

# Natural Radioactivity in Some Selected Powdered Milk Brands Consumed in Nigeria and Corresponding Annual Effective Dose

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## Abstract

*Regulation and monitoring of radionuclides in foodstuff is important for human health maintenance. Powdered milk is rich in minerals and protein contents which are essential for healthy growth and development. Five (5) brands of powdered milk commonly consumed in Nigeria were collected and analysed for their natural radioactivity content. The analysis was carried out using gamma ray spectrometry system consisting of a 3×3 inch NaI(Tl) detector. Results showed that the activity concentration of <sup>40</sup>K varied from 13.19 ± 1.18 to 136.78 ± 13.81 Bq kg<sup>-1</sup> with average of 64.25 ± 7.25 Bq kg<sup>-1</sup>. Activity concentration of <sup>238</sup>U varied from 0.58 ± 0.11 to 5.23 ± 1.90 Bq kg<sup>-1</sup>, with mean value of 2.39 ± 0.60 Bq kg<sup>-1</sup>, while the activity of <sup>232</sup>Th varied from 0.40 ± 0.05 to 3.61 ± 0.47 Bq kg<sup>-1</sup> with average value of 1.93 ± 0.28 Bq kg<sup>-1</sup>. Computed mean Ra<sub>eq</sub> was found to be 10.10 ± 1.57 Bq kg<sup>-1</sup>. The Ra<sub>eq</sub> evaluated was below the UNSCEAR limit of 370 Bq kg<sup>-1</sup>. Total mean annual effective dose due to intake of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K from ingestion of powdered milk by the infants (ages 0 to 2 years), children (ages 2 to 17 years) and adults (>17 years) are lower than the safety limit of 1.0 mSv<sup>-1</sup> for all ages recommended by the International Commission on Radiological Protection. Hence the consumption of milk products does not pose any significant health challenge to the population.*

**Keywords:** *Effective dose, Ingestion, Natural radioactivity, Powdered milk.*

## 1.0 Introduction

Radionuclides are present in the environment due to various sources such as natural background radiation, nuclear accidents or nuclear weapons testing (UNSCEAR, 2008). Primarily; the main sources of radiation exposure to humans through food ingestion are naturally occurring radionuclides, particularly <sup>40</sup>K and the radionuclides from <sup>238</sup>U and <sup>232</sup>Th or their progenies (Duong *et al.*, 2021). The radionuclides present in the environment can contaminate plants, soil, water and food. Radionuclides both of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K may enter the human body mainly through inhaling contaminated air and ingestion of contaminated food or water (Obaid 2018).

Milk is an important source of nutrients in infants, children and adults' diets. Milk contains fats, proteins, carbohydrates and vitamins required by human body for healthy growth and development. Milk is an important basic material for making yoghurt, cheese, chocolate and other cookies (El Mestikou *et al.*, 2018). Humans have domesticated animals such as cows and sheep to get milk. Cow's milk account for the bulk source of milk globally; goat, sheep, camel and buffalo account for the other source of milk (Nandan and Kumar, 2022). Due to liquid's milk short span, it is processed into powdered milk. Powdered milk can be stored for a longer period and is easily transported as a result of weight and volume reduction during conversion from liquid milk. Despite milk been an important food enriched with proteins, carbohydrates, vitamins and calcium (Alamoudi, 2013). However, it is important for milk consumers and producers to note that milk quality can be compromised by contamination from naturally occurring radionuclides (Al-Sharabi, 2020). Powdered milk can be contaminated by radionuclides through soil – grass – cow pathway. The presence of radionuclides in powdered milk can pose a potential health risk to consumers, especially infants and children, who are more sensitive to radiation exposure (Jemii and Alharbi, 2018). Estimation of activity concentration and dose assessment of radionuclides in powdered milk consumed will

significantly contribute to radiation protection. Some brands of powdered milk consumed in Nigeria are imported from different countries. Therefore, it is important to monitor and assess the levels of radionuclides activity in powdered milk and estimate the corresponding radiation effective doses.

