Journal of Materials and Environmental Science
ISSN : 2028-2508
e-ISSN : 2737-890X
CODEN : JMESCN

J. Mater. Environ. Sci., 2025, Volume 16, Issue 2, Page 282-290

http://www.jmaterenvironsci.com

Radiological Investigation of sachet packaged drinking water: A case study of products produced in Sokoto south local government area Sokoto, North west, Nigeria

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Received *25 Oct 2024,*
Revised *05 Feb 2025,*
Accepted *06 Feb 2025*

Keywords:
- ✓ *Radioactivity;*
- ✓ *Injection*
- ✓ *Water;*
- ✓ *Sachet Package Water;*
- ✓ *Gamma Spectrometry;*

Citation*: Omoteji I.B., Omoniyi F.O., Babalola O.A., Imalerio T.I., Amos A. (2025) Radiological Investigation of sachet packaged drinking water: A case study of products produced in Sokoto south local government area Sokoto, North west, Nigeria, J. Mater. Environ. Sci., 16(2), 282-290*

Abstract: In this paper, the radioactivity levels in sachet-packaged drinking water produced in Sokoto South, Nigeria were studied. The study found varying activity concentrations of radionuclides ^{226}Ra, ^{228}Ra, and ^{40}K across different samples. Specifically, ^{226}Ra levels ranged from 0.58 ± 0.20 Bq/L to 11.59 ± 3.20 Bq/L, while ^{228}Ra levels varied from 4.51 ± 2.08 Bq/L to 18.61 ± 2.30p Bq/L. The average values and standard deviation were 6.61±1.16 Bq/L and 12.19 ± 1.52 Bq/L, respectively. Additionally, ^{40}K levels ranged from 21.49±6.33 Bq/L to 142.08 ± 40.74 Bq/L, with an average value and standard deviation of 67.74 ± 14.18 Bq/L. Notably, ^{40}K was not detected in one sample, likely due to geographical or source-related factors. However, the mean AED's from ingesting these radionuclides in the water samples by adults, varied between ^{226}Ra and ^{228}Ra in the water sample varied 1.53 mSv to 9.44 mSv with an arithmetic mean ± standard deviation of 5.38 ± 2.08mSv, which are above the safety limits recommended by WHO, UNSCEAR, and ICRP. The ELCR's values indicate imminent cancer risk. This study suggests that, consuming these sachets water poses significant radiological health risks.

1. Introduction

Water is essential to human and the various activities that ensure a comfortable living. It serves as a natural solvent, facilitating the transport of pollutants (soluble or insoluble), microbes, heavy metals and radionuclides (Errich *et al*., 2021; Alaqarbeh *et al*., 2022; Fatima *et al*., 2024). In recent researches, naturally occurring radionuclides (norms), such as uranium, thorium, and potassium, have been detected in water sources meant for human consumption, raising concerns about their potential health impacts (Mansour *et al*., 2015; Ogedengbe and Oyelakin, 2019; Piñero-García *et al*., 2021). Furthermore, human activities, especially in industries like mining and mineral processing, often release artificially produced radionuclides into the environment, increasing contamination risks in water bodies. This highlights the importance of monitoring both natural and

industrial sources of radionuclides in water to protect public health and the environment (Ijeoma and Almayahi, 2020).

Over the past two decades, access to pipe-borne water in both urban and rural areas have deteriorated, forcing a growing reliance on unreliable water sources. In response, government authorities and international organizations like the World Health Organization (WHO) and Water Aid called for immediate action. These organizations emphasize the need to improve water infrastructure and enforce regulatory adherence to safety standards as critical steps towards achieving Sustainable Development Goal 6 by 2030, which aims to ensure universal access to clean water and sanitation (Adelodun *et al*., 2020). Though, sachet water has become a convenient solution for many Nigerians, the regulatory oversight by agencies like the National Agency for Food and Drug Administration and Control (NAFDAC) which primarily focuses on microbial and chemical aspects, often neglects the radiological safety of the water. This has raised public health concerns, with some sachet water products displaying unpleasant colors, odors, and tastes, indicating potential contamination (Adegoke *et al*., 2020).

