

## ASSESSMENT OF NATURAL RADIOACTIVITY IN SACHET DRINKING-WATER SAMPLES IN NIGERIA, WEST AFRICA

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**Abstract.** *An assessment of the natural radioactivity in sachet drinking-water samples in Ilorin, Nigeria has been carried out. Gamma-ray spectroscopic analysis of the samples revealed the presence of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  radionuclides in high proportions. The activity concentration values range from  $174.20 \pm 34.19$  to  $376.02 \pm 65.03 \text{ Bq l}^{-1}$ ,  $9.36 \pm 2.87$  to  $22.52 \pm 6.20 \text{ Bq l}^{-1}$  and  $9.85 \pm 3.88$  to  $23.88 \pm 7.47 \text{ Bq l}^{-1}$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  respectively. The derived annual effective dose received by the population as a result of the ingestion of  $^{40}\text{K}$  was found to range from  $0.39 \pm 0.08$  to  $0.85 \pm 0.15 \text{ mSv y}^{-1}$ ,  $0.96 \pm 0.38$  to  $2.30 \pm 0.63 \text{ mSv y}^{-1}$  for  $^{226}\text{Ra}$  and  $2.48 \pm 0.98$  to  $6.01 \pm 1.88 \text{ mSv y}^{-1}$  for  $^{228}\text{Ra}$ . Consequently, the effective dose received as a result of the combined ingestion of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  was found to range from 1.27 to  $2.32 \text{ mSv y}^{-1}$ . These contributions of both  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  activities to the committed effective dose from a year's consumption of the drinking-water are higher than the tolerable level of  $1 \text{ mSv y}^{-1}$  to the general public for prolonged exposure as recommended by ICRP, and much more than WHO's recommended level of  $0.1 \text{ mSv y}^{-1}$  for drinking-water. Therefore, it is strongly recommended that calculated efforts should be made by relevant regulatory authorities in Nigeria to protect the populace from adverse radiological health implications.*

**Key words:** radioactivity, drinking-water, gamma spectroscopy, health and Nigeria

### 1. INTRODUCTION

In Nigeria today, more than 70 million (about 42% of the population) people do not have access to potable water, with most of those affected living in rural areas (WHO/UNICEF Joint Monitoring Program, 2013). Many communities meet their daily need for water from polluted and distant rivers, lakes or reservoirs. Recently, the Nigerian Government decried the low access to potable water by Nigerians, which has resulted in a high mortality rate from water borne diseases such as cholera, typhoid, diarrhoea and river blindness. Water borne diseases are among the leading causes of illness and death in

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most developing countries (Reiff, 1995). Estimates by World Health Organization (WHO) in 1996, indicate that as much as 80% of all diseases in the world is associated with water. As a result, the government solicited for actions that would increase access to water supplies in the country in line with the Millennium Development Goals. Since the local water administration is untiringly struggling to meet the increasing daily domestic and industrial demand for water; many homes, organizations and communities now pump their required quantities of water from the ground. Notably, the society has therefore taken to several adaptive measures of alleviating stress resulting from the lack of public drinking water, such as the proliferation of very affordable and easy to serve portable drinking-water in 50cl cellophane sachets. As a result of high demand and minimal cost of production, the market for this packaged water is witnessing tremendous growth, particularly among the lower income populace. Presently it is estimated that about 70 percent of the Nigerian populace live on less than one dollar (US \$1.0) per day (Oyekanmi, 2011). Therefore, there are serious concerns on the quality of these sachet waters in order to protect the vulnerable public.