The radionuclides activity in powdered milk has received attention from researchers across many parts of the world. In Mali, a report showed the activities of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  as  $32.8\pm 2.3$ ,  $12.3\pm 1$ ,  $242.8\pm 6.8$  and  $0.0026\pm 0.0004$   $\text{Bqkg}^{-1}$  respectively with annual effective dose of  $< 1$   $\text{mSvy}^{-1}$  (Coulibaly *et al.*, 2023). Activities of  $114\pm 1.2$   $\text{Bqkg}^{-1}$ ,  $140\pm 1.2$   $\text{Bqkg}^{-1}$ , and  $1826\pm 3.1$   $\text{Bqkg}^{-1}$  for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively were reported in Egypt (Hamid *et al.*, 2017). In Bangladesh, annual effective dose of  $1.30$   $\text{mSvy}^{-1}$  for infants and  $0.5$   $\text{mSvy}^{-1}$  for children were reported (Begam *et al.*, 2020). Similar radionuclides activities and annual effective doses have been reported by Rejah (2017), Al-jnabi (2017), Yii (2019), Silva *et al.* (2019), Duong *et al.* (2021), Uwatse *et al.* (2015) and Sarker *et al.* (2021). The activity concentration assessment of radionuclides in powdered milk consumed is vital in radiation protection, control and monitoring in foods. Data from this study would be added to existing available data in Nigeria which may assist national health policy makers, health agencies and research community in highlighting the radiological safety of powdered milk and other foods in general. Therefore, this research is aimed at determining natural radioactivity in some brands of powdered milk consumed in Nigeria and effective dose estimation.

## 2. Material and Methods

### 2.1 Sample Collection and Preparation

Five (5) commonly consumed brands of powdered milk across Nigeria were obtained from major suppliers for analysis. The collected brands are presented in Table 1.

**Table 1:** Powdered milk brands analysed and its characteristics.

Brand Name	Sample Code	Country of origin	Source	Age group	Manufactured date (dd/mm/yy)
Nunu	A1	Netherlands	Cow milk	All Ages	21-12-22
Cowbell	A2	South Africa	Cow milk (milk + vegetable oil)	All Ages	10-02-23
Peak milk	A3	Netherlands	Cow milk	All Ages	01-02-23
Milky	A4	United Arab Emirate	Cow or Sheep or Camel milk	All Ages	24-06-22
Three Crowns	A5	Netherlands	Cow milk	Children and Adults	23-04-23

The brands of powdered milk were correctly labelled as shown in table 1, neatly packed and transported to the laboratory for radiometric analysis. In the laboratory,  $200\pm 0.2$  g of each sample was accurately weighed into neatly washed cylindrical plastic containers. The containers (with the samples) were then sealed tightly with top cap and doubled sealed by wrapping adhesive tape around their necks. Each brand of powdered milk was prepared in triplicate for analysis. The sealed samples were then stored for about 28 days to allow the parent radionuclides attain secular equilibrium with their respective daughters.

### 2.2 Sample Analysis

The radioactivity concentration levels of the samples were analysed using a  $3 \times 3$  inch NaI(Tl) gamma ray detector at Ladoke Akintola University of Technology (LAUTECH) Ogbomosho, Nigeria. The detector which was housed in cylindrical lead to attenuate the influx of radiation, was coupled to a Gamma Spectacular (model GS-2000 Pro) multichannel analyser and further linked to a computer for display. Data acquisition and analysis of gamma-ray spectra were achieved using Thermo software. Prior to analysis, energy calibration of the detector was carried out

using the RSS8 gamma source (Spectrum Techniques LLC, USA), while the detection efficiency calibration of the detector was carried out using a standard source ( $^{137}\text{Cs}$  and  $^{60}\text{Co}$ ). An empty container was counted for 36000 seconds so as to determine the background gamma-ray distribution count. The sealed samples after attaining a state of secular equilibrium, were each placed on the detector and counted for the same period of time as that of the empty container. Activity concentration of  $^{40}\text{K}$  in the samples was evaluated based on its characteristic  $\gamma$ -line of 1460.0 keV. The gamma ray pick of  $^{214}\text{Bi}$  (1764.5 keV) and that of  $^{208}\text{Tl}$  (2614.7 keV) were used to estimate the activity concentrations of  $^{238}\text{U}$  and  $^{232}\text{Th}$  respectively. The relative activity concentration  $A$  ( $\text{Bq kg}^{-1}$ ) of each identified  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the samples were computed using the equation:

$$A = \frac{C_{net}}{P_{\gamma} \times \varepsilon \times m \times t} \tag{1}$$

where  $C_{net}$  is the net peak count for each radionuclide present in the sample after subtracting the background count from the gross count,  $P_{\gamma}$  is the absolute gamma-ray emission probability of the identified radionuclide,  $\varepsilon$  is the obtained full energy peak efficiency for each identified radionuclide,  $m$  is sample mass and  $t$  is the counting time.

