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Annual effective dose estimation due to gross alpha and beta activities in groundwater samples from breweries in Nigeria

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Abstract

Background: Water makes up about 90 to 95 percent composition of beer products. The presence of natural radionuclides in groundwater samples used in Nigeria breweries' which spontaneously decay with the emission of gross alpha and beta activities ($G\alpha$ and β RAC) when ingested, may result in internal irradiation of the human body. It is therefore imperative to determine the potential radiation hazard (Annual effective dose).

Materials and Methods: Groundwater samples were obtained from fourteen (14) different brewery sites in Nigeria. Fifteen (15) ml/L of 1 mol/dm³ of concentrated Trioxonitrate (v) acid was added into each groundwater sample immediately after collection from the sources to preserve the radionuclides from container wall adsorption. Further sample preparation and analysis were carried out in the laboratory of Ghana Atomic Energy Commission, Accra using Method 900 of the US Environmental Protection Agency. Canberra iMatic™ Automatic low background gas-filled counter was used for determination of $G\alpha$ and β RAC in groundwater samples. IBM SPSS version 20 was used for statistical analysis. Annual Effective Dose (AED) in groundwater samples were estimated using a mathematical formula: $AED_{\alpha/\beta} (mSv y^{-1}) = A_{\alpha/\beta} \times IR_{w/b} \times CF$.

Results: Mean values for $G\alpha$ and β RAC in Nigeria breweries' groundwater samples were 0.370 ± 0.34 Bq/L and 0.193 ± 0.26 Bq/L respectively. Values for $G\alpha$ RAC were higher than the WHO recommended safe limits for most brewery sites whereas the $G\beta$ RAC were within the safe limit established by WHO. Highest AED value due to alpha emitting radionuclides in groundwater samples was recorded in Northern Nigeria (1.3240 mSv/y) while south-south region recorded the least value (0.6209 mSv/y) but were all above the WHO recommended safeguard of 0.1 mSv/y.

Conclusion: Results obtained from this study were above the recommended WHO safe level and therefore may pose radiological health hazards to consumers. Radioactivity water treatment plant as well as routine radiation monitoring are recommended for all breweries' groundwater sources in Nigeria.

Keywords: Effective dose, gross alpha/beta, groundwater, radionuclide and radioactivity

Introduction

Water is an indispensable integral part of our ecosystem and vital for human existence. It is well-known as universal solvent and makes up about 90-95% content of all ingredients used in breweries for brewing beers [1, 3]. Breweries in Nigeria are sited strategically at different geographical locations with possible variations in concentration of gross alpha and beta activities from radionuclides in their local environmental compositions such as groundwater [4, 5]. Indigenous researchers have discovered that most materials in our immediate vicinity such as the food and water we ingest all contain some natural radionuclides with variations in their activity concentrations [6, 9]. The presence and concentration of gross alpha and beta activities emanating from decay of radionuclides dissolved in groundwater we often ingest on regular bases varies geographically due to local geological variations of different environments [7, 4, 10].

Ingestion, absorption and deposition of radionuclides in groundwater inside the body tissue are often excreted via defecation, sweating or urination and are consistently replenished by ingestion of contaminated food and water [10, 12]. Alpha particle emitters (Uranium-238,

Radium-228, Thorium-228 and Polonium-210) and beta emitters (Radium-228, Potassium- 40 and Lead-210) in water, when ingested can pose a number of radiological health hazards such as cancer induction and deoxyribonucleic acid damage^[13, 14]. The hazardous effects of radionuclides in groundwater depend on the nature of the decay product, activity concentration and the extent of distribution when deposited in the tissue of consumers^[11, 19]. When alpha particle-emitting radionuclides are ingested via water or food sources, their average radiological effects are estimated to about twenty (20) times more hazardous than equivalent activity concentration of beta-emitting radionuclide. This is as a result of alpha particle's high relative biological effectiveness to cause radiobiological damages^[7, 15].

Human and animal studies have shown that no detrimental radiological health hazard such as incidence of carcinoma is preponderant from ingestion of groundwater when the radionuclide activity concentration is below the reference dose levels which is equal to committed effective dose value of 0.1mSv/year^[16, 18]. The globally recommended guideline activity concentrations for gross alpha and beta activities are 0.1 Bq/L and 1.0 Bq/L respectively^[17].

