

e-ISSN: 2319-8753 | p-ISSN: 2347-6710

EDIA

International Journal of Innovative Research in SCIENCE | ENGINEERING | TECHNOLOGY

INTERNATIONAL JOURNAL OF INNOVATIVE RESEARCH

800

IN SCIENCE | ENGINEERING | TECHNOLOGY

Volume 12, Issue 5, May 2023

INTERNATIONAL STANDARD SERIAL NUMBER INDIA

Impact Factor: 8.423

9940 572 462

6381 907 438

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|e-ISSN: 2319-8753, p-ISSN: 2347-6710| www.ijirset.com | Impact Factor: 8.423| A Monthly Peer Reviewed & Referred Journal |

|| Volume 12, Issue 5, May 2023 ||

| DOI:10.15680/IJIRSET.2023.1202007 |

Determination of Water Radioactivity Transfer from Telluric origin from Bemanevika Sambirano, Madagascar

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ABSTRACT: Natural radioactivity in water comes from direct contact with the ground it passes through. The water is charged with radioactive elements during its passage through the rocks that can include high levels of natural radioactivity belonging to potassium-40, to the series of uranium-238 and thorium-232. The objectives of this work are to determine radionuclides in the waters, soil and their transfer –factor. Twenty samples have been collecting from various locations at the study site including 10 of water and 10 of soils. The results of the water samples show that ⁴⁰k, ²³⁸U series and ²²²Th series activities vary, respectively from (32±5) to (58±5) Bq. 1⁻¹, (11±2) to (21±2) Bq. 1⁻¹ and (24±2) to (38±4) Bq. 1⁻¹. For soil samples, the activities are in the range of (563±18) to (1069±27) Bq. kg⁻¹, (32±3) to (61±4) Bq. kg⁻¹ and (102 ± 6) to (166±7) Bq. kg⁻¹, respectively for the ⁴⁰k, the ²³⁸U and the ²³²Th series. Concerning the radionuclide soil-water transfer factors, the mean values for the whole study site are 0.04 for ⁴⁰k, 0.37 for ²³⁸U series and 0.24 for ²³²Th series. These results show that the high Sambirano 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 physical-chemical properties of the radionuclides present in the areas.

KEYWORDS: Natural radioactivity, gamma spectrometry, well water, activity concentrations, soil-water transfer factors.

I. INTRODUCTION

Some people believe that well water, because it is natural, is safe to drink; however, many elements both natural and fabricated that are found in the soil can dissolve and transfer groundwater [7]. Natural and artificial radionuclides can therefore be present in the consumed water [8, 10, and 12]. This problem is very serious especially in the study sites, the population of the Bemaneviky Sambirano uses well water for their usual activities. Water is loaded with radioactive elements as it passes through the rocks [21]. Water plays a very important role in human health [13]. Studies on the radioactivity of the water for consumption were carried out in the Bemanevika countryside affirming the existence of radioactive elements in the water consumed well. The present study has as objective to determine the factor of transfer of the specific activities of potassium-40, the uranium-238 and the thorium-232 series in the water in the Bemaneviky Sambirano.



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II. MATERIALS AND METHODS

II-1 Description of the study sites.

This present work was carried out from the Bemaneviky Sambirano in the District of Ambanja. This study site extends between 13°40'01. 80'' S to 13°50'01. 21'' S and 48°31'37. 32" S at 48°24'0. 023", with an average altitude of 56m. The district of Ambanja covers a total area of 5735 km². The study area is bounded by the Benavony countryside (South-West), the Marovato countryside (South East), the Migiko countryside (North-West). The geological characteristic of that study area is formed of gneiss and migmatites with quartzites, cipolins and basic gneisses belongs to the upper complex of the old Precambrian. The soils in this area are mainly of alluvial and hydromorphic types. This alluvium is recent, brought by runoff, and deposited on the plains furrowed by the great Sambirano River. This type of soil is rich in mineral elements such as sandy loam, sandy loam and loam clay. These soils are intensively exploited for the industrial cultivation of cocoa, coffee and ylang-ylang.



Fig 1: Location of study sites and sampling points.

II-2-Sample collection and preparation.