Most of the packaged water is sourced from groundwater and recent studies in North-Central Nigeria showed that groundwater in some areas may have high concentrations of radium isotopes (Nwankwo, 2013). Radium has four isotopes,  $^{226}\text{Ra}$  derived from the  $^{228}\text{U}$  decay chain,  $^{228}\text{Ra}$  and  $^{224}\text{Ra}$  that are part of the  $^{232}\text{Th}$  decay chain, and  $^{223}\text{Ra}$  from  $^{235}\text{U}$  decay chain (Venngosh et al, 2009). These isotopes are carcinogenic (USEPA, 2005) and therefore their occurrence in drinking-water above recommended reference level may increase the long-term incidence of cancer. Moreover, long-term exposure may cause toxic effects to the kidneys (Colorado Department of Public Health and Environment, 2013). However, no deleterious radiological health effects are expected from consumption of drinking-water if the concentrations of radionuclides are below the guidance levels (WHO, 2011). It is therefore vital to consider and, when justified, take remedial action to reduce dose if found to be more than international drinking-water standards.

In this paper, we have presented the data of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  radionuclides measured in major sachet drinking-water samples produced in Ilorin, north-central Nigeria. Similar work done in south-western Nigeria showed that activity concentrations of radium isotopes in selected sachet water samples exceed health standards (Ajayi and Adesida, 2009). Therefore, this work would ascertain whether the level of radioactivity in cellophane sachet drinking-water in Ilorin, north-central Nigeria could pose any significant health hazard to the populace. This would also contribute intelligently to the safety discourse of such packaged water from radiological point of view.

## 2. EXPERIMENTAL PROCEDURE

Cellophane sachet drinking-water samples were obtained from 15 popular vendors within Ilorin metropolis, Nigeria, West Africa (Figure 1) in May 2013. The activity concentrations of the water samples were measured using high-purity germanium (HPGe) detector coupled with a Canberra series 10 plus Multichannel Analyzer (MCA). Environmental shielding of the water was achieved using a Canberra 10cm thick lead castle while counting was done for 10 hours because of the low natural activities of radionuclides in water. The observed spectrum was measured and the area under the photo-peaks was calculated using the algorithm of the MCA. Noticeable photo-peaks observed in the spectra of the samples were evaluated by matching gamma energies at

various energy levels to a library of possible isotopes. Consequently, the radionuclides in the natural decay series of  $^{238}\text{U}$  and  $^{232}\text{Th}$ , and the non-series  $^{40}\text{K}$  were identified. The  $^{226}\text{Ra}$  activity of the samples was determined through the intensities of 351.9 and 609.3 keV gamma-lines of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  respectively, while  $^{228}\text{Ra}$  activity was obtained through the gamma line of  $^{228}\text{Ac}$  at 911.21 keV. The activity of the non-series  $^{40}\text{K}$  was also determined directly from its 1460.8 keV gamma line (Ahmed, 2004; Avwiri et al, 2007; Nasirian et al., 2008; Lydie and Nemba, 2009; Nwankwo, 2010; 2012; 2013).



**Fig. 1** Map of Nigeria showing Ilorin City.

The radionuclide concentration  $C$  and the resulting annual effective dose AED in each water sample were evaluated using the following relations (Lydie and Nemba, 2009):

$$C = \frac{N(E_y)}{\varepsilon(E_y) \cdot I_y \cdot V \cdot t_c} \quad (1)$$

where  $N(E_y)$  is the net peak area of the radionuclide of interest,  $\varepsilon(E_y)$  is the efficiency of the detector for the energy  $E_y$ ,  $I_y$  is the intensity per decay for the energy  $E_y$ ,  $V$  is the volume of the water sample and  $t_c$  is the total counting time in second (36000 s).

$$AED = \sum_i I_i \cdot 365 \cdot D_i \quad (2)$$

where  $I_i$  is the daily intakes of radionuclide  $I$  ( $\text{Bq d}^{-1}$ ) and the ingestion dose coefficient  $D_i$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  is  $6.2 \times 10^{-9}$ ,  $2.8 \times 10^{-7}$  and  $6.9 \times 10^{-7}$   $\text{SvB q}^{-1}$  respectively (ICRP, 1994).

