

Research Article

Determination of ^{238}U and ^{232}Th Levels in Well Water and Associated Radiological Hazards in the Municipality of Ivato Airport, Madagascar

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Abstract

Groundwater always contains uranium and thorium but with varying concentrations depending on the area geological formation. Long-term ingestion of water containing high natural radioactivity levels could cause diverse effects to human health. This is the reason why the study of two radionuclides ^{238}U and ^{232}Th was conducted in the well water in the Municipality of Ivato airport, Madagascar. Measurements have been performed by high-resolution gamma spectrometry system with coaxial HPGe detector to determine the specific activities of two radionuclides in ten samples. The samples were put into 1 litre Marinelli beakers, sealed hermetically and stored for three weeks to reach secular equilibrium between ^{226}Ra and its daughters before analyses. Results obtained showed that the specific activities vary from $(4.08 \pm 0.52) \text{ Bq.kg}^{-1}$ to $(4.48 \pm 0.70) \text{ Bq.kg}^{-1}$ with an average value of $(4.36 \pm 0.12) \text{ Bq.kg}^{-1}$ and from $(4.40 \pm 0.42) \text{ Bq.kg}^{-1}$ to $(4.80 \pm 0.73) \text{ Bq.kg}^{-1}$ with an average value of $(4.61 \pm 0.12) \text{ Bq.kg}^{-1}$ respectively. The effective annual dose rate due to ingestion were also determined, and compared to the worldwide average values, the WHO reference value and the permissible limit value of public. Statistical studies of the data were carried out to understand the behavior of uranium and thorium elements in these waters. Generally, radionuclides present in well water have normal levels of radioactivity. This means that these levels do not present any radiological risks to the local population.

Keywords

Well Water, Natural Radioactivity, Ivato Airport, Specific Activity, Effective Dose Rate

1. Introduction

Water is essential for human life. Several areas of life require water such as construction, sanitation, consumption, irrigation, livestock, etc. There are several types of water intended for consumption, namely tap water, well water, spring water and

river. About 1.1 billion people worldwide have difficulty accessing safe drinking water [4]. Madagascar is no exception, being a developing country, only 41% of Malagasy people have access to drinking water [12]. According to the national database,

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87.7% of the urban population with access to drinking water comes from improved sources while 70.9% of the rural population consumes unimproved drinking water [12]. Unimproved drinking water is generally obtained from unprotected boreholes, unpumped wells, rainwater, surface water, unprotected spring water. Indeed, humans are exposed to ionizing radiation either from artificial or natural sources. According to the literature, exposure to artificial radiation is estimated at 48% and that of natural exposure at 58% [16]. These natural sources of radiation are radioactive elements called radionuclides, radioelements or radioisotopes [27]. Human exposures are mainly due to:

1. either by external exposure by cosmogenic radiation. The collision of cosmic radiation with the atmosphere elements generates cosmogenic radionuclides such as ^3H , ^7Be , ^{14}C , ^{22}Na , ^{32}P , and ^{35}S , with half-lives varying from microseconds to millions of years. Petra and Thibault, reported by the Belgium Federal Agency for Nuclear Control (FANC) [29] in 2018 estimated that external exposure due to cosmogenic radiation in Belgium is 0.3 mSv.a^{-1} .
2. either internal exposure through so-called primordial radionuclides inside the human body. These are radionuclides ^{238}U , ^{235}U , ^{232}Th and their progenies which were formed during the universe creation and are present in rocks [18, 22] and can be present in the form of particles or gases [8]. They enter the human body through the mouth or nose, i.e., by inhalation or ingestion. Air and water are the main transport vectors for these radionuclides. In their natural state, these radionuclides can be composed of several isotopes. For example, uranium isotopes consist of 99.75% ^{238}U , 0.719% ^{235}U and 0.0057% [17]. For these isotopes, however, exposure is mainly due to ^{238}U . As for thorium, it

has 13 radioisotopes with atom masses between 212 and 236 but ^{232}Th series is its main radioisotope. Thorium is 3 to 4 times more abundant than uranium in the Earth's crust. ^{232}Th is present in granites, basalts, schists, sedimentary rocks and more particularly in carbonates, phosphors, silicates and oxides [3]. Thorium has very limited mobility in physical compartments of aquatic environments. Its transfers are mostly linked to solid supports such as suspended matter and sediments. However, thorium is very poorly soluble in water compared to uranium [5].

