diseases; natural radioactivity data in the area is not currently available; and volcanic groundwaters are generally rich in natural radioactivity.

Sample collection and preparation

Field work was carried out in July 2016 at the Andranomitehitehy water spring, at Mahasarika Lake and at Besokatra Dam. At the Besokatra River and catchment, two water samples were collected upstream of the dam and two others were collected nearby (Ref. WBD01/.../04 in Fig. 1b). At the Andranomitehitehy site, upstream of the Besokatra River, two water springs were sampled (Ref. WAS01/02 in Fig. 1c). At Mahasarika Lake, which also supplies the Besokatra Dam, five water samples were collected (Ref. WML01/.../05) and two others were collected from surrounding water springs (Ref. WML06/07),

Table 1
Radionuclides used with their regions of interest.

Radionuclide	Energy (keV)	Energy range (keV)	Intensity [%]	Region of interest	Detection efficiency Cylindrical container (100 cm^3)	Marinelli beaker (1 L)
^{40}K	1461.0	1388.0 – 1534.1	10.67	(1)	0.0181 ± 0.0006	0.0146 ± 0.0005
^{238}U series	1764.5	1676.3– 1852.7	15.36	(2)	0.0168 ± 0.0001	0.0133 ± 0.0001
^{232}Th series	2614.5	2483.8 – 2745.2	99.16	(3)	0.0101 ± 0.0001	0.0067 ± 0.0001

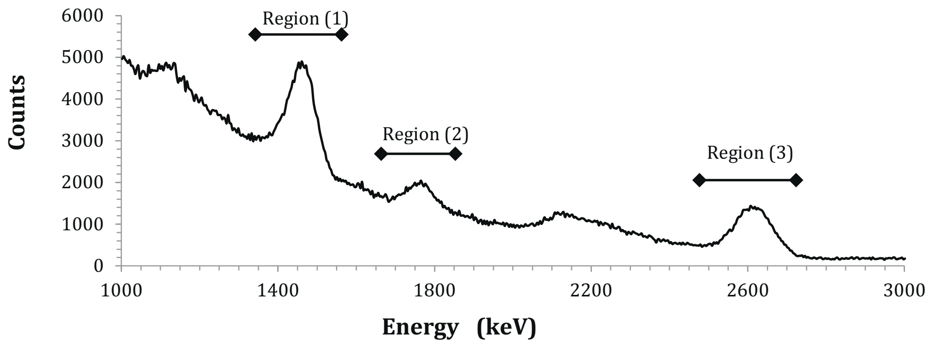

Fig. 2. Regions of interest of ^{40}K, ^{238}U and ^{232}Th series.

as shown in Fig. 1d. To investigate the radionuclide transfer rates, soil samples were taken near the water sampling points (Ref. SBD01/…/12, Ref. SAS01/…/05 and Ref. SML01/…/10 respectively, at Besokatra River, Andranomitehitehy spring and Mahasarika Lake). Each sampling point was localised using an Etrex Garmin GPS device. In all, 40 samples were collected: 13 water and 27 soil samples. Water samples were contained in 1.5 L plastic containers. The soil samples, c.a. 400 g wet mass each, were extracted up to 30 cm in depth, and contained in plastic bags. At the laboratory, the water samples were put directly into Marinelli beakers (1L). They were then sealed hermetically for three weeks to enable the contents to reach secular equilibrium between ^{226}Ra and its daughters before analyses. The soil samples were oven-dried at 80°C for 24 h, manually ground with a pestle mortar and sieved through 2 mm mesh. The samples were then homogenized and also sealed hermetically into 100 cm^3 cylindrical polyethylene containers for three weeks to reach secular equilibrium of the ^{238}U series [11].

Gamma spectrometry measurement

Sample analyses were performed using a gamma spectrometry system with a 3”x3” ORTEC NaI(Tl) detector, 905-4 series, at the department of Nuclear Analyses and Techniques, Institut National des Sciences et Techniques Nucléaires (INSTN-Madagascar). The detector energy resolution (FWHM) at 1332.5 keV (for the ^{60}Co peak) was 7.5%. The detector was placed inside a two-layered stainless steel (10 mm thick) and lead (30 mm thick) shield to minimize the background radiation, as well as the contribution of scattered radiation from the shield [12]. The soil samples were counted for 12 h and water samples for 24 h. Measurements of known masses of samples enabled determination of the specific activity of ^{40}K, ^{238}U series and ^{232}Th series. The method described by Rybach was used to process the spectra obtained [14,13]. This method targets three regions of interest corresponding to three isolated intense total absorption peaks of ^{40}K (1461 keV), ^{214}Bi (1764.5 keV) and ^{208}Tl (2614.5 keV), as shown in Table 1. The two last radionuclides are progenies of ^{238}U and ^{232}Th series, respectively [13,15].

Fig. 2 illustrates the three regions of interest of a soil or water gamma-ray spectrum.

Energy and efficiency calibrations were performed using certified IAEA (International Atomic Energy Agency) reference materials (RGK-1, RGU-1 and RGTh-1).