Radionuclides, also known as radioactive isotopes or radioisotopes, are atoms with unstable nuclei that emit radiation during their decay processes. Cosmogenic radionuclides, including isotopes like carbon-14 and beryllium-7, are generated when cosmic rays collide with atoms in the atmosphere. These isotopes can enter aquatic systems through precipitation. Despite their presence in very small quantities, they play a critical role in understanding groundwater age and flow dynamics (Steinhauser, 2021). Primordial radionuclides, such as Uranium-238 (^{238}U), Thorium-232 (^{232}Th), and Potassium-40 (^{40}K), are key contributors to the natural radioactivity present in water sources (Arunima *et al*., 2021). These isotopes are remnants from Earth's formation and decay extremely slowly due to their extensive half-lives, often spanning millions to billions of years. For instance, ^{238}U undergoes decay, producing radioactive elements like Radium-226 (^{226}Ra), which can be detected in trace amounts in groundwater, posing potential radiation risks to accessible clean water (Alotaibi *et al*., 2024). Understanding these sources is crucial for evaluating potential health risks, especially in areas where natural radionuclide contamination in water is prevalent. Ionizing radiation, which includes alpha particles, beta particles, gamma rays, and X-rays, can cause a range of biological effects depending on the exposure's intensity and duration. These forms of radiation are energetic enough to ionize atoms or disrupt chemical bonds in biological tissues, leading to various health concerns (Brenner, 2021). Considering the potential health risks, it is vital to measure the radiological content in drinking water to prevent potential health hazards. Regular assessment helps mitigate risks associated with radiological contamination (Onyema *et al*., 2020). Stochastic effects which is health risks that arise by chance and typically appear long after exposure to radiation include cancer and genetic mutations, with their likelihood increasing as the radiation dose increases. Radiation exposure has been linked to cancer development many years after the initial exposure. Epidemiological studies, such as those involving atomic bomb survivors, have demonstrated a rise in cancer risk with higher radiation doses, particularly above 100 millisieverts (Boice, 1997). Non-stochastic, effects arise from acute, high-dose radiation exposure and occur only when a certain dose threshold is surpassed. Examples include skin burns, cataracts, and organ damage. These effects manifest when specific exposure levels are reached such as erythema after doses of 3 Gy or higher, temporary or permanent sterility at exposures over 0.15 Gy in males and 0.25 Gy in females. Unlike stochastic effects, the severity of deterministic effects worsens with increasing dose (Shah and Ghosh, 2020, Little and Dorr, 2021). Recent studies emphasize the risks associated with exposure to outdoor

ionizing radiation other than radionuclide concentrations in drinking water. This continuous assessment is essential to prevent excessive exposure and protect public health (Ugbede *et al*., 2021).

In this paper, we investigated naturally occurring radioactive materials (norms) presence in the consumed sachet water in Sokoto South, Nigeria, and the associated health risk indices.

2. Methodology

2.1 Preliminary Survey

A preliminary survey was undertaken to identify and document the widely consumed and locally produced sachet water brands in Sokoto South Local Government Area, in Figure 1. This survey involved direct interviews with manufacturers and consumers of sachet water. The results of the survey revealed that ten (10) sachet water brands are most commonly consumed in the local government area. These brands samples include SS_1, SS_2, SS_3, SS_4, SS_5, SS_6, SS_7, SS_8, SS_9 and SS_{10} water, as shown in Table 1. These brands were identified based on their popularity among local consumer.

2.2 Sample Collection

Ten brands of sachet-packaged water samples were obtained, representing ten brands from ten different producers in the Sokoto South Local Government Area of Sokoto State, Nigeria, with each brand highly demanded. Three samples were prepared from each brand and transferred into 1-liter containers, which were acidified with 10 mL of 1M Hydrochloric acid (HCl) per liter to prevent radionuclide binding to the container walls.

2.3 Sample Preparation

Ten non-radioactive, cylindrical plastic containers were provided to match the dimensions of standard sample containers. These containers were thoroughly washed, rinsed with diluted Sulfuric acid (H_2SO_4), and dried to prevent any contamination. Each container was labelled with the appropriate brand name and filled with a specific volume of acidified water sample. After sealing, the containers were stored for at least four weeks to ensure secular equilibrium between ^{226}Ra (from the ^{238}U series), ^{228}Ra (from the ^{232}Th series), ^{40}K, and their associated short-lived progeny (Dirican *et al*., 2024). In total, ten samples were set up for gamma spectrometry analysis.