Due to their short or long-lived decays, these radionuclides expose the stomach, kidney, liver, bladder, bones and bone marrow [11, 28].

Long-term radiation exposure can cause deoxyribonucleic Acid (DNA) damage and cancers with a risk coefficient of $5.5 \cdot 10^{-2}$ per Sievert [10], and according to the International Atomic Energy Agency (IAEA) Safety Standards for protecting people and the environment, (Part 3.) [9] and World Health Organization, (2017) [30] publication, the permissible limit of annual internal exposure for the public is 1 mSv/year .

Uranium and thorium isotopes are present in varying amounts in air, rocks, plants, soil, animals and water. When groundwater comes into contact with igneous rocks, it contains radionuclides [13]. This is the reason why the present work is interested in studying natural radioactivity, in particular ^{238}U and ^{232}Th series in well water of the Municipality of Ivato airport, Madagascar. Indeed, the objective of this study is to investigate the natural radionuclide ^{238}U and ^{232}Th levels in well water used as drinking water, and to evaluate its radiological hazards to the local population.

2. Materials and Methods

2.1. Study Site and Sampling

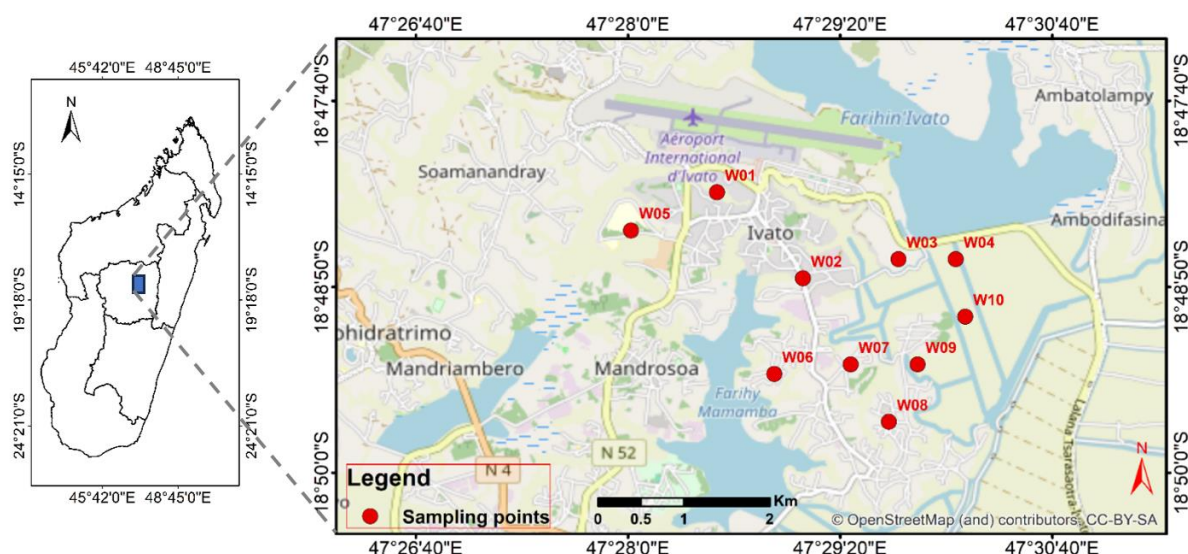


Figure 1. Location of sampling points.