## 3. RESULTS AND DISCUSSION

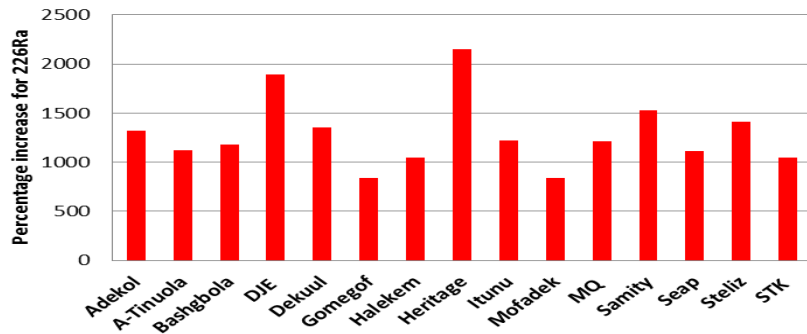
The radionuclides identified in the water samples and quantified from the gamma ray spectra are  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  (from  $^{238}\text{U}$  decay series) and  $^{228}\text{Ra}$  ( $^{232}\text{Th}$  decay series). The activity concentration values range from  $174.20 \pm 34.19$  to  $376.02 \pm 65.03 \text{ Bq l}^{-1}$ ,  $9.36 \pm 2.87$  to  $22.52 \pm 6.20 \text{ Bq l}^{-1}$  and  $9.85 \pm 3.88$  to  $23.88 \pm 7.47 \text{ Bq l}^{-1}$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  respectively. The entire activity of naturally occurring radionuclides for the 15 samples is shown in Table 1. The table shows that Gomegof sachet water has the lowest radionuclide concentrations of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . The highest concentrations of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are found in Steliz, Heritage and Samity sachet water respectively. However, potassium is an essential element in the human body and is absorbed mainly from ingested food.  $^{40}\text{K}$  does not accumulate in the body but is maintained at a constant level independent of intake (WHO, 2011), therefore the ingestion of  $^{40}\text{K}$  may not be harmful to the body since it has little effect on the body content or on the radiation dose received (NCRP, 1967). Consequently,  $^{40}\text{K}$  is not used in the computation of committed effective dose received by the ingestion of the sampled water in this study. Nevertheless, the accepted guidance levels for radionuclide activity concentration of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in drinking-water are 1.0 and 0.1  $\text{Bq l}^{-1}$  respectively (WHO, 2011). This shows that the activity concentration of the radionuclides in the sampled drinking-water extremely exceeds these guidance levels as illustrated in Figures 2 and 3.