Due to a non-uniform distribution of naturally occurring radionuclides in samples; the activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are represented by a weighted factor component called radium equivalent ( $Ra_{eq}$ ).  $Ra_{eq}$  was computed based on the estimation that  $1.0 \text{ Bqkg}^{-1}$  of  $^{238}\text{U}$ ,  $0.7 \text{ Bqkg}^{-1}$  of  $^{232}\text{Th}$  and  $13. \text{ Bqkg}^{-1}$  of  $^{40}\text{K}$  produces equal dose rates (Al-jnabi, 2017).  $Ra_{eq}$  was calculated using the equation (Obaid, 2018):

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.0770A_K \tag{2}$$

where  $A_U$ ,  $A_{Th}$  and  $A_K$  are activities of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively in  $\text{Bqkg}^{-1}$ .

### 2.3 Estimation of Annual Effective Dose

The annual effective dose  $D$  due to ingestion of radionuclides was estimated using the following equation (El Mestikou *et al.*, 2018; UNSCEAR, 2000):

$$D = A \times I \times E \tag{2}$$

where  $D$  is the effective dose ( $\mu\text{Sv y}^{-1}$ ),  $A$  is the activity concentration of the radionuclides ( $\text{Bq kg}^{-1}$ ),  $I$  is the annual milk intake rate with respect to age groups and  $E$  is the dose conversion factor  $\text{Sv Bq}^{-1}$ . The annual milk intake rate and dose conversion factors are presented in Table 2.

**Table 2.** Dose conversion Factors for  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  and Ingestion Rate for Age Groups ((ICRP, 1996; ICRP, 2012; Benaissa *et al.*, 2020).

Category	Ingestion Rate ( $\text{kg y}^{-1}$ )	Dose Conversation Factor ( $\text{Sv Bq}^{-1}$ )		
		$^{40}\text{K}$	$^{238}\text{U}$	$^{232}\text{Th}$
Infants	22.4	$4.2 \times 10^{-8}$	$1.2 \times 10^{-7}$	$4.5 \times 10^{-7}$
Children	14.0	$1.3 \times 10^{-8}$	$6.8 \times 10^{-8}$	$2.9 \times 10^{-8}$
Adults	13.0	$6.2 \times 10^{-9}$	$4.5 \times 10^{-8}$	$2.3 \times 10^{-7}$

### 3. Results and Discussion

#### 3.1 Activity Concentration of Analysed Samples

Measured activity concentrations of  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  in analysed powdered milk samples and the computed  $\text{Ra}_{\text{eq}}$  are presented in Table 3. The activity of  $^{40}\text{K}$  varied from  $13.19 \pm 1.18$  to  $136.78 \pm 13.81$   $\text{Bq kg}^{-1}$  with average value of  $64.25 \pm 7.25$   $\text{Bq kg}^{-1}$ . Three Crown recorded the lowest  $^{40}\text{K}$  activity while Nunu brand has the highest. Activity concentration of  $^{238}\text{U}$  varied from  $0.58 \pm 0.11$  (Three Crown) to  $5.23 \pm 1.09$  (Nunu)  $\text{Bq kg}^{-1}$ , with mean value of  $1.93 \pm 0.28$   $\text{Bq kg}^{-1}$ , while the activity of  $^{232}\text{Th}$  varied from  $0.40 \pm 0.05$  to  $3.61 \pm 0.47$   $\text{Bq kg}^{-1}$  with mean value of  $1.25 \pm 0.19$   $\text{Bq kg}^{-1}$ . Nunu milk (Sample A1) showed the highest-level activity concentration among the sampled milk brands, which could be due to local geology of the milk farming areas and soil-to-plant radionuclide transfer rates.  $^{40}\text{K}$  activity concentrations in all the studied samples were relatively high which may be due to its high mobility, solubility and natural abundance in soils (UNSCEAR, 2000). Continuous uptake of soluble potassium from soil by plants as a result of its biological demand can also contribute to its enhanced concentration in plants which form the basic ingredients for milk production (Uwase *et al.*, 2015, Khandaker *et al.*, 2013b, Sarker *et al.*, 2021)