The well water samples were collected at the Municipality of Ivato airport, north-west of Antananarivo capital city, in 2014. The study area is delimited by the geographic coordinates of 18°48'14"S to 18°49'41"S and 47°30'07"E to 47°48'01"E. This site was chosen because it has the biggest international airport in the country and one of the high population density rates in the surroundings of Antananarivo city which is the capital of the country. Due to the high population density, the drinking water supply infrastructure of the National Water Company is insufficient. This is the reason why the local population uses well water as a source of drinking water and for domestic use.

During the sampling work, 10 well water samples were collected and distributed across densely populated areas (Figure 1). Samples were directly put into plastic containers. The geographic coordinates of sampling points were identified by a GPS device. All collected samples were transported to the Institut National des Sciences et Techniques Nucléaires (INSTN-Madagascar) for physical preparation and analyses.

2.2. Sample Preparation and Measurement

All laboratory work was carried out at the Nuclear Analyses and Techniques Department of INSTN-Madagascar. Samples were put directly into 1 L Marinelli beakers. They were then sealed hermetically for three weeks to reach secular equilibrium between ^{226}Ra and its daughters before analyses.

Sample measurements were performed using a gamma spectrometry system with a Canberra high-purity germanium (HPGe) detector, model 7500 SL. The liquid nitrogen-cooled detector was connected to a preamplifier, a multichannel analyzer (MCA) and a computer equipped with Genie 2000 software. Calibration was done using a multisource gamma (187/93-Type: MBSS 2 X 14/93) which contains ^{241}Am , ^{133}Ba , ^{60}Co , ^{137}Cs and ^{22}Na . To have good counting statistics, each sample was counted for more than 24 hours.

2.3. Specific Activity Calculation

The specific activity A (Bq.kg^{-1}) of these natural radionuclides is determined by the equation (1) [32]:

$$A = \left(\frac{C_s}{T_s} - \frac{C_b}{T_b} \right) \frac{1}{\varepsilon \times I_\gamma \times m} \quad (1)$$

where:

- C_s and C_b : sample and background total counts,
- T_s and T_b : sample and background counting times (s),
- ε : detection efficiency,
- I_γ : intensity of gamma-ray emitter,
- m : analyzed sample mass (kg)

Specific activity of ^{238}U was measured through its progenies ^{214}Pb (295 keV and 352 keV) and ^{214}Bi (609 keV and

1764.5 keV). While ^{232}Th concentration was determined through its daughters ^{228}Ac (338 keV, 911 keV and 969 keV), ^{212}Pb (239 keV), ^{212}Bi (727 keV) and ^{208}Tl (583 keV and 861 keV). However, the specific activities of ^{238}U and ^{232}Th are presented by the average concentrations of its respective progenies. The uncertainties of an average values are represented by the standard deviations.

2.4. Annual Effective Dose Assessment

In this present work, the radiological risk is due to the ingestion of naturally occurring radioactive (^{238}U and ^{232}Th series). Therefore, the health risk to the local population depends on the radionuclide levels present in drinking water. It means that the risk is high when its specific activities are important. As potassium concentration in the human body is being under homeostatic control, the estimation of the annual effective dose is based only on two radionuclides ^{238}U and ^{232}Th .

For ^{238}U , the effective dose coefficients are 0.12 mSv.Bq^{-1} , 0.068 mSv.Bq^{-1} and 0.045 mSv.Bq^{-1} for infants, children and adults respectively. Regarding ^{232}Th , these coefficients are 0.45 mSv.Bq^{-1} , 0.29 mSv.Bq^{-1} and 0.23 mSv.Bq^{-1} respectively [25].

The consumption rates are considered to be 150 kg.a^{-1} , 350 kg.a^{-1} and 500 kg.a^{-1} for infants, children and adults respectively [25].