**Table 1** The mean specific activity concentration of radionuclides detected in the drinking-water samples

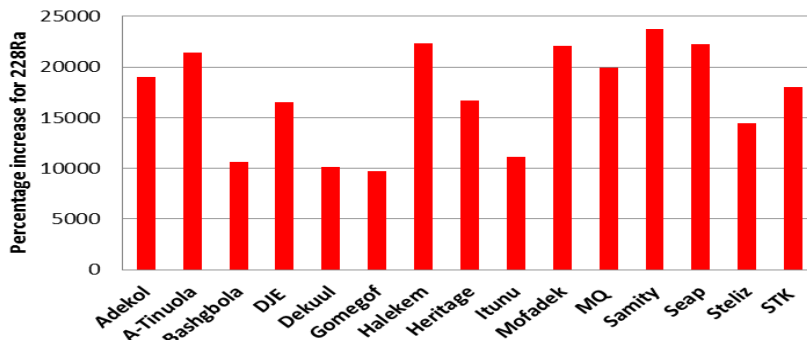
Sample	$^{40}\text{K}$ ( $\text{Bq l}^{-1}$ )	$^{226}\text{Ra}$ ( $\text{Bq l}^{-1}$ )	$^{228}\text{Ra}$ ( $\text{Bq l}^{-1}$ )
Adekol	$233.12 \pm 63.23$	$14.20 \pm 4.82$	$19.14 \pm 6.11$
A-Tinuola	$301.79 \pm 71.18$	$12.26 \pm 3.72$	$21.53 \pm 5.99$
Bashgbola	$253.18 \pm 66.10$	$12.81 \pm 4.31$	$10.74 \pm 4.81$
DJE	$235.56 \pm 43.06$	$19.92 \pm 5.75$	$16.63 \pm 5.01$
Dekuul	$281.05 \pm 47.21$	$14.54 \pm 5.05$	$10.23 \pm 3.98$
Gomegof	$174.20 \pm 34.19$	$9.36 \pm 2.87$	$9.85 \pm 3.88$
Halekem	$250.62 \pm 45.16$	$11.50 \pm 3.64$	$22.48 \pm 6.41$
Heritage	$213.50 \pm 37.89$	$22.52 \pm 6.20$	$16.76 \pm 5.52$
Itunu	$202.82 \pm 47.26$	$13.24 \pm 4.12$	$11.22 \pm 4.01$
Mofadek	$247.87 \pm 68.23$	$9.42 \pm 3.71$	$22.18 \pm 5.95$
MQ	$266.55 \pm 53.21$	$13.16 \pm 5.41$	$20.03 \pm 6.56$
Samity	$257.74 \pm 64.52$	$16.29 \pm 4.30$	$23.88 \pm 7.47$
Seap	$178.26 \pm 39.48$	$12.13 \pm 3.62$	$22.33 \pm 6.33$
Steliz	$376.02 \pm 65.03$	$15.10 \pm 4.34$	$14.54 \pm 5.45$
STK	$176.14 \pm 49.19$	$11.44 \pm 3.21$	$18.16 \pm 4.88$
Range	174.20 - 376.02	9.36 - 22.52	9.85 - 23.88
Mean	$243.23 \pm 51.37$	$13.86 \pm 3.45$	$217.31 \pm 4.78$

Assuming the volume of drinking-water for adult to be 1 litre per day (Ahmed, 2004; Lydie and Nemba, 2009), the daily intake per person of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  through the consumption of the sachet water and the derived annual effective dose are presented in Table 2. The effective doses received by the population as a result of the ingestion of  $^{226}\text{Ra}$  was found to range from  $0.96 \pm 0.38$  to  $2.30 \pm 0.63 \text{ mSv y}^{-1}$  and  $2.48 \pm 0.98$  to

6.01±1.88 mSv y<sup>-1</sup> for <sup>228</sup>Ra. The annual effective dose received as a result of the combined ingestion of <sup>226</sup>Ra and <sup>228</sup>Ra is consequently found to range from 1.27 to 2.32 mSv y<sup>-1</sup> as shown in Figure 4. The figure shows that, expectedly, Gomegof sachet water has the lowest effective dose while Samity has the highest effective dose. Two samples (Gomegof and Itunu) have effective doses less than 1.5 mSv y<sup>-1</sup>, while four samples (Adkol, Heritage, MQ and Samity) have more than 2 mSv y<sup>-1</sup>.



**Fig. 2** Percentage increase of activity concentration above guidance levels for 226Ra in the drinking-water samples.



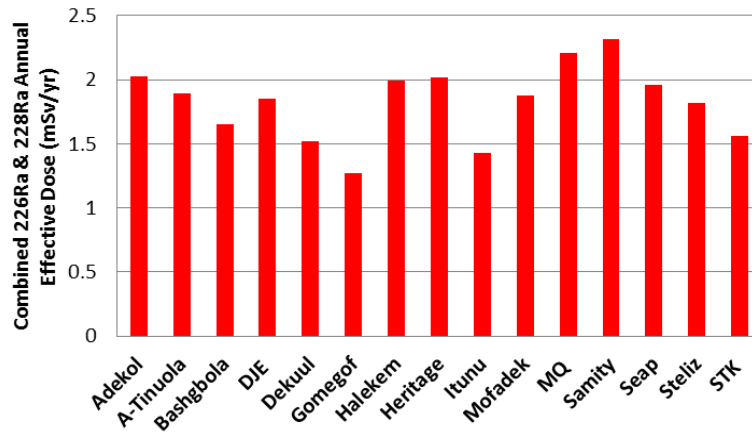
**Fig. 3** Percentage increase of activity concentration above guidance levels for 228Ra in the drinking-water samples.

