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Assessment of Radiological Hazards in Drinking Water (Mineral Water) Consumed by the Malian Population Using Gamma Spectrometry

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Abstract: *Background*: Qualitatively risk to human health is the product of the probability that contaminated water or food will be ingested and the radiological consequence or damage due to the intake. The human being activities can increase the deposition and transportation of naturally occurring radioactive materials (NORM) in the groundwater and surface water bodies. The determination of radionuclide concentration in twelve (12) drinking water in Mali, the calculation of annual effective dose due to their ingestion, the calculation of risk assessment, etc. will permit to the AMARAP to avoid any over exposition (determinist effects) and minimize as well the associated risk due to low doses (stochastic effects). *Materials and Methods*: The health impact due to ingestion of radionuclides from these drinking waters was evaluated by the determination of activity concentration of radionuclides U-238, Th-232 and K-40 using gamma spectrometry analysis. The concentration of gross alpha/beta counting was also evaluated and the radiological hazards were calculated in in these drinking waters. *Results*: The range of activity concentrations for U-238 vary from 0.24 ±0.02 to 9.42 ± 0.8 Bq/l, for Th-232 from 0.28 ± 0.02 to 5.54 ± 0.28 Bq/l and for K-40 from 0.44 ± 0.03 to 4.23± 0.23 Bq/l. The highest value of activity concentration for gross αβ radionuclides was reported in samples Emin05 (CRISTALINE) 2,4 Bq/l. The mean values of radiological hazard such as risk assessment (RA) and annual committed effective dose (AED) from this work were within the dose criteria limits given by international organizations (ICRP and UNSCEAR) and national standards. *Conclusion*: Based on the obtained results in this study, these drinking waters are safe for human consumption even if the risk (stochastic effect) associated with internal exposure due to low dose intakes exists. Based on obtained values, the probability of someone dying of cancer due to the ingestion of these drinking waters is less than 10^{-5} in the Malian population.

Keywords: Activity Concentration, Annual Committed Effective Dose, Drinking Water, Risk Assessment

1. Introduction

Safe food production and consumer protection are among the main tasks of legal authorities in every country. The food quality can be influenced by many factors and one of them is certainly the level of radioactivity. The major sources of public exposure to natural radiation are cosmic and terrestrial radiation, inhalation of air/dust and ingestion of water or food contaminated by radionuclides. Qualitatively risk to human health is the product of the probability that contaminated water or food will be ingested and the radiological consequence or damage due to the intake. The mining activities by human being can increase the deposition and transportation of naturally occurring radioactive materials (NORM) in the groundwater and surface water bodies [1].

The supply of drinking water to large cities is generally ensured with surface water reservoirs, including artificial lakes built at main rivers. Groundwater provides a supplementary contribution to water supply in some regions. Worldwide, countries implement water treatment and water

quality control programs to ensure the distribution of potable drinking water to the population (WHO 2004). In this process, a wide number of water parameters can be easily and routinely controlled, such as acidity, turbidity, major ion concentrations, etc. while other parameters may require more specialized analysis, such as NORM and man-made radionuclides [2].

The main mission of Malian Radiation Protection Agency called (AMARAP) is to regulate any activity that can potentially cause radiological or nuclear damage. This can be NORM producing from mines (planned exposure situations) and legacy sites (existing exposure situations).

Currently, AMARAP has a regulatory standard in term of radionuclide concentrations in environmental samples, water and foodstuff.

The determination of radionuclide concentrations in drinking waters in Mali, the calculation of annual effective dose due to the ingestion of them, the calculation of risk assessment will permit to avoid any over exposition (determinist effects) and minimize as well the associated risk due to low doses (stochastic effects).

This study is the first national radiological analyze in drinking waters in Mali, it will be as a reference for AMARAP and others national authorities.

2. Materials and Methods

2.1. Sampling, Samples Preparations and Measurement

Twelve (12) samples of drinking water were collected from mall and shops around Bamako. Most of them are manufactured in Mali. For each sample, two samples were prepared for analyses: one for gross αβ total counting using Thermo-SCIENTIFIC RadEye HEC (serial n°00147) counter and one sample for gamma spectrometry counting using sodium iodine (NaI) detector coupled to the Genie 2000 software.

2.2. Calculations of Activity Concentration and Annual Committed Effective Dose

Activity concentration

After the counting time (1 hour) of gross αβ total, the results were expressed automatically in cps (count per second), which is egal to 1 Bq.

For gamma analysis, the activity concentrations of U-238 and Th-232 were assessed by averaging the peaks of different daughters for the U-238 and Th-232 series and K-40 were calculated using the photopeaks in the spectrum.

Therefore, the activity concentration ($A_{E,i}$) in Bq/l, for a radionuclide *i* with a photopeak at energy E, was calculated by equation (1).

$$A_{E,i} = \frac{N_{Ei}}{\varepsilon_E.t.\gamma_d.V} \quad (1)$$

where $N_{E,i}$ is the net peak-area of *i* radionuclide at energy E, ε_E is efficiency at energy E, t is counting time (s), γ_d is the gamma emission probability, and V is the volume (l)

Annual committed effective dose (AED)

The AED (Sv/year) was calculated using the activity concentration of all radionuclides detected in the samples, see equation (2).

$$AED = \Sigma e(g)_j . A_j . U_j \quad (2)$$

Where $e(g)_j$ is the effective dose conversion factor for ingestion of nuclide j (Sv/Bq), A_j is the activity concentration of nuclide j (Bq/kg) and U_j is the consumption rate (l/year or kg/year).