2.4 Gamma Spectrometric Measurement

The activity concentration of water samples was evaluated using gamma spectrometry at the Centre for Energy Research and Development (CERT), Obafemi Awolowo University in Ile-Ife, Nigeria. The gamma spectrometric system utilized a 7.6 x 7.6 Sodium Iodide (NaI)- Titanium (Tl) scintillation detector (Model Bircom). This detector was integrated with several electronic components, including a pre-amplifier (Model 2001), an amplifier (Model 2020), and an Analog-to-Digital Converter (ADC) (Model 8075), all were connected to Canberra S100 Multi-Channel Analyzer (MCA). The quantification of the concentrations of radionuclides in the water samples, the specific channel numbers corresponding to gamma-ray energies was employed: 40K (1460 keV), ^{232}Th (2614.74 keV, via ^{208}Ti), and ^{226}Ra (1764.5 keV, via ^{215}Bi) (International Atomic Energy Agency (IAEA), 2020). Calibration was performed using a standard water sample (IAEA MBSS 197-92-16-1010, NO: 09-92). Each water sample was placed on the detector for 25,200 seconds to collect a spectrum featuring distinct peaks. Background counts were measured using an empty container of

identical geometry to the samples. Net counts for each sample and standard were computed by subtracting the background counts (IAEA, 2020). The block diagram of the spectrometry is shown in Figure 2.

Figure 1: Map of Sokoto state, showing the Sokoto South (blue color).

Table 1: Sample location and their Identification

Sample Location	Label
Sokoto South	SS_1
Emir Area	SS_2
Kalfu Area	SS_3
Bello Way	SS_4
Old Market	SS_5
Sokoto Stadium	SS_6
Ya'Rakija Area	SS_7
Sahara Area	SS_8
Hajia Halima Area	SS_9
Diploma Area	SS_{10}

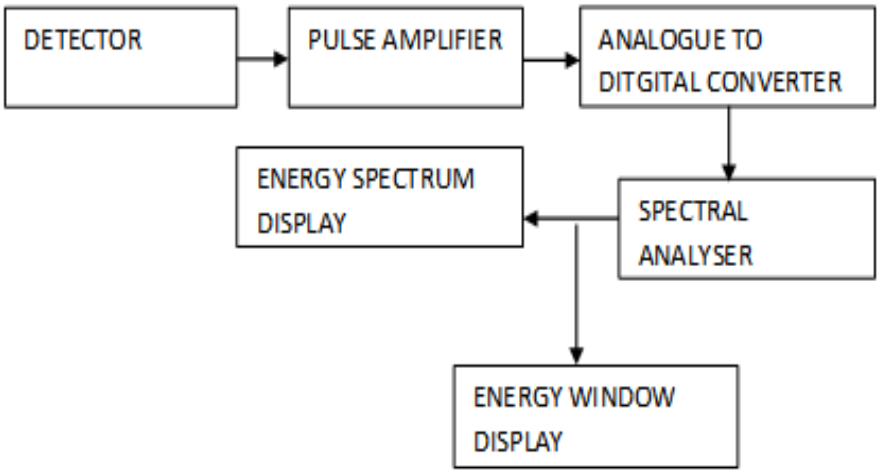

Figure 2: A block diagram of Spectrometry technique.

2.5 Activity Concentration

The activity concentrations of radionuclides in water samples were analyzed using the comparative method (Okedeyi *et al*., 2012). This method relates required parameters as:

$$\frac{A_S}{A_{SD}} = \frac{N_S}{N_{SD}} \qquad \text{Eqn. 1}$$

Where:

A_S = Activity concentration (Bq/L) of radionuclides in water samples
A_{SD} = Activity concentration (Bq/L) of radionuclides in standard sample
Ns = Net count rate under region of interest for each sample
N_{SD} = Net count rate under region of interest for standard sample

2.6 Annual Effective Dose (AED)

$$AED = \sum_{i=1}^{3} F(^{226}Ra, ^{228}Ra\ ^{40}K)A_s \times I \qquad \text{Eqn. 2}$$

Where F (^{226}Ra ^{228}Ra ^{40}K) is the dose conversion factor of the radionuclides presence in the water samples. The values are 2.8 x 10^{-7} SvBq^{-1} for ^{226}Ra, 2.3 x 10^{-7} SvBq^{-1} for ^{228}Ra and 5.0 x 10^{-9} SvBq^{-1} for ^{40}K (ICRP, 2020, UNSCEAR, 2020). The activity concentration of each of the radionuclides in the water samples and "*I*" is daily water consumption rate considered to be 2L/day or 730L/y.

The annual effective dose (AED) due to ingestion of ^{226}Ra, ^{228}Ra, and ^{40}K was estimated using:

$$AED\ (mSv/y) = F((^{226}Ra)\ A_s(^{226}Ra) + (\ ^{228}Ra)\ A_s(^{226}Ra) + (\ ^{40}K)\ A_s(^{40}K)) \times I \qquad \text{Eqn. 3}$$

2.7 Radium Equivalent Activity Index (Ra_{eq})

The Radium Equivalent Activity Index is estimated with Equation 4.