Figure 5 shows that in the area under investigation, the effective dose received as a result of the combined ingestion of <sup>226</sup>Ra and <sup>228</sup>Ra from each of the sampled sachet water are above tolerable limits. According to ICRP recommendation (ICRP, 1991), the public should not be exposed to more than an average of 1 mSv y<sup>-1</sup>, while in WHO's recent publication, 0.1 mSv y<sup>-1</sup> (WHO, 2011) is recommended for drinking-water. Therefore, the doses from consumed sachet drinking-water exceed ICRP recommended safe level by 27 to 132% and WHO by 1,170 to 2,220%, as shown in Figures 6 and 7. These outcomes have revealed that the contributions of both <sup>226</sup>Ra and <sup>228</sup>Ra activities to the committed effective dose from a year's consumption of the drinking-water from all the samples are higher than the tolerable level of 1 mSv y<sup>-1</sup> to the general public for prolonged exposure as recommended

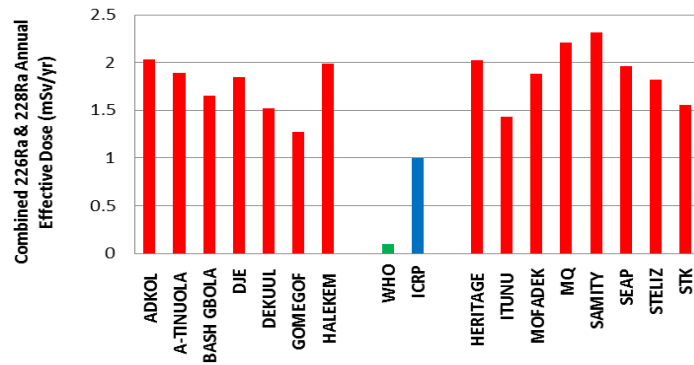
by ICRP (ICRP, 1991) and extremely more than WHO's recommended level of  $0.1 \text{ mSv y}^{-1}$  (WHO, 2011) for drinking-water. The average annual effective dose received by the populations as a result of the combined ingestion of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in sachet drinking-water therefore exceeds health standards significantly in north-central Nigeria.

**Table 2** Daily intake of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  and the estimated Annual Effective Doses from the water samples

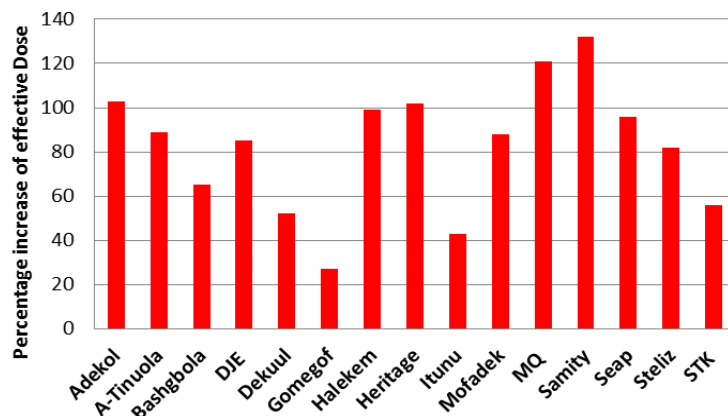
Sample	Intake per person ( $\text{Bq d}^{-1}$ )		Annual Effective Dose ( $\text{mSv y}^{-1}$ )	
	$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{226}\text{Ra}$	$^{228}\text{Ra}$
Adekol	14.20±4.82	19.14±6.11	1.45±0.49	4.82±1.54
A-Tinuola	12.26±3.72	21.53±5.99	1.25±0.38	5.42±1.51
Bashgbola	12.81± 4.31	10.74±4.81	1.31±0.44	2.70±1.21
DJE	19.92±5.75	16.63±5.01	2.04±0.59	4.19±1.26
Dekuul	14.54±5.05	10.23±3.98	1.49±0.52	2.58±1.00
Gomegof	9.36±2.87	9.85±3.88	0.96±0.29	2.48±0.98
Halekem	11.50±3.64	22.48±6.41	1.18±0.37	5.66±1.61
Heritage	22.52±6.20	16.76± 5.52	2.30±0.63	4.22±1.39
Itunu	13.24±4.12	11.22±4.01	1.35±0.42	2.83±1.01
Mofadek	9.42±3.71	22.18±5.95	0.96±0.38	5.59±1.50
MQ	13.16±5.41	20.03±6.56	1.34±0.55	5.04±1.65
Samity	16.29± 4.30	23.88±7.47	1.66±0.44	6.01±1.88
Seap	12.13±3.62	22.33±6.33	1.24±0.37	5.62±1.59
Steliz	15.10±4.34	14.54±5.45	1.54±0.44	3.66±1.37
STK	11.44±3.21	18.16±4.88	1.17±0.33	4.57±1.23
Range	9.36 - 22.52	9.85 - 23.88	0.96 - 2.30	2.48 - 6.01
Mean	13.86±3.45	217.31±4.78	1.42±0.35	4.36±1.20