Fieldwork was carried out in December 2021 in Bemaneviky Sambirano. To investigate the radionuclide transfer rates, soil samples were taken near the water sampling points. The sampling points were located using an Etrex Garmin GPS device. The sampling campaigns made it possible to collect 20 samples, 10 of water and 10 of soils. The samples were prepared in the Analyses and Techniques Nucléaires laboratory of INSTN-Madagascar. They were placed in cylindrical sample holders made of 200-cm3 polyethylene. After having prepared, he samples were then homogenized and sealed hermetically into 200 cm³ cylindrical polyethylene containers for three weeks to reach secular equilibrium of the ²³⁸U series [11]. Using a gamma spectrometer equipped with a NaI (Tl) detector, the prepared samples were analysed. To have a good resolution of the spectrum, the counting time of each sample is fixed at 24 hours, i.e. to have a 95% level of confidence and relative uncertainties lower than 1%. The obtained spectra were processed using ScintiVision software. Next, the Activity concentrations (Bq. kg⁴) of potassium-40, of the uranium-238 and of thorium-232 series were determined.



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II-3- Gamma spectrometry measurement

The chains of gamma spectrometry with the NaI (Tl) detector have rather limited determinations but the energy efficiency of a very good quality. This allows them to be used attentively in the analyses of environmental samples. 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, National Institute of Sciences and Technics, Nuclears (INSTN-Madagascar). The detector energy resolution (FWHM) at the 1332.5 Kiev (for the 60Co 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, 19, 22, and 24]. The water samples were counted for 24 h. Measurements of known masses of samples enabled determination of the specific activity of ⁴⁰K, ²³U series and ²³²Th series. The method described by Rybach was used to process the spectra obtained [14, 13, 18]. This method targets three regions of interest corresponding to three isolated intense total absorption peaks of ⁴⁰K (1461 Kev), 214Bi (1764.5 Kev) and ²³⁸Tl (2614.5 Kev), as shown in the Table 1. The two last radionuclides are progenies of ²³⁸U and ²²²Th series.



Fig 2: NaI (Tl) analysis device from the INSTN-Madagascar laboratory.

II.4. Energy calibration.

In order to be able to identify the radionuclides involved, the spectrum must be calibrated in energy. This consists of establishing a polynomial relationship between the channel number and the energy deposited in the crystal [4, 6, and 2]. To do this, reference materials are used as radioactive sources emitting known gamma radiation. For each peak of the spectrum of reference materials, the channel number C of maximum accumulation of peak counts is noted and the corresponding energy E is associated with it.

$$E = aC + b$$

Or

E: energy in keV;

C: maximum hit accumulation channel number;

a: slope of the curve or gain in keV/channel;

b: energy of the first channel.

The coefficients a and b are determined by the method of least squares. Thus, we have a formula of the form:



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$$a = \frac{\sum (C_i - \overline{C}) (E_i - \overline{E})}{\sum (C_i - \overline{E})^2}$$
[2]

And

$$b = \overline{E} - a\overline{C}$$
[3]

By taking into account the channel number that corresponds to each radionuclide energy given in the table below then by applying the equation above, we obtain an equation of a following straight line: a=3, 61Channel -31, 02

II.5. Calculation of specific activity of natural radionuclides in samples.

Referring to the net counting rates of the three regions of interest (R_k , R_u and R_h) as well as the counting efficiencies, the activities of 40K (A_k), 238U (A_u) and 232th (A_h) of samples can be calculated using the equation system below. Thus, the activities are given by the equation system below in matrix form

$$\begin{pmatrix} R_{K} \\ R_{U} \\ R_{Th} \end{pmatrix} = \begin{pmatrix} e_{1,1} & e_{1,2} & e_{1,3} \\ 0 & e_{2,2} & e_{2,3} \\ 0 & e_{3,2} & e_{3,3} \end{pmatrix} \times \begin{pmatrix} A_{K} \\ A_{U} \\ A_{Th} \end{pmatrix}$$
[4]

The activity concentration are determined from the following relationship:

$$\begin{pmatrix} A_{K} \\ A_{U} \\ A_{Th} \end{pmatrix} = \begin{pmatrix} \frac{1}{e_{1,1}} & \frac{e_{1,3}e_{3,2} - e_{1,2}e_{3,3}}{e_{1,1}\left(e_{2,2}e_{3,3} - e_{2,3}e_{3,2}\right)} & \frac{e_{1,2}e_{2,3} - e_{1,3}e_{2,2}}{e_{1,1}\left(e_{2,2}e_{3,3} - e_{2,3}e_{3,2}\right)} \\ 0 & \frac{e_{3,3}}{e_{2,2}e_{3,3} - e_{2,3}e_{3,2}} & \frac{e_{2,3}}{e_{2,3}e_{3,2} - e_{2,2}e_{3,3}} \\ 0 & \frac{e_{3,2}}{e_{2,3}e_{3,2} - e_{2,2}e_{3,3}} & \frac{e_{2,2}}{e_{2,2}e_{3,3} - e_{2,3}e_{3,2}} \end{pmatrix} \times \begin{pmatrix} R_{K} \\ R_{U} \\ R_{Th} \end{pmatrix}$$