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \qquad \text{Eqn. 4}$$

where A_{Ra}, A_{Th} and A_k are the activity concentration of ^{226}Ra, ^{232}Th (^{228}Ra) and ^{40}K respectively. The maximum recommended value is 370Bql^{-1}for safe use building and other purposes. The mean value as estimated in our result is safe.

2.8 Radiation Hazard Indices

The internal and external radiation hazard (H_{in} and H_{ex} respectively) were estimated with equations 5 and 6 respectively.

$$\boldsymbol{H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}} \quad \text{Eqn. 5}$$

$$\boldsymbol{H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}} \quad \text{Eqn. 6}$$

Where H_{int} and H_{ext} must be less than unity for safety from respiratory diseases.

2.9 Excess Life Cancer Risk (ELCR)

The excess life cancer risk (ELCR) was evaluated with the formula in Equation 7.

$$\boldsymbol{ELCR = AED \times DL \times RF \times 1000} \quad \text{Eqn. 7}$$

Where AED is the annual effective dose, DL is the average duration of life (70 years) and RF is the risk factor. The ICRP RF value of public exposure is 0.05 for Stochastic effects. The 1000 is the conversion coefficient from Sv to mSv

2.10 Gamma Radiation Hazard Index (I_γ)

Gamma radiation hazard indices of the radionuclides in the water samples were determined using Equation 8.

$$\boldsymbol{I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}} \quad \text{Eqn. 8}$$

For safety, the value of I_γ must be less than or equal to unity ($I_\gamma \leq 1$).

3. Results and Discussion

The Table 2 shows significant variability that is influenced by the mineralogical and geological composition of the soil and rock in the environment of the obtained samples (Nguelem, 2013). Notably, ^{226}Ra activity concentration in all samples, except Sample SS_1, exceeds the 1.00 Bq/L limit set by IAEA (2020), consistent with previous studies in the region (Ajayi and Adesida, 2009). The computed ^{228}Ra activity concentration for all samples, surpasses the 0.1 Bq/L of safe drinking water recommended by WHO (2021). It is also higher than the reported values for adult by Ajayi and Adesida (2009) of 1.58 mS/y, and 93.99 µSv/y by Levi (2010). The AED from ^{226}Ra and ^{228}Ra ingestion ranges from 1.0021 mSv to 5.8863 mSv, with mean value of 3.6443 mSv which exceeds recommended doses by ICRP (2020), UNSCEAR (2020), and WHO (2021), which are 0.1 mSv/y, and 1 mSv/y respectively. The AED values are in unit multiples of standard values, and it signals hazard to consumers. The Figure 3 showed the bar chart of the samples with location SS_3 having the highest concentration of radionuclides.

Table 2: Activity Concentrations (Bq/L) and Annual Effective Doses (mSv/y) of Radionuclides in Sachet Water Samples.

Location of Sachet Water Samples	ACTIVITY CONCENTRATIONS(Bq/L)			AED (mSv/y)
	^{226}Ra	^{228}Ra	^{40}k	
SS_1	0.58 ± 0.20	4.51 ± 2.08	34.60 ± 9.86	1.0021
SS_2	5.89 ± 2.21	5.56 ± 1.87	48.39 ± 12.70	2.3141
SS_3	11.59 ± 3.20	18.61 ± 6.07	107.59 ± 33.97	5.8863
SS_4	10.60 ± 4.11	14.48 ± 2.30	63.03 ± 12.86	4.8279
SS_5	5.12 ± 2.01	15.20 ± 4.90	142.08 ± 40.74	4.1172
SS_6	8.46 ± 3.20	16.26 ± 4.66	21.49 ± 6.33	4.5377
SS_7	2.71 ± 1.17	12.18 ± 2.75	Nil	2.5989
SS_8	7.87 ± 2.55	10.62 ± 2.30	65.50 ± 10.52	3.6308
SS_9	3.57 ± 1.10	13.98 ± 5.10	75.02 ± 12.72	3.3508
SS_{10}	9.66 ± 2.22	10.52 ± 4.64	119.65 ± 41.06	4.1775
MEAN±SD	**6.61 ± 2.20**	**12.19 ± 3.67**	**67.74 ± 18.08**	**3.6443**