Specific activity and soil-water transfer factor

Each radionuclide specific activity was calculated from a sample γ-spectrum, using Eq. (1):

$$A = N_{net}/\left(\varepsilon \times P_{\gamma} \times t_c \times m\right) \quad (1)$$

where A (Bq.kg^{-1}) or (Bq.l^{-1}) is the specific activity for a given radionuclide, N_{net} (count) is the total absorption peak net area at energy E, ε is the detection efficiency at energy E, P_e is the gamma-ray emitter probability at energy E, t_c (s) is the measurement time and m (kg) is the analysed sample mass.

Table 2
Specific activities of ^{40}K, ^{238}U and ^{232}Th series in water samples from the Amber Mountain catchment.

Study sites	Sample reference	Specific activity ($Bq.l^{-1}$)* ^{40}K	^{238}U	^{232}Th
Besokatra River	WBD01	19.3 ± 1.3	2.4 ± 1.4	0.7 ± 0.3
	WBD02	2.7 ± 1.6	3.0 ± 0.2	0.4 ± 0.3
	WBD03	3.8 ± 2.2	8.2 ± 0.3	0.6 ± 0.3
	WBD04	3.9 ± 2.2	2.2 ± 1.3	0.6 ± 0.3
	Mean [min – max]	7.4 [2.7 - 19.3]	4.0 [2.2 – 8.2]	0.6 [0.4 – 0.7]
Andranomitehitehy Spring	WAS01	8,8 ± 1,1	2,2 ± 1,0	3,3 ± 0,3
	WAS02	3,8 ± 2,1	5,7 ± 0,3	0,6 ± 0,3
	Mean [min – max]	6.3 [3.8 – 8.8]	4.0 [2.2 – 5.7]	2.0 [0.6 – 3.3]
Mahasarika Lake	WML01	8,0 ± 2,2	4,9 ± 0,3	0,6 ± 0,3
	WML02	6,5 ± 3,7	3,3 ± 1,9	1,1 ± 0,6
	WML03	11,0 ± 1,0	1,7 ± 1,0	0,6 ± 0,3
	WML04	7,0 ± 2,3	2,1 ± 1,2	0,6 ± 0,3
	WML05	7,2 ± 1,1	5,2 ± 0,3	0,6 ± 0,4
	WML06	12,0 ± 1,1	5,0 ± 0,3	1,3 ± 0,3
	WML07	19,8 ± 1,1	4,3 ± 0,3	0,6 ± 0,3
	Mean [min – max]	10.2 [6.5 – 19.8]	3.8 [1.7 – 5.2]	0.8 [0.6 – 1.3]

* Value ± 1σ

However, the absolute absorption peak net area N_{net} was determined by subtracting the background total count from the sample at the same energy E [4,11]. In cases of different counting time between background and sample measurements, total count rate (count per second) was used.

The uncertainties for the ^{40}K, ^{238}U series and ^{232}Th series activities were calculated from the error propagation formula as shown in Eq. (2) [4,15].

$$\sigma_A = A \times \sqrt{(\sigma_{N_{net}}/N_{net})^2 + (\sigma_\varepsilon/\varepsilon)^2 + (\sigma_m/m)^2} \quad (2)$$

where $\sigma_{N_{net}}$, σ_ε and σ_m are the total absorption peak net area, the detection efficiency and analysed sample mass uncertainties, respectively.

The Minimum Detectable Activity (MDA) was determined by Eq. (3) [16].

$$MDA = \left(8.76 \times \sqrt{R_{BG} \times \Delta E}\right) / \left(\varepsilon \times P_\gamma \times t_c \times m\right) \quad (3)$$

where R_{BG} is the count rate from the background spectrum and ΔE is the full width at half of maximum.

The radioactivity of soils and rocks in contact with interstitial or surface waters contributes to the contamination of the latter. This phenomenon can be estimated by the radionuclide soil-water transfer factor (TF) given by the following Eq. (4) [17]:

$$TF = A_{water}/A_{sol} \quad (4)$$

where A_{water} ($Bq.l^{-1}$) is the water sample specific activity and A_{soil} ($Bq.kg^{-1}$) is the soil sample specific activity.

Results and discussions

Radioactivity of waters

The specific activities of the three radionuclides were determined from Amber Mountain catchment waters. The results are presented in Table 2. Extracts from worldwide results similar to the present study are shown in Table 3.

Comparing with the other results found in Madagascar and elsewhere in the world (see Table 3), the following observations are highlighted, considering the sites as a whole.

The average ^{40}K activity was slightly higher than the value obtained from mineral waters in Cameroon and in Romania, and from groundwater in Saudi Arabia [18,19]. On the other hand, it is comparable to the value found from drinking water in Ghana, from river water in Tanzania and from groundwater in Egypt [22–24]. The activity is lower compared to the results determined from hot spring water in Ethiopia, from river water in north-eastern Madagascar and in Nigeria, from mineral water in Bangladesh, from groundwater in Yemen and in Iraq, and from drinking water in Malaysia [9,25–30].

Regarding the ^{238}U series, the average activity obtained was comparable to the value found in Bangladesh, in Yemen, in Nigeria and in Malaysia [26–28,30]. However, on one hand, this value is much higher than the one obtained in Cameroon, in Romania, in Germany, in Saudi Arabia, in Ghana, in Tanzania and in Ethiopia [18–23,25], on the other hand, it is very low compared to the results determined in north-eastern Madagascar, in Egypt and in Iraq [9,24,29].