**Table 3.** Activity Concentration ( $\text{Bqkg}^{-1}$ ) of  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  in Powdered Milk Samples and  $\text{Ra}_{\text{eq}}$  Index ( $\text{Bqkg}^{-1}$ )

Sample Code	Brand Name	Activity Concentration ( $\text{Bqkg}^{-1}$ )			$\text{Ra}_{\text{eq}}$ ( $\text{Bqkg}^{-1}$ )
		$^{40}\text{K}$	$^{238}\text{U}$	$^{232}\text{Th}$	
A1	Nunu	$136.78 \pm 13.81$	$5.23 \pm 1.09$	$3.61 \pm 0.47$	$20.92 \pm 2.83$
A2	Cowbell	$90.32 \pm 12.52$	$3.76 \pm 1.14$	$2.88 \pm 0.52$	$14.83 \pm 2.85$
A3	Peak Milk	$42.05 \pm 7.70$	$1.31 \pm 0.59$	$1.47 \pm 0.33$	$6.65 \pm 1.65$
A4	Milky	$38.92 \pm 1.02$	$1.08 \pm 0.08$	$1.29 \pm 0.05$	$5.92 \pm 0.23$
A5	Three Crowns	$13.19 \pm 1.18$	$0.58 \pm 0.11$	$0.40 \pm 0.05$	$2.17 \pm 0.27$

Pictorial representation of activity concentrations of primordial radionuclides in all the powdered milk samples investigated is shown in Figure 1. Data obtained from this study is compared with those of similar studies around the world as expressed in table 4. Average radioactivity values obtained herewith were found to compare favourably with those from other countries round the world. The  $\text{Ra}_{\text{eq}}$  computed for all the studied powdered milk samples as presented in table 1 varied from  $2.17 \pm 0.27$  to  $20.92 \pm 2.83$   $\text{Bq kg}^{-1}$  with average of  $10.10 \pm 1.57$   $\text{Bq kg}^{-1}$ . The radium equivalent is below the permissible limit of  $370$   $\text{Bq kg}^{-1}$ . (UNSCEAR, 2000; Al-jnabi, 2017).