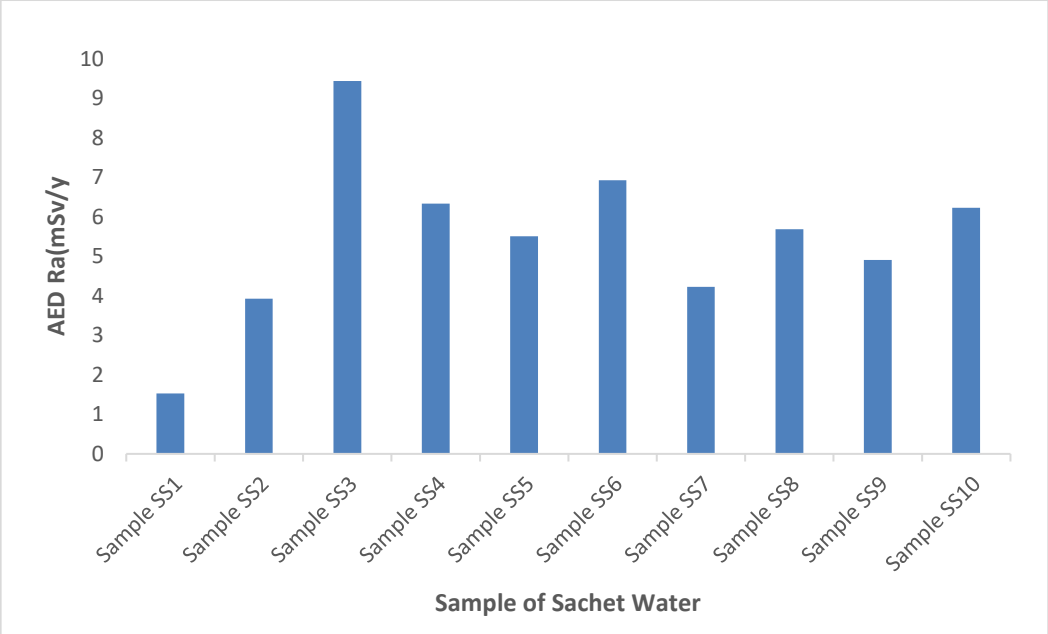

Figure 3: Graph of Annual effective dose in some sachet water in Sokoto South

The mean Ra_{eq} is less than the recommended value of 370 Bq/kg as well as the value for each water sample used, as shown in Table 3. The mean H_{int} and H_{ext} of the samples were estimated to be 0.0969 and 0.0790 respectively. The H_{ext} is higher than 0.465 reported by Levi (2010). The hazard indices (H_{int} and H_{ext}) values are less than unity and this implies that, the hazard effects of the radionuclides with their short life span, are negligible. Nevertheless, the ELCR values of all the samples tested were unit multiples of the recommended values by radiation regulatory bodies which is 0.2×10^{-3}. The mean ELCR index 12.7552 mS/y is higher than 1.012 mSv/y reported by Muhammad *et al.* (2024) for Hadejia town. It is high due to relatively high AED caused by ^{40}K radionuclide thus, poses cancer threat. The representative gamma index I_γ of 0.2111 Sv/y is less than unity, and this is within the safe limit.

Table 3. Radiation hazard parameters for water samples

Sample Location	AED (Adults) (mSv/y)	Ra_{eq}	H_{ex}	H_{in}	ELCR (mS/y)	I_γ
SS_1	1.0021	9.6935	0.0262	0.0277	3.5072	0.0720
SS_2	2.3141	17.5668	0.0474	0.0634	8.0992	0.1271
SS_3	5.8863	46.4867	0.1255	0.1569	20.6021	0.3351
SS_4	4.8279	36.1597	0.0977	0.1263	16.8976	0.2575
SS_5	4.1172	37.7962	0.1021	0.1159	14.4102	0.2809
SS_6	4.5377	33.3665	0.0901	0.1130	15.8820	0.2333
SS_7	2.5989	20.1274	0.0544	0.0617	9.0963	0.1399
SS_8	3.6308	28.1001	0.0759	0.0972	12.7078	0.2023
SS_9	3.3508	29.3379	0.0792	0.0889	11.7277	0.2136
SS_{10}	4.1775	33.9167	0.0916	0.1177	14.6214	0.2494
Mean	**3.6443**	**29.2552**	**0.0790**	**0.0969**	**12.7552**	**0.2111**

Conclusion

The investigation the radioactivity level in sachet-packaged drinking water in Sokoto South, Nigeria was successfully done. The results revealed significant variations in radionuclide activity concentrations (^{226}Ra, ^{228}Ra, and ^{40}K) among different brands. The annual effective doses due to ingestion of ^{226}Ra and ^{228}Ra exceeded recommended limits set by WHO, UNSCEAR, and ICRP. The AED values were elevated by high ^{40}K. The high AED's was set by high ^{40}K value and the ELCR's values revealed imminent cancer and safety from ionizing radiation. Nevertheless, the gamma index (I_γ) was in the safe limit.

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