Regarding the ^{232}Th series, the average activity is relatively higher than the value found in Cameroon, in Romania, in Saudi Arabia, in Ghana, in Yemen and in Nigeria [18,19,21,22,27,28]. However, it is lower than the results obtained in north-eastern Madagascar, in Tanzania, in Egypt, in Ethiopia, in Bangladesh, in Iraq and in Malaysia [9,23–26,29,30].

Table 3
Extracts from worldwide water activities of the three radionuclides.

Country	Source type	Specific activity (Bq.l^{-1})* ^{40}K	^{238}U	^{232}Th	References
Madagascar	Spring, lake, river	8.8 [2.7 – 19.8]	3.9 [1.7 – 8.2]	0.9 [0.4 – 3.3]	Present work
Cameroon	Mineral water	0.11 [0.007 – 0.156]	0.022 [0.007 –0.038]	0.036 [0.005 – 0.121]	[18]
Romania	Mineral water	0.51 [0.14 – 0.87]	0.085 [0.050 – 0.095]	0.025 [MDA – 0.047]**	[19]
Germany	Drinking water	n.d.***	0.003 [< 0.001 – 0.320]	n.d.	[20]
SaudiArabia	Groundwater	4.58 [1.47 – 8.90]	0.56 [0.05 – 1.63]	0.20 [0.52 – 0.69]	[21]
Ghana	Borehole, river	7.76 [1.65 – 11.99]	0.54 [0.11 – 1.03]	0.41 [0.21 – 0.56]	[22]
Tanzania	River water	10.1 [9.2 – 11.0]	2.35 [2.2 – 2.5]	1.85 [1.8 – 1.9]	[23]
Egypt	Groundwater	10.3 [5.3 – 22.1]	45.6 [0.0 – 92.0]	2.9 [1.1 – 5.7]	[24]
Ethiopia	Hot spring water	17.70 [10.63 – 24.33]	2.30 [1.21 – 3.85]	1.70 [0.74 – 2.65]	[25]
Madagascar	River water	17.9 [6.9 – 36.2]	9.2 [0.6 – 19.5]	13.0 [3.4 – 17.7]	[9]
Bangladesh	Mineral water	18.3 [10.9 – 32.2]	3.3 [1.9 – 5.0]	6.4 [1.4 – 9.7]	[26]
Yemen	Groundwater	34.9 [26.73 – 43.70]	2.95 [2.25 – 3.45]	0.72 [0.30 – 1.37]	[27]
Nigeria	River water	120.45 [31.48 – 206.93]	5.49 [1.15 – 8.50]	0.14 [BDL – 0.51]****	[28]
Iraq	Drinking water	129.88 [6.38 – 253.86]	12.88 [1.40 – 55.79]	5.98 [0.59 – 11.95]	[29]
Malaysia	Drinking water	152 [53 – 222]	2.86 [0.55 – 8.64]	3.78 [0.70 – 7.03]	[30]

* Mean value [minimum value – maximum value].
** MDA: Minimum Detectable Activity.
*** n.d: not determined.
**** BDL: Below Detection Limit.

The differences in results compared to values from other regions can be explained by the interaction with geological and pedological formations in the study sites [31]. Reference studies indicate that basaltic rocks in contact with catchment waters can contain natural radioactivities around of 300 Bq.kg^{-1} for ^{40}K, 10 to 15 Bq.kg^{-1} for ^{238}U series and 7 to 10 Bq.kg^{-1} for ^{232}Th series [32]. In general, radioactivity is higher in mineral waters or when the water is in contact with radioactive minerals. It is increased in confined aquifer samples and diluted in open surface water. The results obtained reveal that the radioactivity levels of the three natural radionuclides are not uniformly distributed in the waters from the Amber Mountain catchment, the activities varying from one place to another.

Radioactivity of soils

Table 4 shows the activity values of ^{40}K, ^{238}U, and ^{232}Th series in soil samples collected nearby to the related water samples from the Amber Mountain catchment. Table 5 presents the extracts of similar study results.

Comparing the current study with similar studies presented in Table 5, considering the sites as a whole, allows us to make the following observations.

The ^{40}K average activity is higher than the values found from ferrallitic soil in Ewekoro Nigeria [33]. This activity is relatively similar to the results found from ferrallitic soil in Douala Bassa Cameroon [34]. On the other hand, it is lower than the worldwide average value and the results found from volcanic soil of Yemen, from basement and volcanic soil in Fongo-Tongo Cameroon, from volcano-sedimentary area in northern Madagascar, in Rize and Erzincan Provinces of Turkey, from clayey soil in Itagunmodi Nigeria and from ferruginous soil in Egypt [4,35–41].

It is noted that in the current study, the average activity for the ^{238}U series is higher than the world average value and the ones found from ferrallitic soils in Ewekoro Nigeria and in Douala Bassa Cameroon, from soil in Rize and Erzincan Provinces of Turkey [4,33,34,38,39]. This activity is relatively similar to the results obtained from basaltic rock terrains in Yemen, from clayey soil in Itagunmodi Nigeria and from ferruginous soil in Egypt [35,40,41]. On the other hand, it is lower than the values obtained from ferrallitic soil in Fongo-Tongo Cameroon and from a volcano-sedimentary area in northern Madagascar [36,37].