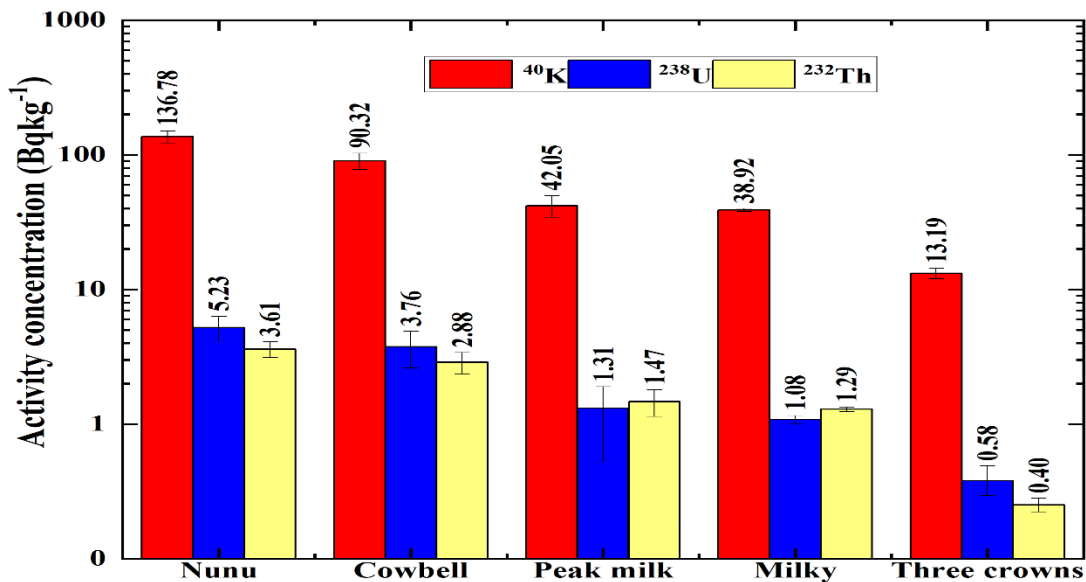


Figure 1: Activity Concentration of Radionuclides in Powdered Milk Samples

Table 4. Comparison of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K Activity Concentration with Literature Values

Region	<sup>226</sup> Ra	<sup>238</sup> U	<sup>232</sup> Th	<sup>40</sup> K	Reference
Mali	—	32.8±2.3	12.3±1.0	242.±6.8	Coulibaly <i>et al.</i> (2023)
Tunisia	BDL	—	BDL	160.83±83	El Mestikou <i>et al.</i> (2018)
Vietnam	2.45±0.24	—	1.21±0.24	387±7.0	Duong <i>et al.</i> (2021)
Egypt	114±1.2	—	140±1.2	1826±3.1	Hamid <i>et al.</i> (2017)
Baghdad	—	5.92	18.75	673.94	Rejah (2017)
Iraq	—	3.881±0.563	1.588±0.396	387.187±6.299	Al-jnabi (2017)
Jordan	—	1.01±0.3	0.42±0.13	346±20	Tawalbeh <i>et al.</i> (2020)
Saudi Arabia	9.64	—	6.77	74.51	Alamoudi (2013)
Bangladesh	—	2.98±0.38	3.94±0.29	155.7±7.5	Begam <i>et al.</i> (2020)
Malaysia	1.65±0.70	0.95±0.86	1.02±0.52	294±73	Yii (2019)
	3.05±1.84	—	2.55±2.48	99.1±69.5	Uwatse <i>et al.</i> (2015)
Thailand	3.20±0.51	—	1.24±0.10	231.20±25.50	Poltabtim and Saenboonruang (2019)
Brazil	5.49±1.08	—	1.73±0.63	242.51±9.47	Silva <i>et al.</i> (2019)
Turkey	7.3±0.7	—	4.9±0.4	482.1±48.1	Ağuş (2017)
Yemen	31.29	—	5.37	96.84	Al-Sharabi (2020)
Nigeria	—	2.93±0.60	1.93±0.28	64±7.25	Current Study

Note: BDL- below detection limit (not investigated)

### 3.2 Annual Effective Dose (AED) due to Ingestion

The estimated annual effective dose computed for all the ICRP age groups is presented in Table 5. The ingestion dose due to the intake of  $^{40}\text{K}$  by infants, children and adults respectively ranged between 12.41 and 127.94  $\mu\text{Sv y}^{-1}$  with average value of 60.30  $\mu\text{Sv y}^{-1}$ ; 2.41 and 24.75  $\mu\text{Sv y}^{-1}$  with average value of 11.66  $\mu\text{Sv y}^{-1}$  and 1.06 to 10.96  $\mu\text{Sv y}^{-1}$  with average value of 5.16  $\mu\text{Sv y}^{-1}$ . Range of annual effective dose due to intake of  $^{238}\text{U}$  by infants, children and adults is 1.56 to 14.05, 0.55 to 4.97 and 0.34 to 3.05  $\mu\text{Sv y}^{-1}$  respectively, while the mean annual effective dose due to intake of  $^{232}\text{Th}$  is respectively 19.45, 0.78 and 5.77  $\mu\text{Sv y}^{-1}$  for infants, children and adults.