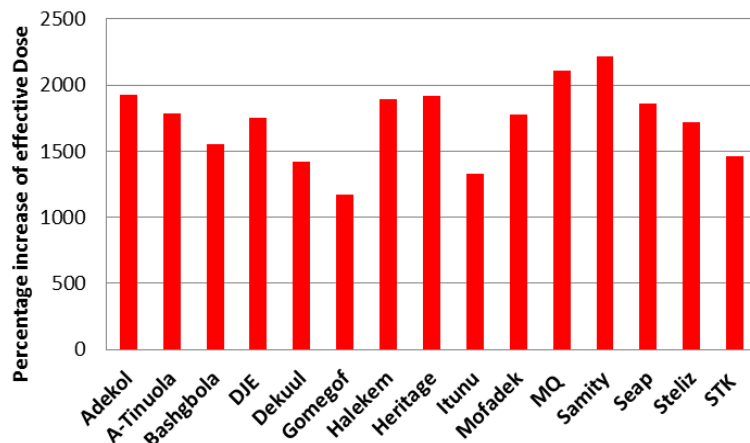
**Fig. 4** Combined ( $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ) annual effective doses for the drinking-water samples.



**Fig. 5** Combined (226Ra and 228Ra) annual effective doses for the drinking-water samples compared with international radiological standards.



**Fig. 6** Percentage increase of committed effective dose above ICRP limit of 1 mSv/year from combined 226Ra and 228Ra activities in the drinking-water samples.



**Fig. 7** Percentage increase of committed effective dose above WHO limit of 0.1 mSv/year from combined 226Ra and 228Ra activities in the drinking-water samples.

#### 4. CONCLUSION

A study of the radioactivity in some cellophane packaged drinking-water samples produced in Nigeria has been carried out. Gamma-ray spectroscopic analyses of the samples revealed the presence of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  (U-series) and  $^{228}\text{Ra}$  (Th-series) radionuclides in very high proportions. The measured activity concentration values range from  $174.20 \pm 34.19$  to  $376.02 \pm 65.03$   $\text{Bq l}^{-1}$ ,  $9.36 \pm 2.87$  to  $22.52 \pm 6.20$   $\text{Bq l}^{-1}$  and  $9.85 \pm 3.88$  to  $23.88 \pm 7.47$   $\text{Bq l}^{-1}$  for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  respectively. The derived annual effective dose received by the population as a result of the ingestion of  $^{226}\text{Ra}$  was found to range from  $0.96 \pm 0.38$  to  $2.30 \pm 0.63$   $\text{mSv y}^{-1}$  and  $2.48 \pm 0.98$  to  $6.01 \pm 1.88$   $\text{mSv y}^{-1}$  for  $^{228}\text{Ra}$ . Consequently, the effective dose received as a result of the combined ingestion of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  was found to range from 1.27 to 2.32  $\text{mSv y}^{-1}$ . These contributions of both  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  activities to the committed effective dose from a year's consumption of the drinking-water are higher than the tolerable level of  $1 \text{ mSv y}^{-1}$  to the general public for prolonged exposure as recommended by ICRP, and much more than WHO's recommended level of  $0.1 \text{ mSv y}^{-1}$  for drinking-water. The mean contribution of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  activities to the committed effective dose from a year's consumption of the sachet drinking-water in the study is therefore up to 132% higher than the tolerable level of  $1 \text{ mSv y}^{-1}$  to the general public for prolonged exposure as recommended by ICRP, and more than 2,000% above the WHO recommended level of  $0.1 \text{ mSv y}^{-1}$  for drinking-water.