Regarding the ^{232}Th series in the current study, the mean activity is also higher than the world average value and the ones found from ferrallitic soils in Ewekoro Nigeria and in Douala Bassa Cameroon, from basaltic rock terrain in Yemen, from soils in Rize and in Erzincan Provinces of Turkey, from clayey soil in Itangunmodi Nigeria and from ferruginous soil in Egypt [4,33–35,38–41]. Nevertheless, this activity is lower than the results obtained from basaltic rock terrain in Fong-Tongo Cameroon and from a volcano-sedimentary area in northern Madagascar [36,37].

In addition, the fluctuation of radioactivity levels in soils from the Amber Mountain catchment can also be explained by the geological structures of the study area [40,42]. The regional geology of the northern part of Madagascar, in particular the Amber Mountain basaltic formation, is characterised by the rarity of uranium-thorium minerals. This study shows that the three radionuclide activities vary spatially in soils around the mountain catchment.

Table 4
Specific activities of ^{40}K, ^{238}U and ^{232}Th series in soil samples from the Amber Mountain catchments.

Study sites	Sample reference	Specific activity (Bq.kg^{-1})* ^{40}K	^{238}U	^{232}Th
Besokatra River	SBD01	253 ± 11	64 ± 3	123 ± 6
	SBD02	253 ± 12	73 ± 3	134 ± 7
	SBD03	287 ± 12	26 ± 2	61 ± 5
	SBD04	185 ± 9	71 ± 3	72 ± 5
	SBD05	183 ± 10	46 ± 2	82 ± 5
	SBD06	193 ± 10	48 ± 2	296 ± 10
	SBD07	175 ± 9	58 ± 2	402 ± 12
	SBD08	154 ± 9	59 ± 2	386 ± 12
	SBD09	202 ± 11	72 ± 3	63 ± 6
	SBD10	126 ± 9	30 ± 2	26 ± 3
	SBD11	182 ± 9	42 ± 2	80 ± 4
	SBD12	210 ± 10	45 ± 2	74 ± 5
	Mean [min – max]	200 [126 – 287]	53 [26 – 73]	150 [26 – 402]
Andranomitehitehy Spring	SAS01	321 ± 15	36 ± 3	68 ± 7
	SAS02	174 ± 10	40 ± 3	23 ± 5
	SAS03	221 ± 10	48 ± 2	26 ± 4
	SAS04	262 ± 12	43 ± 3	25 ± 5
	SAS05	176 ± 10	26 ± 2	38 ± 5
	Mean [min – max]	229 [174 – 312]	39 [26 – 48]	36 [23 – 68]
Mahasarika Lake	SML01	224 ± 12	39 ± 3	32 ± 5
	SML02	258 ± 13	22 ± 3	10 ± 5
	SML03	148 ± 10	45 ± 3	24 ± 5
	SML04	231 ± 12	35 ± 3	25 ± 5
	SML05	239 ± 12	38 ± 3	18 ± 5
	SML06	191 ± 9	14 ± 2	25 ± 4
	SML07	200 ± 11	44 ± 3	27 ± 5
	SML08	276 ± 13	32 ± 3	58 ± 5
	SML09	231 ± 12	43 ± 3	112 ± 6
	SML10	327 ± 14	42 ± 3	110 ± 6
	Mean [min – max]	233 [148 – 327]	35 [14 – 45]	44 [10 – 112]

* Value ± 1σ.

Table 5
Extracts from worldwide soil activities from the three radionuclides.

Country (city)	Type of soil	Specific activity (Bq.kg^{-1}) * ^{40}K	^{238}U	^{232}Th	References
Madagascar	Ferruginous/ferrallitic	218 [126 – 327]	44 [14 – 73]	90 [8 – 402]	Present work
Nigeria (Ewekoro)	Ferrallitic	16.5 [6.3-37.9]	8.1 [4.9-13.4]	8.3 [3.6-17.6]	[33]
Cameroon (Douala Bassa)	Ferrallitic	215.9 [47.4-271.8]	24.5 [21.9-27.7]	66.7 [52.6-78.9]	[34]
Yemen (Sana'a)	Clay	939 [505 - 1230]	48 [24 - 69]	42 [18 - 53]	[35]
Cameroon (Fongo-Tongo)	Ferrallitic	671 [136-1269]	99 [ND - 215] **	157 [59 - 272]	[36]
Madagascar (Antsiranana)	Ferruginous	313 [112 – 565]	139 [77 – 190]	126 [81 – 161]	[37]
Turkey (Rize)	Not mentioned	344.9 [35.7 – 913.8]	24.5 [7.4 – 79.8]	51.8 [9.5 – 170.8]	[38]
Turkey (Erzincan)	Not mentioned	281.9 [64.7 – 977.8]	8.9 [1 – 23]	11.4 [1.2 – 29.4]	[39]
Nigeria (Itagunmodi)	Clay	505.1 [200.5 – 901.2]	55.3 [18.5 – 90.3]	26.4 [12.5 – 52.4]	[40]
Egypt (El Sahu)	Ferruginous	672.3 [646.9-707.4]	44.6 [28.8-78.2]	58.6 [55.5-66.3]	[41]
World average		400 [140 – 850]	35 [16 – 110]	30 [11 – 64]	[4]

Soil-water transfer factors of natural radionuclides

Table 6 summarizes the estimated soil-water transfer factors for the three natural radionuclides from Amber Mountain catchment. The extracts from worldwide transfer factors are given in Table 7.