The National Agency for Food and Drug Administration and Control (NAFDAC) in Nigeria has formulated and enforced safety standards for different consumed products such as drugs, food and packaged water. However, these standards only laid emphasize the physico-chemical and microbial parameters without attention on radionuclide activity concentration^[2].

Consequently, there is great concern about the possible radiological health hazards to consumers of groundwater used as part of ingredients used in brewing beer products at different geographical locations in Nigeria, most especially in southern parts where consumption is regarded as part of socio-cultural norm. Currently, there is paucity of data on estimated annual effective dose due to ingestion of gross alpha and beta activities from radionuclides in groundwater samples used in different breweries at different geographical locations in Nigeria. This will create awareness and justify the need to purify radioactive groundwater sample in different breweries to guarantee safe low radiological hazards to consumers if necessary, thus the rationale for this study.

Materials and Methods.

Study locations: All registered and operating brewery in Nigeria which included:

1. Brewery A factories located in:

- Lagos State- Abebe village road, Iganmu, Lagos state, Industry Road
- Abia State- Ogbor hill industrial Layout, Obiangwu, Aba.
- Kaduna State-No1 Makera Road, industrial layout and No 1 Kudenda industrial area, Nnamdi Azikiwe Expressway Kaduna bypass.
- Enugu State- Amaeke in 9th Mile.
- Ogun State- Epe Road, Imagbon village Ijebu-ode and Km 38 Lagos Abeokuta Expressway Sango Ota both in Ogun state.
- Imo state- KM 24 Owerri/Onitsha road, Awo-Omama.

2. Brewery B factories located in:

- Lagos State- Km 24 Oba Akran Avenue.

- Edo State- 49 Oregbeni industrial Estate, Ikpoba Hill, Benin city.

3. Brewery C factories located in

- Osun state- Lawrence Omole Way, Omi-Asoro, Ilesa.
- Anambra State- SABMiller Drive, Harbour Industrial Layout, Onitsha.
- River State- 186/187 Trans-Amadi Industrial Layout, Oginigba, Port Harcourt

4. Breweries D factory located in

- Akwa-Ibom State- Industrial Layout Aka Offot, Uyo.

Ethical Clearance

Ethical approval was obtained in line with Helsinki declaration of 1964 from the research ethics committee of the Faculty of Health Sciences and Technology, Nnamdi Azikiwe University, Nnewi Campus.

Sample Size

- Eight (8) groundwater samples from brewery 'A' were obtained from eight (8) locations.
- Two (2) groundwater samples were obtained from two locations at breweries 'B' located.
- Three (3) groundwater samples were obtained from three locations at breweries 'C' located
- One (1) groundwater sample was obtained from brewery 'C' from one location.

Therefore the sample size was fourteen (14) groundwater samples^[15].

Sample collection

During sample collection and preparation, method 900.0 of the US Environmental Protection Agency was adopted as follow: Groundwater samples were collected directly into two liters (2 L) of plastic kegs (polyethylene containers) after being properly washing and rinsed with the water sample to be collected. About 15 ml/L validated sample of 1 mole per cubic decimeter (1 mol/dm³) of concentrated Trioxonitrate (V) acid (HNO₃) was added into the collected ground water samples at the point of collection using a syringe, gloves and a face-mask. The addition of concentrated Trioxonitrate (V) acid (HNO₃) served to preserve the radionuclides present in the water samples from adsorption to the inner walls of the containers. Prior to collection of groundwater samples, the taps was turned on at its full capacity for about three (3) minutes to purge the plumbing system of any water impurity and radon gas. The taps flow rate was subsequently reduced to attain steady turbulence and radon loss before collecting the water into the kegs^[15]. Groundwater samples were finally sent to the laboratory of Radiation Protection Institute (RPI), Ghana Atomic Energy Commission (GAEC), Kwabenya Accra, Ghana, where further preparation and analysis were carried out to determine the gross alpha (α) and beta (β) activity concentration.

Sample preparation.

Apparatus: The following apparatus were used for preparation of Groundwater sample:

Laboratory beakers, petri-dishes, hot plate, infra-radiator lamp, Experimental (Digital) weighing balance, planchet and cotton wool.