3. Results and Discussions

3.1. Specific Activity

The analytical results obtained are shown in Table 1. The specific activity variabilities of ^{238}U and ^{232}Th . The specific activity of ^{238}U varies from $(4.08 \pm 0.52) \text{ Bq.kg}^{-1}$ to $(4.48 \pm 0.70) \text{ Bq.kg}^{-1}$ with an average value of $(4.36 \pm 0.12) \text{ Bq.kg}^{-1}$. The minimum value is found in sample W02, while that of the maximum is determined in samples W03 and W07. For ^{232}Th , the specific activity is ranged from $(4.40 \pm 0.42) \text{ Bq.kg}^{-1}$ to $(4.80 \pm 0.73) \text{ Bq.kg}^{-1}$ with an average value of $(4.61 \pm 0.12) \text{ Bq.kg}^{-1}$. The maximum value is measured in sample W09. Like ^{238}U , the minimum value is found in sample W02. The average activities of ^{238}U and ^{232}Th are significantly higher than the reference values of 1 mBq.kg^{-1} and 0,05 mBq.kg^{-1} respectively [26].

Nevertheless, the average value of ^{238}U obtained in the present study is relatively close to that obtained in spring water from the Amber Mountain catchment, in the north of the country, which is $(3.9 \pm 1.7) \text{ Bq.kg}^{-1}$ [6].

The $^{232}\text{Th}/^{238}\text{U}$ ratio is also calculated to be able to predict the depositional environment of uranium and thorium and describe their radiochemical behavior. The results obtained show that the ratio vary from (1.02 ± 0.23) to (1.10 ± 0.27) with an average of (1.06 ± 0.03) . These results are all less than

2, meaning that uranium deposited in the tetravalent state (U^{4+}) in a reducing environment. It should be remembered that uranium possesses several oxidation states such an insolubil-

ity of U^{4+} and its fixation under reducing environment whereas hexavalent state (U^{6+}) is soluble and mobilized in aquatic medium [15].

Table 1. Specific activity of ^{238}U and ^{232}Th .

Sample Code	Specific Activity ($Bq.kg^{-1}$)		$^{232}Th/^{238}U$ ratio
	^{238}U	^{232}Th	
W01	4.30 ± 0.71	4.74 ± 0.86	1.10 ± 0.27
W02	4.08 ± 0.52	4.40 ± 0.42	1.08 ± 0.17
W03	4.48 ± 0.70	4.70 ± 0.88	1.05 ± 0.26
W04	4.39 ± 0.69	4.56 ± 0.72	1.04 ± 0.23
W05	4.39 ± 0.69	4.59 ± 0.73	1.05 ± 0.23
W06	4.39 ± 0.69	4.57 ± 0.85	1.04 ± 0.25
W07	4.48 ± 0.70	4.56 ± 0.72	1.02 ± 0.23
W08	4.47 ± 0.73	4.67 ± 0.79	1.04 ± 0.25
W09	4.39 ± 0.69	4.80 ± 0.73	1.09 ± 0.24
W10	4.28 ± 0.79	4.54 ± 0.95	1.06 ± 0.30
Mean $\pm \sigma$	4.36 ± 0.12	4.61 ± 0.12	1.06 ± 0.03
Coefficient of Variation (CV)	2.8%	2.5%	2.5%

σ : Standard Deviation

Figure 2 below shows the graphical representation of the specific activities of ^{238}U and ^{232}Th and their correlation between two radionuclides. In general, the ^{232}Th activity is slightly higher than that of ^{238}U (Figure 2a). Correlation analysis was carried out to determine the mutual relationships and association between two radionuclides. A strong positive

correlation is observed between ^{232}Th and ^{238}U (Figure 2b), meaning that their origin and behavior in the groundwater are the same [14]. In other words, the two radionuclides present in well water at the study area could come probably from the same source rock.