The results show that the average transfer rates of ^{40}K, ^{238}U series and ^{232}Th series from the study sites are 4%, 10% and 3%, respectively. Comparing to the results reported in Table 7, it is observed that:

- The soil-water transfer factors of 40K are relatively similar to the results found in river water around a gold mining site in Ghana, in thermal spring water in Ethiopia, in irrigation water in Yemen and in water from Qarun Lake in Egypt [22,25,43,44].
- The soil-water transfer factors of 238U series, are higher than values found in Ghana, in Ethiopia and in Yemen [22,25,43]. Conversely, it is low compared to the value found in South Africa and in Egypt [17,44].

Table 6
Soil-water Transfer Factors for ^{40}K, ^{238}U and ^{232}Th series in the Amber Mountain catchment.

Study sites	Transfer factor reference	Transfer factor value ^{40}K	^{238}U	^{232}Th
Besokatra River	TF-01	0,03	0,06	0,00
	TF-02	0,03	0,05	0,00
	TF-03	0,03	0,15	0,01
	TF-04	0,04	0,06	0,01
	TF-05	0,04	0,09	0,01
	TF-06	0,04	0,08	0,00
	TF-07	0,04	0,07	0,00
	TF-08	0,05	0,07	0,00
	TF-09	0,04	0,05	0,01
	TF-10	0,06	0,13	0,02
	TF-11	0,04	0,09	0,01
	TF-12	0,04	0,09	0,01
	Mean [min – max]	0.04 [0.03 – 0.06]	0.08 [0.05 – 0.15]	0.01 [0.00 – 0.02]
Andranomitehitehy Spring	TF-13	0,02	0,11	0,03
	TF-14	0,04	0,10	0,08
	TF-15	0,03	0,08	0,08
	TF-16	0,02	0,09	0,08
	TF-17	0,04	0,15	0,05
	Mean [min – max]	0.03 [0.02 – 0.04]	0.11 [0.08 – 0.15]	0.06 [0.03 – 0.08]
Mahasarika Lake	TF-18	0,05	0,10	0,02
	TF-19	0,04	0,17	0,10
	TF-20	0,07	0,08	0,03
	TF-21	0,04	0,11	0,03
	TF-22	0,04	0,10	0,04
	TF-23	0,05	0,27	0,03
	TF-24	0,05	0,09	0,03
	TF-25	0,04	0,12	0,01
	TF-26	0,04	0,09	0,01
	TF-27	0,03	0,09	0,01
	Mean [min – max]	0.05 [0.03 – 0.07]	0.12 [0.08 – 0.27]	0.03 [0.01 – 0.10]

Table 7
Extracts of soil-water transfer factors for ^{40}K, ^{238}U and ^{232}Th series in the world.

Country	Sample type	Average activity ^{40}K	^{238}U	^{232}Th	Transfer factor ^{40}K	^{238}U	^{232}Th	References
South Africa	water	n.d.	63.9	0.7	n.d.	0.49	0.04	[17]
	soil	n.d.	129	18.1				
Ghana	water	7.76	0.54	0.41	0.05	0.04	0.02	[22]
	soil	157	15	27				
Ethiopia	water	17.70	2.30	1.70	0.03	0.01	0.03	[25]
	soil	577	249	60				
Yemen	water	18.34	1.44	1.20	0.03	0.02	0.02	[43]
	soil	698	59	71				
Egypt	water	31.30	6.40	3.20	0.03	0.27	0.20	[44]
	soil	933	24	16				

n.d.: not determined.

- The soil-water transfer factors of 232Th series, are relatively similar to values found in South Africa, Ghana, Ethiopia and Yemen [17,22,25,43] but, it is on average 7 times lower than value obtained in Egypt [44].

The results illustrate that the soil-water transfer rates are relatively low, especially for ^{40}K and the ^{232}Th series. Regarding the ^{238}U series, the transfer factor rate is weak but uranium compounds are more mobile than the other two radionuclides. The magnitude of the radionuclide transfer is related to the geochemical properties of the site. The soil acidity in this area facilitates the mobility of the ^{238}U series. From a geochemical point of view, the uranyl complexes are very soluble in the acidic media. This is not the case for the ^{40}K and the ^{232}Th series which are less mobile under local geochemical conditions. Frequent rainfall in the region has also permanently leached and depleted the soil radionuclides, resulting in a reduced magnitude of the process at present. The thick humic soil covers that limit the erosion phenomena also leads to low transfer within sites, despite the rugged relief and the steep slopes which generally cause torrential streams. Moreover, basaltic soils and rocks covering the site do not have significant levels of radioactivity.