Reagents: Acetone and vinyl acetate.

Procedure: The beakers, petri-dishes, planchet and spatula were properly washed and rinsed with clean water. The apparatus were subsequently sterilized using acetone and dried inside an oven.

The beaker was rinsed twice with little quantity of groundwater sample to avoid cross contamination before evaporation. About one liter (1L) of groundwater sample was filtered on a filtration system set up and transferred into a one liter (1L) beaker. Two millimeter (2 ml) of HNO₃ was added to all the groundwater samples, allowed to stay overnight liberate metals and dissolve organic particles. For each groundwater sample, 300 ml of the filtrate was measured into Pyrex glassware and set on electric hot plate in a fume chamber at steady temperature below boiling point 600 - 700 for three hours. They are allowed to gradually evaporate to avoid excessive loss of the residue until a volume of 20-30 ml were obtained. The remaining filtrate was transferred into 47 mm stainless-steel planchet at 100C – 200 C. This process of heating continues until the volume of the groundwater samples were evaporated to dryness and placed in desiccator to prevent them from absorbing moisture and to cool down to room temperature before counting. This process is known as surface drying. Sample residues were dried and weighted using a weighing balance [15]. The weight of the total residue obtained from the total volume evaporated groundwater sample were calculated with the formula below:

$$W_r = W(d+s) - W_d \quad (3.1)^{[15]}$$

Where:

W (d+s) = Weight of the dish with sample's residue.

W_d = Weight of empty dish.

W_r = Weight of the total residue.

About 0.0770g of the residue will be added to the sterilized planchet and the exact volume that produced this required weight will be calculated by the use of the formula below:

$$0.0770g \times V_{tr} = W_{tr} \times V \quad (3.2)^{[15]}$$

Where:

$$\text{Activity } (\alpha) \text{ (Bq/L)} = \frac{\text{Net count (CPM)} (\alpha)}{DE \times 60 \times \text{Sample Size (Volume)} \times \text{Sample Efficiency}} \quad (3.4)^{[15]}$$

Where:

D.E = Detector's efficiency.

Net counts is given by:

Net counts = Raw counts (CPM) – Background (CPM).
..... (3.5)^[15].

$$\text{Activity } (\beta) \text{ (Bq/L)} = \frac{\text{Net count (CPM)} (\beta)}{DE \times 60 \times \text{Sample Size (Volume)} \times \text{Sample Efficiency}} \quad (3.6)^{[15]}$$

The parameters remain the same as for equation 3.5.

Determination of Gross alpha and beta activities in groundwater samples

The residues were counted for 200 minutes duration with regards to the procedure selected during the calibration of the instrument to investigate alpha/beta activity concentration as recommended by the World Health

V_{tr} = Volume that generated total residue,

W_{tr} = Weight of the total residue obtained

V = Volume that yielded the required residue

For sample efficiency to be 100%, residue obtained should be greater than or equal to 0.0770g. Samples with residue less than 0.0770g, its efficiency can be obtained using the expression below;

$$\text{Sample efficiency} = X \times 100 \% \dots \dots \dots (3.3)^{[15]}$$

Instrumentation and Calibration

Determination of gross alpha and beta activity concentration were carried out using low background automatic gross alpha/beta count system (Canberra IMatic™, USA). This system was calibrated with standard sources such as Americium-241 and Strontium-90. It was counted 10 minutes later to determine the efficiency of alpha and beta counter. The counting efficiency of beta and alpha counters were 31.01 % ± 2.18% and 69.01% ± 4.39% respectively. Americium-241, a standard calibration source has a higher alpha particle energy (5.49MeV) than those emitted by naturally occurring uranium. It is therefore the prescribed radionuclide for gross alpha calibration. Another calibration source, trontium-90 when at equilibrium with daughter Yttrium-90 are the correct radionuclides for gross beta calibration. Calibration was used to count the background radioactivity in the environment. The gross alpha and beta activity counting modes often operated at voltages of 1,600 and 1,700 volts respectively are employed to count the prepared groundwater samples. The counting system will be employed in counting clear empty planchet in all the counting modes to obtain the background radioactivity of the environment which is useful in the subsequent measurements [15].