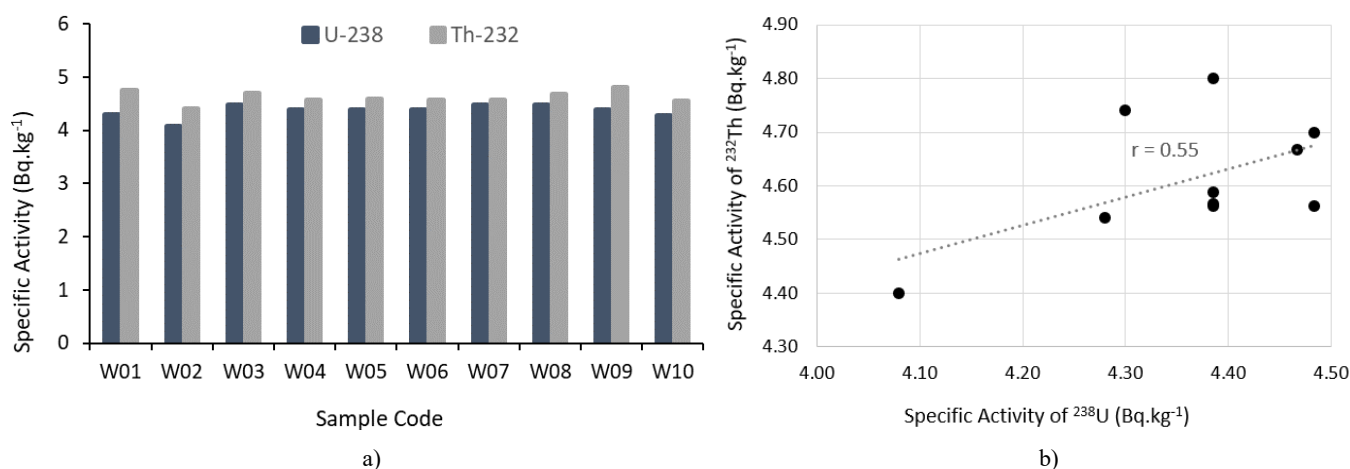


Figure 2. a) Histogram and b) correlation of two radionuclides.

Basic descriptive statistics are used to show characteristics of two radionuclide specific activities in the well water in the study area. Statistical parameters are estimated and summarized in Table. The results show that the basic statistic parameters such as mean, standard deviation, minimum, median, mode, maximum values of two radionuclide activities are close to each other (Table 2). Other parameters such as skewness and kurtosis are also determined. Skewness is used to determine the asymmetry of the probability distribution of a real-valued random variable about its mean. Normal distribution is obtained when the skewness value is zero, which is not possible for environmental data. The experimental skewness value is either positive or negative. Therefore, these negative results of -1.48 for ^{238}U and -0.08 for ^{232}Th show a distribution with an asymmetric long tail to the left. As for kurtosis, it is used to distinguish the normal distribution shape, i.e., a peaked distribution for a positive value and a flat distribution for a negative value. In this study, these positive kurtoses show a peaked distribution.

Table 2. Statistical distribution of specific activities.

Parameter designations	^{238}U	^{232}Th
Mean	4.36	4.61
Median	4.39	4.58
Mode	4.39	4.56
Standard deviation	0.12	0.12
Kurtosis	2.75	0.13
Skewness	-1.48	-0.08
Minimum	4.08	4.40
1 st quartile (Q1)	4.30	4.56
3 rd quartile (Q3)	4.47	4.70
Maximum	4.48	4.80
Total sample number	10	10

The median is positioned close to the center of Q1 and Q3 for both radionuclides, confirming a skewed distribution data set.

In addition, the independent samples *t*-test was applied in order to examine and compare the mean values of ^{238}U and ^{232}Th specific activities. The results obtained show that the calculated *p*-value ($p = 0.0002$) is below the significance value of $\alpha = 0.05$. It means that there is a significant difference between the values of two radionuclides. The difference is, however, due to the behavior and mobility of the uranium and thorium elements in the environment. Mobilization is accelerated by human activities such as agriculture, urbanization, mining, etc. Nevertheless, thorium is less mobile than uranium, especially at low temperatures. Moreover, thorium is considered the most immobile chemical element due to its very low solubility in aqueous solution. As for uranium, it is due to the geochemical mobility of uranium which ensures that its spatial distribution remains generally uniform.