Conclusion

Spring, lake and river water samples were collected from Amber Mountain catchment, in northern Madagascar. Measurements indicate that the ^{40}K, the ^{238}U series, and the ^{232}Th series concentrations are well quantified in the water from the site. Natural radioactivity levels in waters and soils vary from one place to another, and are not spatially uniform. This can be explained by the geological characteristics of the area of interest with low levels of radioactive minerals, the geographical and tropical environmental diversities of the sites studied. The results show that the averages of soil-water transfer rates are estimated at 4%, 10% and 3% for ^{40}K, ^{238}U series and ^{232}Th series, respectively. These results are relatively low, especially for ^{40}K and the ^{232}Th series. Migration of the ^{238}U series is also weak but the latter is more mobile, compared to the former two, because of the soil acidity in the study sites.

To conclude, knowledge of these data makes it possible to evaluate the transfer magnitude and the amount of natural radionuclides in the waters of the Amber Mountain catchment, located in northern Madagascar. The results of the study are important because the natural radioactivity contents in drinking water used by the local population are determined. The potential radiological risks related to ingestion need further investigation in order to help the decision-makers to mitigate the related health risks.

For the African perspective, and from the literature review conducted during this work, it can be noted that, compared to other regions, scientific data production lags behind in Africa. This is especially true in the radio-ecological field, due to the lack of specialized equipment such as nuclear spectrometry and other radio-analytical tools. It is therefore important that for regional countries and research institutions capable of carrying out such investigation, strengthening research activities and data production in the radio-ecological field is necessary throughout the African continent to fill the gap, allowing better contribution to the protection of the African population against the harmful effects of ionizing radiation. The data obtained makes it possible to consolidate and to build a regional database, therefore allowing having a clear idea of the situation at the African level. Knowledge and skill exchanges are also advised to strengthen pan-African scientific capacities.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

Acknowledgments

The present work in general, and the sample analytical activities in particular, were performed at the Institut National des Sciences et Techniques Nucléaires (INSTN-Madagascar). Therefore, our thanks go to the Institute Director General and staff, such as, (i) the Nuclear Analyses and Techniques Department scientific and technical team for facilitating access to the preparation and measurement equipments, for assisting in their use, and for helping in the data treatment and interpretation, (ii) the Institute administrative staff, who created pleasant and carefree fellowship at the Institute, and the National Liaison Officer with I.A.E.A for giving opportunities to participate in training courses, under the framework of IAEA TC projects RAF5063 and RAF7017. The knowledge gained has strengthened the scientific skills in environmental radioactivity.

References

[1] Madagascar National Parks (MNP), Mémorandum parc national montagne d'Ambre (2014) pp. 1–14. URL: Memorandum parc national montagne d'ambre (doczz.fr).
[2] P. Ségalen, Notice sur la carte pédologique de reconnaissance au 1/200000^{e}, feuille n 1, Diego Suarez. Mémoires de l'Institut Scientifique de Madagascar, série D, Tome VII, 1956.
[3] C. Barat, La montagne d'Ambre (nord de Madagascar), Revue Géogr. Alpine 46 (4) (1958) 629–681.
[4] United Nation Scientific Committee on the Effects of Atomic (UNSCEAR). Sources and effects of ionizing radiation: exposures from natural radiation sources. Report to the General Assembly with scientific annexes B. United Nations, New York, USA. 2000, pp. 84–156.
[5] M. Amrane, L. Oufni, Determination for levels of uranium and thorium in water along OumEr-Rabia river using alpha track detectors, Morocco, J. Radiat. Res. Appl. Sci. 10 (2017) 246–251, doi:10.1016/j.jrras.2017.05.002.
[6] M. Rasolonirina, Etude de la Radioactivité des Eaux de Consommation en Milieu Uranifère: Cas de Vinaninkarena et ses Environs, Université d'Antananarivo, 2003 Mémoire de DEA.
[7] H.N. Ravoson, Contribution à l'étude de la Radioactivité des Eaux de Consommation àMadagascar: Cas de la Localité d'Alasora, Université d'Antananarivo, 2007 Mémoire de DEA.
[8] F.E. Sahoa, Etude de la Radioactivité des eaux de Consommation : Cas des eaux de puits et eaux de sources d'Ambohimangakely, Université d'Antananarivo, 2007 Mémoire de DEA.
[9] T.H. Randriamora, H.A. Razafindramiandra, R. Andriambololona, S.D. Ravelomanantsoa, M.A.L. Ralaivelo, M. Rasolonirina, J.L.R. Zafimanjato, H.F. Randriantseheno, Determination of natural radioactivity in the North East Beach Sands of Madagascar, Am. J. Phys. Appl. 5 (1) (2017) 6–12, doi:10.11648/j.ajpa.20170501.12.
[10] P. Lemoine, in: Etudes géologiques dans le Nord de Madagascar : Roches éruptives basaltiques récentes, XIX, Paris, Librairie scientifique A. HERMANN, 1906, pp. 278–296. Chap.
[11] International Atomic Energy Agency, in: Measurement of radionuclides in food and the environment. A guidebook, Technical reports series, 1989, p. 170.
[12] H. El-Gamal, H. Negm, M. Hasabelnaby, Detection efficiency of NaI(Tl) detector based on the fabricated calibration of HPGe detector, J. Radiat. Res. Appl. Sci. 12 (1) (2019) 360–366, doi:10.1080/16878507.2019.1672313.
[13] P. Chiozzi, P. De Felice, A. Fazio, V. Pasquale, M. Verdoya, Laboratory application of NaI (Tl) gamma-ray spectrometry to the studies of natural radioactivity in geophysics, Appl. Radiat. Isotopes 53 (2000) 127–132, doi:10.1016/s0969-8043(00)00123-8.
[14] J. Bezuidenhout, Measuring naturally occurring uranium in soil and minerals by analysing the 352 keV gamma-ray peak of ^{214}Pb using a NaI(Tl)-detector, Appl. Radiat. Isotopes 80 (2013) 1–6, doi:10.1016/j.apradiso.2013.05.008.
[15] AndrianjafitrimoEtablissement d'un dispositif d'analyse spectrométrique gamma au NaI(Tl): application à la caractérisation de la radioactivité des sols, Thèse de 3^{e}cycle, Université d'Antananarivo, 2001.
[16] C.H. Gong, G.Q. Zeng, L.Q. Ge, X. Tang, C.J. Tan, Minimum detectable activity for NaI(Tl) airborne g-ray spectrometry based on Monte Carlo simulation, Sci. China Tech. Sci. 57 (9) (2014) 1840–1845, doi:10.1007/s11431-014-5553-x.