Gross Alpha Counting

For gross alpha activity counting, voltage was set at 1600V and groundwater samples were counted for 3 cycles of 2700s (45mins) per cycle. The results were displayed as raw counts and count rate (count/min). The data were acquired for alpha mode and the specific activity for alpha in the samples were also calculated using the formula below.

Gross Beta Counting

The voltage for gross beta counting was set at 1700V and samples were counted for 3 cycles over a preset period of 2700s in beta mode. The specific activities were calculated using the formula below.

Organization [18]. All the samples were counted twice to ensure accuracy and validity before the results were recorded.

Annual Effective Dose Estimation (potential radiological hazards) in groundwater Sample

Annual effective dose estimation of gross alpha and beta activity emanating from groundwater sampled from

breweries in Nigeria were carried out with the aid of mathematical formula:

$$AED_{\alpha/\beta} (\text{mSv } y^{-1}) = A_{\alpha/\beta} \times IR_{w/b} \times CF \dots\dots 3.7^{[16]}$$

Where:

$A_{\alpha/\beta}$ = Gross alpha/beta activity concentration of the sample (Bq L^{-1}).

CF = Age- dependent effective dose factor.

$IR_{w \text{ or } b}$ = Annual ingested volume of drinking water per annum.

In this research work, $IR_{w \text{ or } b} = 730 \text{L } y^{-1}$ according to WHO for an adult person^[18].

Gross alpha activity concentrations in water are mainly due to emitter radionuclides such as U-238, U-234, Th-230, Th-232, Po-210 and Ra-226.

The gross beta activities were due to the following emitter radionuclides: Pb-210, Ra-228 and K-40.

Thus, we applied the annual dose conversion factors as follow:

- $CF_{U-238} = 4.5 \times 10^{-5} \text{ mSv Bq}^{-1}$ Gross alpha emitter.
- $CF_{U-234} = 4.9 \times 10^{-5} \text{ mSv Bq}^{-1}$ Gross alpha emitter.
- $CF_{Th-230} = 2.1 \times 10^{-4} \text{ mSv Bq}^{-1}$ Gross alpha emitter.
- $CF_{Th-232} = 2.1 \times 10^{-3} \text{ mSv Bq}^{-1}$ Gross alpha emitter.
- $CF_{Po-210} = 1.2 \times 10^{-3} \text{ mSv Bq}^{-1}$ Gross alpha emitter.
- $CF_{Ra-226} = 2.8 \times 10^{-4} \text{ mSv Bq}^{-1}$ Gross alpha emitter.

- $CF_{Pb-210} = 6.9 \times 10^{-4} \text{ mSv Bq}^{-1}$ Gross alpha emitter.
- $CF_{Ra-228} = 6.9 \times 10^{-4} \text{ mSv Bq}^{-1}$ Gross beta emitter.
- $CF_{K-40} = 6.2 \times 10^{-6} \text{ mSv Bq}^{-1}$ Gross beta emitter^[16].

The annual effective dose were estimated with the aid of above mathematical formula using respective conversion factors for each radionuclide and finally summing up the outcomes for either alpha or beta emitters in each sample being calculated^[8].

Data Analysis

Data obtained from sample analysis were saved on a computer Microsoft excel spread sheet and categorized into names of sample and name of breweries where samples were collected. The brewery locations and groundwater samples were categorized into site by site and regions in Nigeria such as South-east, South-west, Northern and South-south. They were analyzed using descriptive statistics (range, mean standard deviation, frequency, percentage, and tables). Data analysis were carried out with IBM, SPSS version 20.0 (IBM Corp. Armonk NY, 2011).

Results

Table one and two shows the gross alpha and beta activities in groundwater samples across several breweries in Nigeria.

Table 1: Gross alpha and beta activity from Groundwater samples across the various site of breweries in Nigeria.