This study shows that people may suffer substantial internal exposure from these sampled sachet drinking-waters, therefore it may not be radiologically safe to consume the cellophane packaged drinking-water in the study area since they exceed health standards. The populace face the risk of some health effects that may result from the significant accumulation of radium in their bones and other vulnerable or radiosensitive soft body tissues. This raises concerns about the radiological safety of sachet drinking-water in Nigeria; therefore, it is strongly recommended that calculated efforts should be made by relevant regulatory authorities in Nigeria to protect the populace from adverse radiological health implications.

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## **PROCENA PRIRODNE RADIOAKTIVNOSTI U UZORCIMA PIJEĆE VODE U NIGERIJ; ZAPADNA AFRIKA**

*Procena prirodne radioaktivnosti u uzorcima pijaće vode je izvršena u Ilorin, Nigeriji. Spectroscopska analiza gama zraka uzoraka otkrila je prisustvo  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  i  $^{228}\text{Ra}$  radionuklida u visokim proporcijama. Koncentracije vrednosti aktivnosti u rasponu od  $174.20 \pm 34.19$  do  $376.02 \pm 65.03 \text{ Bk l}^{-1}$ ,  $9.36 \pm 2.87$  do  $22.52 \pm 6.20 \text{ Bk l}^{-1}$  i  $9.85 \pm 3.88$  do  $23.88 \pm 7.47 \text{ Bk l}^{-1}$  za  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  i  $^{228}\text{Ra}$  respektivno. Izvedena godišnja efektivna doza koju primi stanovništvo kao rezultat uzimanja  $^{40}\text{K}$  pronađena je u rasponu od  $0.39$  do  $0.08 \pm 0.85 \pm 0.15 \text{ mSv y}^{-1}$ ,  $0.96 \pm 0.38$  do  $2.30 \pm 0.63 \text{ mSv y}^{-1}$  za  $^{226}\text{Ra}$  i  $2.48 \pm 0.98$  do  $6.01 \pm 1.88 \text{ mSv y}^{-1}$  za  $^{228}\text{Ra}$ . Shodno tome, nađeno je da efektivna doza dobijena kao rezultat kombinovanog uzimanja  $^{226}\text{Ra}$  i  $^{228}\text{Ra}$  je u rasponu od  $1.27$  do  $2.32 \text{ mSv y}^{-1}$ . Ovi doprinosi obe  $^{226}\text{Ra}$  i  $^{228}\text{Ra}$  aktivnosti na učinjenoj efektivnoj dozi iz potrošnje pijaće vode za godinu dana su veći od podnošljivog nivoa od  $1 \text{ mSv y}^{-1}$  za javnost za produženo izlaganje po preporuci ICRP, i mnogo veće od preporučenog nivoa WHO koji iznosi  $0.1 \text{ mSv y}^{-1}$  za pijaću vodu. Zbog toga se preporučuje da se ulože napori od strane nadležnih regulatornih organa u Nigeriji da zaštite stanovništvo od negativnih implikacija radiološke zaštite.*

**Ključne reči:** radioaktivnost, pijaća voda, spektroskopija gama zraka, zdravlje i Nigerija