[17] S.G. Dlamini, M.M. Mathuthu, V.M. Tshivhase, Radionuclides and toxic elements transfer from the pincess dump to water in Roodepoort, South Africa, J. Environ. Radioact. 153 (2016) 201–205, doi:10.1016/j.jenvrad.2015.12.026.

[18] M.M. Ndontchueng, A. Simo, E.J.M. Nguelem, J.F. Beyala, D. Kryeziu, Preliminary study of natural radioactivity and radiological risk assessment in some mineral bottledwater produced in Cameroon, Int. J. Sci. Technol. 3 (5) (2013) 2224–3577 http://www.ejournalofsciences.org.

[19] M.R. Calin, I. Radulescu, A.C. Ion, F. Sirbu, Radiochemical investigations on natural mineral waters from Bucovina region, Romania, Romanian J. Phys. 61 (2016) 1051–1066 https://www.researchgate.net/publication/306314854_Radiochemical_investigations_on_natural_mineral_waters_from_Bucovina_region_Romania.

[20] M. Beyermann, T. Bünger, K. Schmidt, D. Obrikat, Occurrence of natural radioactivity in public water supplies in Germany: 238U, 234U, 235U, 228Ra, 226Ra, 222Rn, 210Pb, 210Po and gross a activity concentrations, Radiat. Prot. Dosim. 141 (1) (2010) 72–81, doi:10.1093/rpd/ncq139.

[21] F.A. Alseroury, T. Almeelb, A. Khan, M.A. Barakata, J.H. Al-Zahrani, W. Alali, Estimation of natural radioactive and heavy metals concentration in underground water, J. Radiat. Res. Appl. Sci. 11 (2018) 373–378, doi:10.1016/j.jrras.2018.07.004.

[22] A. Faanu, J.H. Ephraim, E.O. Darko, Assessment of public exposure to naturally occurring radioactive materials from mining and mineral processing activities of Tarkwa Gold mine in Ghana, Environ. Monit. Assess. 180 (2011) 15–29, doi:10.1007/s10661-010-1769-9.

[23] N.K. Mohammed, M.S. Mazunga, Natural Radioactivity in Soil and Water from Likuyu Village in the Neighborhood of Mkuju Uranium Deposit, Int. J. Anal. Chem. (2013) 4 2013, doi:10.1155/2013/501856.

[24] M. Yehia, A. Baghdady, F.M. Howaric, S. Awad, A. Gad, Natural radioactivity and groundwater quality assessment in the northern area of the Western Desert of Egypt, J. Hydrol. Reg. Stud. 12 (2017) 331–344, doi:10.1016/j.ejrh.2017.06.002.

[25] A.S. Pradeep, H. Geremew, B. Getachew, Natural Radio Activity Levels in Water and Soil at Kemessie Hot Spring, North-Eastern Ethiopia, Radiat. Sci. Technol. 2 (1) (2016) 1–5, doi:10.11648/j.rst.20160201.11.

[26] Moshiur Rahman, A.M. M., M.A. Kabir, K. Asaduzzaman, Assessment of Natural Radioactivity Levels and Radiological Significance of Bottled Drinking Water in Bangladesh, Am. J. Phys. Appl. 3 (6) (2015) 203–207, doi:10.11648/j.ajpa.20150306.13.

[27] A.I.A. El-Mageed, A.E.H. El-Gamel, A.E.B. Abbacy, S. Herb, I.I. Sale, Natural radioactivity of ground and hot spring water in some areas in Yemen, Desalination 321 (2013) 28–31, doi:10.1016/j.desal.2011.11.022.

[28] F.O. UGBEDE, B.C. ADUO, O.N. OGBONNA, O.C. EKOH, Natural radionuclides, heavy metals and health risk assessment in surface water of Nkalagu river dam with statistical analysis, Sci. Afr. 8 (2020) e00439, doi:10.1016/j.enmm.2021.100503.