**Table 5.** Annual Effective Dose due to Specific Activity of  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  for Age Groups

Brand Name	$^{40}\text{K}$ ( $\mu\text{Svy}^{-1}$ )			$^{238}\text{U}$ ( $\mu\text{Svy}^{-1}$ )			$^{232}\text{Th}$ ( $\mu\text{Svy}^{-1}$ )		
	Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults
Nunu	127.94	24.75	10.96	14.05	4.97	3.05	36.38	1.46	10.79
Cowbell	84.97	16.43	7.27	10.11	3.58	2.19	29.03	1.17	8.61
Peak Milk	39.56	7.65	3.39	3.52	1.24	0.77	14.82	0.59	4.39
Milky	36.62	7.08	3.13	2.91	1.03	0.63	13.01	0.52	3.86
Three Crowns	12.41	2.41	1.06	1.56	0.55	0.34	4.03	0.16	1.19

Table 6 give a cumulative summary of age-dependent annual effective dose to the ICRP groups due to radionuclide ingestion from the studied powdered milk samples.

**Table 6.** Age-dependent Annual Effective Dose due to the Intake of  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  from Powdered Milk.

Radionuclides	Annual effective dose ( $\mu\text{Svy}^{-1}$ )			
	Infants	Children	Adults	
$^{40}\text{K}$	Minimum	12.41	2.40	1.06
	Maximum	128.68	24.89	11.02
	Mean	60.45	11.69	5.18
	Standard deviation	6.82	1.32	0.58
$^{238}\text{U}$	Minimum	1.56	0.55	0.34
	Maximum	14.06	4.98	3.06
	Mean	6.43	2.28	1.40
	Standard deviation	1.62	0.57	0.35
$^{232}\text{Th}$	Minimum	4.03	0.16	1.20
	Maximum	36.39	1.47	10.79
	Mean	19.45	0.78	5.77
	Standard deviation	2.86	0.12	0.85
	Total mean	86.33	14.75	12.35

Mean effective dose due to intake of  $^{40}\text{K}$  by infants is 60.45  $\mu\text{Sv y}^{-1}$ , while that by the children is 11.69  $\mu\text{Sv y}^{-1}$ . The adults recorded the least mean annual effective dose of 5.18  $\mu\text{Sv y}^{-1}$  due to intake of  $^{40}\text{K}$ . Similarly, average contribution of  $^{238}\text{U}$  to the total effective dose is 7.46%, 15.43% and 11.35% respectively for infants, children and adults. Furthermore, the contribution of  $^{232}\text{Th}$  radionuclide due to milk ingestion is 22.51%, 5.30% and 46.80% for

infants, children and adults respectively. Cumulatively, annual effective dose received by infants is approximately 5.9 and 7 times higher than that received by children and adults respectively. Comparatively, results of annual effective dose for all age groups as seen in table 6 are below the recommended ICRP permissible limit of  $1 \text{ mSv}^{-1}$  due to ingestion of natural radiation sources (ICRP, 2007). Infants and children are however, susceptible to radiation exposure because of their greater intestinal absorption than adults and a lower threshold for adverse effects (Uwate *et al.*, 2015).

#### 4. Conclusion

Activity concentration of naturally occurring radionuclides,  $^{40}\text{K}$ ,  $^{238}\text{U}$ , and  $^{232}\text{Th}$  in commonly consumed brands of powdered milk in Nigeria has been determined using a  $3 \times 3$  inch NaI(Tl) gamma ray detector. Mean activity concentration of  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  in the studied samples are  $64.25 \pm 7.25$ ,  $1.93 \pm 0.28$  and  $1.25 \pm 0.19 \text{ Bq kg}^{-1}$  respectively, with corresponding average  $R_{\text{eq}}$  value of  $10.10 \pm 1.57 \text{ Bq kg}^{-1}$ . The annual effective dose for all age groups were below the permissible limit of  $1 \text{ mSv}^{-1}$ . Therefore, the analysed brands of powdered milk are radiologically safe and may not cause immediate radiological health hazards to the consumers. Although the results of this study did not suggest any immediate or urgent intervention, data generated will provide baseline radiometric values for natural radioactivity in powdered milk available for consumption in Nigerian markets. It will also assist regulatory agencies in the setting up of guidelines and regulations for radiological protection Nigeria population. Furthermore, continuous monitoring of the radiological safety of other milk brands consumed in Nigeria is advised to prevent any substantial radiologically induced health risks to milk consumers.

#### Acknowledgement

The authors are highly indebted to the staff and management of radiological measurement unit of Ladoke Akintola University of Technology (LAUTECH) Ogbomosho, Nigeria.

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