$$\begin{bmatrix} F_{K} \\ F_{K} \\$$

With A_{k} , A_{u} and A_{th} representing the activity concentrations and R_{k} , R_{u} and R_{th} the recorded net count rates. The radioactivity of soils in contact with interstitial or surface waters contributes to the contamination of the water. This phenomenon can be estimated by the radionuclide soil-water transfer factor (TF) given by the following:



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$$TF = \frac{A_{Water}}{A_{sol}}$$

Where A_{water} (Bq.l⁻¹) is the water sample specific activity and A_{soil} (Bq.kg⁻¹) is the soil sample specific activity.

III. RESULTS AND DISCUSSIONS

III.1. Specific Activity of natural radionuclides in the Bemaneviky Sambirano.

After analysing each sample taken from the field, the activity concentrations of potassium-40, of the uranium-238 series and of thorium-232 series are presented in the following table, along with their standard deviation.

Table 1. Specific Activity of the three natural radionuclides in the Bemaneviky sambirano countryside

	Specific Activity (Bq.kg ⁻¹)			
Sample code	K-40	U-238	Th-232	
BMVK1	1062±29	53±3	151±7	
BMVK2	692±21	33±3	118±6	
BMVK3	1069±27	61±4	166±7	
BMVK4	886±24	53±4	124±6	
BMVK5	837±27	44±3	119±7	
BMVK6	900±28	64±4	139±7	
BMVK7	783±21	43±4	132±7	
BMVK8	563±18	32±3	102±6	
BMVK9	781±23	42±3	137±7	
BMVK10	632 ± 21	45±3	115±6	
Mean value $\pm \sigma$	820±167	47±11	130±19	
[Min – Max]	[632 - 1069]	[32-61]	[102 - 166]	
Reference value	400	35	30	

BMVK :Bemanevika ; o : standard deviation ; Min : minimum value ; Max : maximum value



Fig 3: Activity concentrations of K-40, U-238 and Th-232 series



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Table 2: Specific Activity of the three natural radionuclides in the Bemaneviky sambirano countryside

Construction of the	Specific Activity (Bq.kg ⁻¹)			
Sample code	K-40	U-238	Th-232	
BMVK1	1062±29	53±3	151±7	
BMVK2	692±21	33±3	118±6	
BMVK3	1069±27	61±4	166±7	
BMVK4	886±24	53±4	124±6	
BMVK5	837±27	44±3	119±7	
BMVK6	900±28	64±4	139±7	
BMVK7	783±21	43±4	132±7	
BMVK8	563±18	32±3	102±6	
BMVK9	781±23	42±3	137±7	
BMVK10	632 ± 21	45±3	115±6	
Mean value $\pm \sigma$	820±167	47±11	130±19	
[Min – Max]	[632 – 1069]	[32-61]	[102 – 166]	
Reference value	400	35	30	

BMVK : Bemanevika ; σ : standard deviation ; Min : minimum value ; Max : maximum value



Fig 4: Activity concentrations of K-40, U-238 and Th-232 series in well water.



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III.2. Soil-water transfer factors of natural radionuclides.

Table 3. Soil-water Transfer Factors for 40K, 238U and 232Th series in the Bemaneviky Sambirano countryside.

Sample code	transfer factors			
	⁴⁰ K	²³⁸ U	²³² Th	
FTBMVK1	0,04	0,32	0,22	
FTBMVK2	0,07	0,52	0,28	
FTBMVK3	0,04	0,20	0,20	
FTBMVK4	0,07	0,38	0,30	
FTBMVK5	0,04	0,41	0,20	
FTBMVK6	0,05	0,31	0,24	
FTBMVK7	0,05	0,49	0,29	
FTBMVK8	0,06	0,53	0,28	
FTBMVK9	0,05	0,26	0,23	
FTBMVK10	0,05	0,36	0,21	
Mean of transfert factor	0.04	0.37	0.24	



Fig5: soil-water transfer factor of potassium-40, uranium-238 and Thorium-232 series.