[29] A.H. Al-Mashhadani, A.M. Saleh, Assessment of radioactivity and associated hazards in drinking water in Al-Sadar City, Baghdad, Int. J. Geol. Agr. Environ. Sci. 2 (4) (2014) www.woarjournals.org/IJGAES. ISSN2348-0254.

[30] B. Almayahi, A. Tajuddin, M. Jaafar, Radiation hazard indices of soil and water samples in Northern Malaysian Peninsula, Appl. Radiat. Isotopes 70 (11) (2012) 2652–2660, doi:10.1016/j.apradiso.2012.07.021.

[31] T. Manickum, W. John, S. Terry, K. Hodgson, Preliminary study on the radiological and physicochemical quality of the Umgeni Water catchements and drinking water sources in KwaZulu-Natal, South Africa, J. Environ. Radioact. 137 (2014) 227–240, doi:10.1016/j.jenvrad.2014.07.015.

[32] National Academy PressEvaluation of Guidelines for Exposures to Technologically Enhanced Naturally Occurring Radioactive Materials. Committee on Evaluation of EPA Guidelines for Exposures to Naturally Occurring Radioactive Materials. Board on Radiation Effects Research. Commission on Life sciences, National Research Council, Washington, D.C, 1999 https://www.ncbi.nlm.nih.gov/books/NBK230656/.

[33] A.M. Gbadebo, A.J. Amos, Assessment of radionuclide pollutants in bedrocks and soils from Ewekoro Cement Factory, Southwest Nigeria, Asian J. Appl. Sci. 3 (2) (2010) 135–144, doi:10.3923/ajaps.2010.135.144.

[34] J.C.S. Guembou, M.M. Ndontchueng, J.E.M. Nguelem, G.C.S. Kayo, O. Motapon, D. Strivay, Elemental quantification and radioactive characterization of soil from Douala Bassa area: littoral region of Cameroon using X- and γ-rays spectrometry, Environ. Res. Commun. 1 (2019) 065001, doi:10.1088/2515-7620/ab1d72.

[35] E.E. Saleh, A.I. El-Mageed, A.H. El-Kamel, A. Abbady, S. Harb, in: Natural radioactivity in the volcanic field north of Sana'a, 34, Radiat Prot Environ, Yemen, 2011, pp. 275–281, doi:10.4103/0972-0464.106205.

[36] E.J.M. Nguelem, M.M. Ndontchueng, O. Motapon, in: Determination of ^{226}Ra, ^{232}Th, ^{40}K, ^{235}U and ^{238}U Activity Concentration and Public dose Assessment in Soil Samples from Bauxite Core Deposits in Western Cameroon, 5, Springer Plus, 2016, p. 1253, doi:10.1186/s40064-016-2895-9.

[37] B. Kall, T. Tombo, M. Rasolonirina, N. Rabesiranana, G. Rambolamanana, Study of the dosimetric impact due to the gamma radioactivity of the soil in the border of the "baie des français", Antsiranana, Madagascar, Afr. Sci. 11 (1) (2015) 122–135 ISSN 1813-548X http://www.afriquescience.info .

[38] A. Durusoy, M. Yildirim, Determination of radioactivity concentrations in soil samples and dose assessment for Rize Province, Turkey, J. Radiat. Res. Appl. Sci. 10 (2017) 348–352, doi:10.1016/j.jrras.2017.09.005.

[39] P. Yalcin, H. Taskin, E. Kam, H. Taskin, M. Terzi, A. Varinlioglu, A. Bozkurt, A. Bastug, B. Tasdelen, Investigation of radioactivity level in soil and drinking water samples collected from the city of Erzincan, Turkey, J. Radioanal. Nucl. Chem. 292 (2012) 999–1006, doi:10.1007/s10967-011-1596-7.

[40] A.K. Ademola, A.K. Bello, A.C. Adejumobi, Determination of natural radioactivity and hazard in soil samples in and around gold mining area in Itagunmodi, south-western, Nigeria, J. Radiat. Res. Appl. Sci. 7 (2014) 249–255, doi:10.1016/j.jrras.2014.06.001.

[41] S.H. Taha, O.R. Sallam, A.E.A. Abbas, N.S. Abed, Radioactivity and environmental impacts of ferruginous sandstone and its associating soil, Int. J. Environ. Anal. Chem. (2020), doi:10.1080/03067319.2020.1715377.

[42] N. Rabesiranana, Analyse multi-groupe des spectres gamma: Application à l'étude de la radioactivité des sols, Thèse de 3ecycle, Université d'Antananarivo, 2001.

[43] S. Harb, A.H. El-Kamel, E.M. Zahran, A. Abbady, F.A. Ahmed, Assessment of Agriculture Soil Primordial Radionuclide Concentrations in Aden Governorate South of Yemen Region, Int. J. New Horizons Phys. 2 (2) (2015) 81–86, doi:10.12785/ijnhp/020207.

[44] M.A. Rafat, Radioactivity levels in some sediments and water samples from Qarun Lake by Low–Level Gamma Spectrometry, Int. J. Sci. Res. 4 (2) (2015) www.ijsr.net. Paper ID: SUB151080 ISSN: 2319-7064.