The average specific activities obtained were also compared to some studies in the country and results in some countries in the world (Table 3). It should be noted that some studies express the specific activity in Bq. L⁻¹. But we considered that one liter of water is equivalent to one kilogram.

The average ^{238}U activity is significantly higher than that found in groundwater from Saudi Arabia [2] and in drinking water from Serbia [24]. It is slightly higher than the results found in groundwater from Bangladesh [15], in drinking water from Iraq [1] and in ground and hot spring water from Yemen [20]. This average activity is relatively similar to those obtained in well water from Nigeria [7] and in spring and surface water from Madagascar [6]. On the other hand, it is significantly lower than the results found in the river from Madagascar [23], in well water from Iran [19] and in groundwater from Egypt [31]. It should be noted that these last three sites are located in rich areas of natural radioactive ores.

Regarding the ^{232}Th , the specific activity is higher than the results found in Saudi Arabia, in Serbia and in Yemen [2, 20, 24]. Nevertheless, it is significantly lower than that obtained in Madagascar surface water flowing through a zone rich in zirconium and monazite ores [23].

Table 3. Extracts from worldwide average activity of ^{238}U and ^{232}Th series.

Country	Type	Specific Activity (Bq.kg ⁻¹)		References
		^{238}U	^{232}Th	
Madagascar	Well water	4.36	4.61	Present work
Madagascar	Spring, lake, river	3.90	0.90	Donné et al., 2021
Nigeria	Well water	3,62	2,62	Godwin et al., 2021
Bangladesh	Groundwater	2.59	2.45	Islam et al., 2021
Serbia	Drinking water	1.20	0.41	Alseroury Todorović et al., 2020

Country	Type	Specific Activity (Bq.kg ⁻¹)		References
		²³⁸ U	²³² Th	
Saudi Arabia	Underground water	0.56	0.20	et al., 2018
Egypt	Groundwater	92.00	5.70	Yehia et al., 2017
Madagascar	River	9.20	13.01	Tiana Harimalala et al., 2017
Iraq	Drinking water	2.86	3.78	Al-Mashhadani & Saleh, 2014
Iran	Well water	32.4	-	Saghatchi & Salouti, 2012
Yemen	Ground and hot spring water	2,96	0,72	Saleh et al., 2011

3.2. Annual Effective Dose

The results obtained show that the annual effective doses due to ingestion vary from (0.37 ± 0.03) mSv.a⁻¹ to (0.40 ± 0.05) mSv.a⁻¹ with an average value of (0.39 ± 0.01) mSv.a⁻¹, from (0.54 ± 0.04) mSv.a⁻¹ to (0.59 ± 0.08) mSv.a⁻¹ with an average value of (0.57 ± 0.01) mSv.a⁻¹ and from (0.60 ± 0.03) mSv.a⁻¹ to (0.65 ± 0.09) mSv.a⁻¹ with an average value of (0.63 ± 0.01) mSv.a⁻¹ for infants, children and adults respectively (Table 4). The average values obtained fluctuate around 2% to 3%. These weak fluctuations confirm the natural radioactivity levels of well waters in the studied area. The minimum and maximum values are always found at samples W02 and W09 respectively. It is noted that the effective doses obtained in adults are greater than in infants and children. In addition, the values found in children are higher than those evaluated in infants. The calculated average values are higher than the worldwide average values of 0.26 mSv.a⁻¹ for infants, of 0.20 mSv.a⁻¹ for children and of 0.11 mSv.a⁻¹ for adults [21].

Similarly, the values obtained are significantly higher than the Individual dose criterion (IDC), reference value of 0.1 mSv.a⁻¹ [30]. Nevertheless, according to IAEA Safety Standards Publication (No. GSR Part 3) [9] and World Health Organization (2017) [30], these average values are below the admissible effective dose value of 1 mSv.a⁻¹ for the public. It means that the well water in the area presents no danger for the local population for its consumption and uses.

Table 4. Effective dose estimated by ingestion.