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Sample code Specific Activity (Bq.I¹) 40K 232**Th** BMVK1 17±2 45±6 33±5 **BMVK2** 45±6 17±2 33±3 **BMVK3** 48±5 12±2 34±3 BMVK4 58±5 20±2 37±2 **BMVK5** 31±5 18±2 24±5 BMVK6 49±7 20±2 33±5 BMVK7 40±6 21±2 38±4 BMVK8 33±5 17±2 29±3 BMVK9 39±5 11±2 31±2 BMVK10 32 ± 5 16±2 24±2 Mean± σ 42±8 17±3 32±5 [Min – Max] [31 - 58][11 - 21][24 - 37]

Table 4: Activity concentrations of the three natural radionuclides in well water.

BMVK : Bemanevika σ : standard deviation; **Min**: minimum value; **Max**: maximum value

IV. DISCUSSION

• Soil-water Transfer factors

The soil-water transfer factors in the samples reveal that the potassium -40 of Bemaneviky Sambirano countryside is low at 0.04. Other studies conducted in Ambodimanga [1], Amber Mountain [22], Ethiopia [10], Egypt [7], South Africa [13], Yemen [11], and Ghana [14] do not exceed 0.05. Those studies are a priori similar to our study. The result could be linked to the use of organic fertilizers by the local population, which is practically still used in traditional farming methods. In the case of ²³⁸U 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 ²¹⁸U series. The sampling factors are high 0.37 was observed to be higher than the value founded in transfer factors samples from Ambodimanga 0.06[1], Amber Mountains 0.10 [22], Ethiopia 0.01[10]; Egypt 0.27 [7], Yemen 0.02[11] and Ghana 0.04 [14]. On the other hand, those of South Africa are higher by Bemaneviky sambirano, which is 0.48.

In groundwater, from a geochemical point of view, the uranyl complexes are very soluble in the acidic media. This is not the case for the "K and the ²⁵²Th series, which are less mobile under local geochemical conditions. With regard to thorium, it has been observed that the transfer factors are high, which is 0.24. This study is similar to that carried out in Egypt [7], which is 0.20 but, it is on average 2 times lower than value obtained in Ambodimanga, Amber Mountain, Ethiopia, Yemen, SouthAfrica, Ghana . Can be explained by the geological structure and geographical condition of this area.

V. CONCLUSION

By way of conclusion, this work makes it possible to approach the essential subjects in the study of the natural radioactivity of the water wells of the Bemaneviky countryside in the district of Ambanja. The method developed by rybach has been applied to the processing of spectra. Twenty (20) well water samples and soil were collected and analyzed by Na (Tl) gamma spectrometry. The results illustrate that potassium-40, the uranium-238 and the thorium-232 series are detected in the samples in the study areas. The levels of radioactivity of these three natural radionuclides in well water variability in the study area and are no longer uniformly distributed. The differences in the results obtained can be explained by the interaction with the geological and pedological formations of these sites. The activities of natural radionuclides present in



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well water are dominated by uranium-238, followed by the thorium-232 series, as well as that of potassium-40. The results show that the averages of soil-water transfer rates are estimated at 38%, 24% and 4%, for^{. 238}U, ²²³Th series and ⁴⁰k, respectively. These results are relatively low, especially for ⁴⁰k and the ²³²th series. Migration of the ²³⁸U series is also weak, but the latter is more mobile, compared to the former two, because of the soil acidity in the study sites. In the field of the study of environmental radioactivity, the results obtained can help as data and reference for future research on the radioactivity of water and which makes it possible to evaluate the possible radiological risks linked to ionizing radiation of the population in this zone.

VI. ACKNOWLEDGMENTS

The present work in general and the sample analytical activities in particular, were performed at the National Institute of Sciences and Technics, Nuclears (INSTN-Madagascar). Therefore, our thanks go to the Institute Director Technics Development. (1) the Nuclear Analyses and Technics Department Scientific and Technical team for facilitating access to the preparation and measurement equipment's, for assisting in their use, and for helping in the data treatment and interpretation, (2) the Institute administrative staff, who created pleasant and carefree fellowship at the Institute.

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|e-ISSN: 2319-8753, p-ISSN: 2347-6710| www.ijirset.com | Impact Factor: 8.423| A Monthly Peer Reviewed & Referred Journal |

|| Volume 12, Issue 5, May 2023 ||

| DOI:10.15680/IJIRSET.2023.1202007 |

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