In this study, the international values of annual consumption rate were used [3].

Risk assessment (RA) calculation

Risk is estimated by the assumption that a linear dose-effect relationship has no threshold according to the IAEA and ICRP 60 [4]. Lower doses have a fatal cancer risk factor of 0.05 Sv^{-1} reported by International Atomic Energy Agency (IAEA) [5]. This risk factor determines the likelihood of an individual dying of cancer by a 5% increase of 1 Sv dose received throughout his entire life.

The Annual Committed Effective Dose, AED (Sv/year) in drinking water is estimated to determine the cancer risk of an adult using equation (3).

$$RA = AED.LE.RF \quad (3)$$

Where, LE is Life Expectancy in Mali which is 59.3years [6] and RF is the Risk Factor, its value is 0.05 Sv^{-1} [5] for low doses.

3. Results and Discussions

3.1. Results

Activity concentration Counting of gamma emitters radionuclides

Twelve (12) DW samples were analyzed, table 1 shows the activity concentrations of U-238, Th-232 and K-40. The minimum detectable activity (MDA) obtained for U-238, Th-232 and K-40 were 0.014, 0.056 and 0.052 Bq/l, respectively.

Table 1. *Results of gamma emitters counting.*

N°	Sample code	Brand	Activity Concentration of NORM (Bq/l)								
			U-238			Th-232			K-40		
1	EMin01	DRAAL	0.24	±	0.02	0.28	±	0.02	4.01	±	0.21
2	EMin02	HEPAR	0.49	±	0.03	< 0.056			4.23	±	0.23
3	EMin03	ASSINIE	4.96	±	0.25	4.37	±	0.22	0.44	±	0.03
4	EMin04	EVA	< 0.014			< 0.056			1.14	±	0.07
5	EMin05	CRISTALINE	9.42	±	0.48	5.54	±	0.28	< 0.052		

N°	Sample code	Brand	Activity Concentration of NORM (Bq/l)								
			U-238			Th-232			K-40		
6	EMin06	EVIAN	< 0.014			< 0.056			< 0.052		
7	EMin07	ARAWANE	< 0.014			< 0.056			1.85	±	0.1
8	EMin08	KATI	0.33	±	0.02	0.85	±	0.05	3.86	±	0.2
9	EMin09	OASIS	< 0.014			< 0.056			< 0.052		
10	EMin10	CULINAN	< 0.014			< 0.056			< 0.052		
11	EMin11	DIAGO	0.53	±	0.03	< 0.056			4.01	±	0.21
12	EMin12	TOMBOUCTOU	0.66	±	0.04	0.41	±	0.03	4.10	±	0.22
Malian standard [7]			1,000			1,000			100,000		

Counting of gross αβ total emitters

For this counting, the volume 50cc of each sample were prepared using hotplate and appropriate cup (geometry). This process was to evaporate the drinking water, the temperature of the hotplate was fixed to 70°C.

Twelve (12) DW samples were analyzed, the results in term of activity concentration (Bq/l) of gross αβ total emitters in each sample were expressed in table 2. Known the values in 50cc for drinking waters, the values in 1000cc were estimated.

Table 2. *Results of gross αβ total emitters counting.*

N°	Sample Code	Brand	αβ total Counter (cps or Bq)		
			αβ total	Value of αβ total in 50cc	Estimated Value in 1000cc (Bq/l)
Background			**0,80**		
1	EMin01	DRAAL	0,82	0,02	0,4
2	EMin02	HEPAR	0,82	0,02	0,4
3	EMin03	ASSINIE	0,80	0	0
4	EMin04	EVA	0,80	0	0
5	EMin05	CRISTALINE	0,92	0,12	2,4
6	EMin06	EVIAN	0,82	0,02	0,4
7	EMin07	ARAWANE	0,85	0,05	1
8	EMin08	KATI	0,84	0,04	0,8
9	EMin09	OASIS	0,83	0,03	0,6
10	Emin10	CULINAN	0,85	0,05	1
11	Emin11	DIAGO	0,85	0,05	1
12	Emin12	TOMBOUCTOU	0,86	0,06	1,2

Comparison of our obtained activity concentrations with others works around the world in UNSCEAR [8].

The comparison was carried out using the le mean values of U-238 and Th-232 in this work.