Breweries water sites	Region of the Country	Alpha concentration in water (Bq/L)	Inference	Beta concentration in water (Bq/L)	Inference
		Mean	(α)	Mean	(β)
Ogun (Ilesha)	South-West	0.5220	Not Safe*	0.0083	Safe
Ogun (Ijebu Ode)	South-West	0.5600	Not Safe*	0.0103	Safe
Ogun (Sango Ota)	South-West	1.0000	Not Safe*	0.0604	Safe
Lagos (Oba Akan)	South-West	0.0030	Safe	0.0092	Safe
Lagos (Iganmu)	South-West	0.2230	Not Safe*	0.0500	Safe
Uyo (Aka Ufot)	South-West	0.5010	Not Safe*	0.0164	Safe
Kaduna (Kakuri)	North-West	0.2580	Not Safe*	0.5670	Safe
Kaduna (Kudenda)	North-West	0.6750	Not Safe*	0.4200	Safe
Benin(Ikpoba Hill)	South-South	0.1540	Not Safe*	0.6125	Safe
Port Harcourt (Oginigba)	South-South	0.0014	Safe	0.0064	Safe
Enugu (Ama)	South-East	0.2045	Not Safe*	0.0535	Safe
Anambra (Onitsha)	South-East	0.0404	Safe	0.6945	Safe
Imo (Awo-omama)	South-East	0.9530	Not Safe*	0.0629	Safe
Aba (Ogbo Hill)	South-East	0.0831	Safe	0.0916	Safe

*The recommended safe reference level activity concentration for ground water is 0.1 Bq/L for gross alpha^[18].

Most groundwater samples obtained from different brewery sites in Nigeria had higher alpha activity concentration which were above the safe recommended level in water. Highest value was obtained from the Ogun (Sango Ota) brewery site (1.0000 Bq/L) while the least value was recorded in Port Harcourt (0.0014 Bq/L). Gross beta activity

concentration from groundwater samples across all the sampled breweries were found to be within the recommended safe levels, with higher values recorded from Anambra (Onitsha) (0.6945 Bq/L) and the least value from Port Harcourt (0.0064 Bq/L), as shown in table 1.

Table 2: Annual effective dose estimation due to intake of radionuclides activities from groundwater samples from the various brewery sites in Nigeria

Breweries water sites	Alpha concentration in beer(Bq/L) Mean	Ann. Eff. Dose (α) (mSv/y)	Beta concentration in beer(Bq/L) Mean	Ann. Eff. Dose (β) (mSv/y)
Ogun (Ilesha)	0.5220	1.7430*	0.0083	0.0042
Ogun (Ijebu Ode)	0.5600	1.8700*	0.0103	0.0052
Kaduna (Kudenda)	0.6750	2.2540*	0.4200	0.2134*
Benin (Ikpoba Hill)	0.1540	0.5140*	0.6125	0.3113*
Enugu (Ama)	0.2045	0.6830*	0.0535	0.0272
Ogun (Sango Ota)	1.0000	3.3390*	0.0604	0.0307
Anambra (Onitsha)	0.0404	0.1349*	0.6945	0.3530*
Kaduna (Kakuri)	0.2580	0.8615*	0.5670	0.2882*
Lagos (Oba Akan)	0.0030	0.0100	0.0092	0.0005
Lagos (Iganmu)	0.2230	0.7446*	0.0500	0.0254
Port Harcourt (Oginigba)	0.0014	0.0047	0.0064	0.0033
Uyo (Aka-Ufot)	0.5010	1.6728*	0.0164	0.0083
Imo (Awo-omama)	0.9530	3.1921*	0.0629	0.0320
Aba (Ogbo Hill)	0.0831	0.2775*	0.0916	0.0466

*WHO recommended safe limit for total annual effective dose due to gross alpha and gross beta activity concentration is 0.1mSv per year for water^[18].

Annual Effective Dose Estimation due to intake of radionuclides from groundwater samples from brewery sites in Nigeria were calculated from gross alpha and beta activity concentration and presented on table 2. Brewery site in Nigeria with highest Estimated Annual Effective Dose from gross alpha activity concentration was Sango-Ota, Ogun state (3.3390 mSv/y) and brewery in Port Harcourt,

River state recorded the least value of annual effective dose (0.0014 mSv/y). Brewery site in Nigeria with highest estimated annual effective dose from gross beta activity concentration was Onitsha (0.3530 mSv/y) while the lowest dose from gross beta activity concentration was obtained from Oba Akan, Lagos state (0.0005 mSv/y) groundwater sample.