Sample Code	Total effective dose (mSv.a ⁻¹)		
	Infants	Children	Adults
W01	0.40 ± 0.06	0.58 ± 0.09	0.64 ± 0.10
W02	0.37 ± 0.03	0.54 ± 0.04	0.60 ± 0.03
W03	0.40 ± 0.06	0.58 ± 0.09	0.64 ± 0.10

Sample Code	Total effective dose (mSv.a ⁻¹)		
	Infants	Children	Adults
W04	0.39 ± 0.05	0.57 ± 0.08	0.62 ± 0.08
W05	0.39 ± 0.05	0.57 ± 0.08	0.63 ± 0.09
W06	0.39 ± 0.06	0.57 ± 0.09	0.62 ± 0.10
W07	0.39 ± 0.05	0.57 ± 0.08	0.63 ± 0.08
W08	0.40 ± 0.06	0.58 ± 0.08	0.64 ± 0.09
W09	0.40 ± 0.05	0.59 ± 0.08	0.65 ± 0.09
W10	0.38 ± 0.07	0.56 ± 0.10	0.62 ± 0.11
Mean $\pm \sigma$	0.39 ± 0.01	0.57 ± 0.01	0.63 ± 0.01

σ : standard deviation

4. Conclusion

Two natural radionuclides ²³⁸U and ²³²Th were investigated in well water samples by using gamma spectrometry system with coaxial HPGe detector. The results obtained show that the specific activities vary from (4.08 ± 0.52) Bq.kg⁻¹ to (4.48 ± 0.70) Bq.kg⁻¹ with an average value of (4.36 ± 0.12) Bq.kg⁻¹ and from (4.40 ± 0.42) Bq.kg⁻¹ to (4.80 ± 0.73) Bq.kg⁻¹ with an average value of (4.61 ± 0.12) Bq.kg⁻¹ respectively. The ²³²Th/²³⁸U ratios are also determined to predict the oxidation states of thorium and uranium elements in the studied samples. The estimation of the annual effective dose rates due to ingestion vary from (0.37 ± 0.03) mSv.a⁻¹ to (0.40 ± 0.05) mSv.a⁻¹ with an average value of (0.39 ± 0.01) mSv.a⁻¹ for infants, from (0.54 ± 0.04) mSv.a⁻¹ to (0.59 ± 0.08) mSv.a⁻¹ with an average value of (0.57 ± 0.01) mSv.a⁻¹ for children and from (0.60 ± 0.03) mSv.a⁻¹ to (0.65 ± 0.09) mSv.a⁻¹ with an average value of (0.63 ± 0.01) mSv.a⁻¹ for adults. These results have exceeded the average worldwide values of the United Nation Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 2000 and the WHO reference value. Nevertheless, the annual effective dose rates are still

below the permissible international standard limit value of 1 mSv.a⁻¹, meaning that the local population is not expected to be exposed to serious radiological complications from the ground-water system. Statistical analysis indicates that both radionuclides in the area are probably come from the same source rock. However, this first database on the natural radioactivity levels in the local drinking water can be used to assess possible changes due to urbanization and other human activities.

Abbreviations

DNA	Deoxyribonucleic Acid
FANC	Federal Agency for Nuclear Control
IAEA	International Atomic Energy Agency
WHO	World Health Organization
INSTN-Madagascar	Institut National des Sciences et Techniques Nucléaires-Madagascar
HPGe	High-purity Germanium
MCA	Multichannel Analyzer
IDC	Individual Dose Criterion
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation

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Author Contributions

Charles Oyverné Randriamaholisoa: Conceptualization, Investigation, Methodology, Writing – original draft, Writing – review & editing

Frank Elliot Saho: Conceptualization, Methodology, Data curation, Writing – review & editing

Jean Remi Randriantsivry: Writing – original draft, Visualization, Writing – review & editing

Martin Rasolonirina: Conceptualization, Methodology, Writing – review & editing, Supervision, Validation

Conflicts of Interest

The authors declare no conflicts of interest.

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