Table 3. *Comparison with others works around the world.*

Country	Sample	U-238	Th-232	Reference
Morocco	Tap water	0.0025 to 0.0157	-	[9]
USA	Potable water	0.0003 to 0.077	0.00005	[10]
Brazil	Underground water	0.00025 to 0.186	-	[11-12]
China	Potable water	0.0001 to 0.700	0.00004 to 0.012	[13]
Germany	Springs and wells	0.0004 to 0.600	0.0001 to 0.004	[14-15]
Finland	Groundwater	0.0005 to 1500000000	-	[16]
Czech Republic	Public supply	0.002 to 1.080	-	[17]
Mali	Mineral water	0.24 to 9.42	0.28 to 5.54	[This work]

3.2. Radiological Hazard Indices

Annual Committed Effective Dose (AED) due to the ingestion and Risk assessment (RA)

The AED were calculated for gamma emitters based on the conversion factor e(g) of detected radionuclides.

According to UNSCEAR [8], the annual intake of drinking water for adult is 500l/year (consumption rate). The AED was calculated using the values of conversion factor for ingestion of specific radionuclide e(g) of age > 17 years. See table 4 for more details.

The risk assessment (RA) was calculated using the mean value AED of gamma emitters in drinking waters.

Table 4. *Results of AED and RA.*

N°	Sample code	Brand	AED (µSv/y) for adult (Age >17a)
1	EMin01	DRAAL	50,04
2	EMin02	HEPAR	24,14
3	EMin03	ASSINIE	615,5
4	EMin04	EVA	3,53

N°	Sample code	Brand	AED (µSv/y) for adult (Age >17a)
5	EMin05	CRISTALINE	849,05
6	EMin06	EVIAN	-
7	EMin07	ARAWANE	5,74
8	EMin08	KATI	117,14
9	EMin09	OASIS	-
10	EMin10	CULINAN	-
11	EMin11	DIAGO	24,36
12	EMin12	TOMBOUCTOU	74,73
Mean values			196,03
Risk assessment (RA)			5,81E-04

3.3. Discussions

The range of activity concentrations for U-238 vary from 0.24 ±0.02 to 9.42 ± 0.48 Bq/l, for Th-232 from 0.28 ± 0.02 to 5.54 ± 0.28 Bq/l and for K-40 from 0.44± 0.03 to 4.23± 0.23 Bq/l.

The peak of U-238 wasn't detected in five samples (EMin04, EMin05, EMin06, EMin09 and Emin10), the peak of Th-232 wasn't detected in seven samples (EMin02, EMin04, EMin06, EMin07, EMin09, EMin10 and EMin11), the peak for K-40 wasn't detected in four samples (EMin05, 06, 09 and 10). More details are in table 1.

The lowest values of activity concentration for U-238 and Th-232 were reported in samples EMin01 and EMin04 for K-40. The highest values of U-238, Th-232 were reported in samples EMin05 and EMin02 for K-40. The results proved that the sample EMin05 (CRISTALINE) contained the maximum value of activity concentrations of NORM. This drinking water was imported from France. Special or more investigation must be carried out on it.

The highest value of activity concentration for gross αβ radionuclides was reported in samples Emin05 (CRISTALINE) 2,4 Bq/l. No values of gross αβ total were reported in two (02) samples (Emin03 and Emin04).

In general, the sample EMin05 (CRISTALINE) was the sample containing the highest value in term of activity concentration of gamma and gross αβ total radionuclides.

The activity concentrations in this work are much lower than the Malian standard which are: 1,000 Bq/l for U-238 and Th-232 and 100,000 Bq/l for K-40.

The comparison with others works revealed the obtained results in this work are also closed to some results carried out by some countries in drinking water, see table 3 for more details.

The AED from ingestion is calculated only for adult (age > 17years). The range of values for AED (µSv/y) of samples was from 3.53 to 849.05. The highest value of AED was reported again in EMin05. The mean value AED for ingestion of these drinking waters were around 0.20 mSv/year respectively for gamma and gross αβ total emitters. The sum of the both AEDs was around 0.95 mSv/year. See table 4 for more details.

The calculated RA using the sum of AEDs was 5.81 x 10^{-4} (see table 4). It is 10.32 times less than the risk (6.0 × 10^{-3}) from all natural ionizing radiation exposition based on the global average annual radiation dose to man (2.4 mSv.year^{-1}) [8].

ICRP 60 cancer risk is evaluated using annual dose limit of 1 mSv for the public, which gives an annual death probability of 10^{-5}, i.e., 1 in 100,000 reported by ICRP 60 [4]. That means the death probability by cancer due to the ingestion of drinking waters in this work is below than the estimated value reported by ICRP 60.

4. Conclusion

The NORM (U-238, Th-232 and K-40) were the detected radionuclides in this research. The analysis revealed that the results of activity concentrations of them in samples were lower than the national standard [7] and approximatively closed to the results of some countries. The AED was also under the authorized limit for the public (1 mSv/year). The mean radiological parameters such as Risk assessment (RA) and annual committed effective dose (AED) were under the dose criteria limits of international organizations (ICRP and UNSCEAR).

Based on the results in this study in term of radiological analysis, these drinking waters are safe for human consumption even if the risk (stochastic effect) associated with internal exposure due to low dose intakes exists. The obtained radiological hazard indices revealed that the probability of someone dying of cancer due to the ingestion of these waters is less than 10^{-5} in the Malian population.

Special attention and more analyzes must be done on the drinking water EMin05 (CRISTALINE) for more protection of public health.

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