Table 3: Annual Effective Dose estimation due to intake of radionuclides in groundwater samples from different geographical areas in Nigeria

Geographical region	Number of Samples	Ann. Eff. Dose (a) (mSv/y)	Ann. Eff. Dose (b) (mSv/y)	Inference
Northwest	2	1.3240	2.5106	Not Safe*
South south	3	0.6209	1.1537	Not Safe*
Southeast	4	0.9073	1.1333	Not Safe*
Southwest	5	1.3099	1.4230	Not Safe*
Annual Average	14	1.0490	0.9760	Not Safe*

*WHO recommended safe limit for total annual effective dose estimation due to gross alpha and gross beta activity concentration is 0.1mSv per year for water^[18].

The estimated annual effective dose emanating from radionuclides from in groundwater samples across different locations in Nigeria were all above the safe annual recommended levels of 0.1mSv per year, with higher readings obtained from northern part of Nigeria (1.3240mSv/ year), as shown in table 3.

Discussion

The results obtained from this study revealed that the mean value of gross alpha activity concentration for most groundwater samples from brewery sites in Nigeria were above the recommended safe value of 0.1 Bq/L^[18, 15]. Similar findings were also reported in Poland by Skwarzec^[7], who found high radionuclides activity concentration in water samples obtained from breweries. These findings may not be unconnected with the geochemistry and the geological formation of the various locations where the groundwater samples were collected across different site locations in the country^[3, 11]. In most groundwater samples, the gross beta activity concentration were however within the recommended value of 0.1 Bq/L^[18, 20]. The inverse outcome obtained shows that gross beta activity may statistically pose little or no radiological hazard on the

consumers.

The brewery site in Nigeria with highest estimated annual effective dose from gross alpha activity concentration was Sango- Ota, Ogun state while the site with the least estimated annual effective doses was from Port Harcourt. Brewery site in Nigeria with highest estimated annual effective dose from gross beta activity concentration was Onitsha while the lowest activity was recorded from gross beta activity concentration found in groundwater sample from Oba Akan, Lagos. This could be connected to variation in concentration of radionuclides in our different environments such as in soil, rocks and water which primarily varies geographically due to local geological variations of different environment^[18, 19]; with the geochemistry and geophysics of such environments playing a vital part in radionuclides transition from the rock to water^[4, 10]. This shows that groundwater samples from the sampled areas poses radiological hazard to the consuming populace since they are above the recommended safe limit. The North-western breweries sites recorded higher estimated annual effective dose due to ingestion of groundwater containing gross alpha and beta emitting radionuclides, while lower concentrations were recorded in

the south-south breweries in Nigeria. The variations in the radionuclides concentration across the various geographical regions could be attributed to geological characteristics of water, rocks and mineral components as well as the geographical formation of these regions as studies have shown variations in activity concentration levels of alpha and beta activities at different altitude and geomorphic characteristics of different locations ^[13, 12].

Annual effective dose estimation due to gross alpha and beta radionuclide activity concentrations in groundwater samples collected at various brewery site locations were significantly high. The values were all higher than the WHO recommended safe value of 0.1mSv per annum for gross alpha and gross beta activities ^[18]. This shows that groundwater from all the breweries in Nigeria are not safe for consumption and consequently can poses radiological hazard to the consuming populace.

Conclusion

Estimated annual effective dose (AED) due to gross alpha activity concentration from groundwater samples used in different breweries at different geographical locations in Nigeria were generally not within the recommended safe limits at most brewery sites whereas the AED due to gross beta activity concentration were found to be within the recommended safe limits set by WHO for consumption.

Recommendation

In respect to linear no-threshold model, ion exchange water softeners and reverse osmosis system technologies should be employed for groundwater radioactivity treatment in all breweries especially in the northern part of Nigeria with strict enforcement by government health regulatory agencies to ensure low level radionuclide concentrations before channeling for brewing in other to reduce radiation hazards to consuming population.

Nigerian government should create a radiation monitoring unit under the Ministry of Health and National Agency for Food and Drug Administration and Control (NAFDAC) to routinely check the water samples used by breweries and table drinking drink factories to ensure safety of their products to human life as regard to radionuclide activity concentrations

Conflict of interest: None.

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