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Assessment of Radioactivity Levels in Bottled Drinking Water Consumed by Different Age Groups of People in Kampala City, Uganda

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ABSTRACT

The activity concentrations of natural radionuclides in drinking water may be enhanced depending on the geological strata from which it is extracted. The demand and consumption of bottled drinking water is increasing every year in Uganda mainly in Kampala city. Therefore, the study was purposed to assess the levels of natural radioactivity using a gamma spectrometer in commonly bottled drinking water in Kampala City. The mean specific activities for Ra-226, Th-232, and K-40 in bottled drinking water were 4.45 ± 0.20 , 1.32 ± 0.27 , and 46.33 ± 0.24 Bq/l respectively which exceeded the specific World Health Organization recommended guidance level of 1.0 Bq/l except for K-40. The mean activity concentrations were however comparable to the mean concentration of tap water used in Kampala City, that is 4.00 ± 0.16 , 1.4 ± 0.19 , and 47.05 ± 0.21 Bq/l for Ra-226, Th-232, and K-40 respectively. The annual effective dose contributed by two radionuclides of Ra-226 and Th-232 in the bottled water exceeded the maximum dose limit of 0.1 mSv/y recommended by the World Health Organization (WHO). The mean estimated excess lifetime cancer risk due to Ra-226 (0.00177) exceeded the recommended limit of 0.001 while that due to Th-232 (0.0001) was below the recommended limit. Therefore, the main contributor to the total annual ingestion dose was Ra-226. Due to the high solubility and toxicity of Ra-226, there is a significant concern warranting further investigation. Finally, the results from the study revealed that there was not any trace of contamination from artificial radionuclides in the bottled drinking water manufactured and consumed within Kampala City. With the growing populational demand for bottled drinking water, it's recommended that radioactivity levels comply with the WHO recommended radioactivity levels. Therefore, this study contributes to the revision of standards and development of

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regulations for radiological protection of the public and the environment from risks of radiation exposure associated with bottled drinking water.

Keywords: Radionuclide; Activity concentration; Gamma spectrometry; Annual effective dose; Excess lifetime cancer risk; Natural radioactivity

1. Introduction

The World Health Organization (WHO) recommends a daily intake of at least 1–2 liters (l) of water for adult human beings, which makes up about 60% of our human bodies^[1]. Therefore, this requires the supply of clean, safe, and quality drinking water. In the past, people used to drink water directly extracted from underground geological water sources like boreholes, springs, wells, dams, rocks, and others. However, this has changed over recent years with the preferential acceptance of bottled drinking water amidst the growing society's demands. The consumption of bottled drinking water in Uganda has continually increased since the 2000s, which corresponds to the growing bottled water market projected at a compound annual growth rate of 9.2% over the analysis period of 2021–2027^[2]. Amidst the growth of the bottled drinking water industries, the use of tap water supplied through piped water systems by the National Water and Sewerage Cooperation (NWSC) still prevails. However, there are several concerns about the safety and cleanliness of tap water as highlighted by^[3]. In this regard, most Ugandans opt to boil the tap water before drinking, which is inconvenient, time-consuming, and very expensive for most urban dwellers. Therefore, the best alternative is to go for bottled drinking water; that has raised the projected average volume per person in the Bottled Water market to 12.89 l in 2024^[2].

According to the East African Standard on potable water specifications for Uganda^[4], packaged drinking water refers to suitably treated water fit for human consumption, sealed in containers. The packaged water may contain natural radionuclides or intentionally added natural minerals and carbon dioxide but with no sugars, sweeteners, or any other foodstuffs added. Packaged drinking water is obtained from several sources, for example, springs, boreholes, underground rocks, sand, and wells. It is then purified by several processes including distillation, deionization, reverse osmosis, or other suitable processes before being packaged. There are variable amounts of radionuclides of natural origin present in the packaged water depending on the mineral

content and the geological subsurface from which the water is extracted^[5–8]. Through processes of erosion and dissolution, the radionuclides are transferred from rocks to the water as discussed by^[7,9]. The radionuclide concentration depends on the infiltration time, the geochemical and mineralogical composition of the rocks, seasonal precipitation, soil conditions through which the water flows, weathering, and exhalation. Such radionuclides include Potassium-40 (K-40), the decay products in the series of Uranium-238 (U-238) and Thorium-232 (Th-232), and their radioactive daughter elements like Radium-226 (Ra-226), Radon-222 (Rn-222), Tritium (H-3) (cosmogenic), and Carbon-14 (C-14). These are further classified based on the radiation-emitting energy transfer, for example, K-40, H-3, and C-14 emit radiation of low energy transfer (LET) while U-238 and Th-232 and their radioactive daughter elements generally emit radiation of high energy transfer (HET). However, some studies have found artificial radionuclides for example H-3, C-14 (also can be naturally occurring), and Cs-137 in drinking water samples^[10]. This maybe a result of contamination along the process chain from manufacturing to distribution.

The radioactivity levels of the above natural radionuclides in most cases are relatively small. However, in some cases, the radioactivity levels may exceed the guidance limits provided by the^[11] and may increase the risk of radiation exposure to the members of the public who consume such packaged water. However, as discussed by^[11,12], the guidance limits should be interpreted to trigger a further investigation but not taken as mandatory limits for unsafe drinking water. Noting that, there are several radionuclides in drinking water, the total individual dose is obtained by adding the contribution from each radionuclide and compared to the recommended Individual Dose Criterion (IDC) of 0.1 mSv/year^[12]. The assumption is that any level of radiation exposure involves some risk, therefore ingestion of drinking water with radionuclides even of low radioactive levels over longer periods may increase the cancer risk in humans^[12–14]. This is because the linear relationship that exists between

exposure and risk has no threshold below which there is no risk. In this case, the World Health Organization (WHO) opts for a strategy based on the Individual Dose Criterion (IDC) of an annual effective dose of 0.1 mSv. This involves taking several assumptions and conditions that associate the final result with bias associated with a small sample size corrected from a small geographical area as discussed in the methodology and the results sections.

Therefore, the purpose of this study is to determine the radionuclides and their activity concentrations in the common brands of bottled drinking water commercially available on the market within Kampala and the central region and assess their possible radiological risks to the consumers. This research is significant for informing the regulatory processes to establish standards of radioactivity in bottled drinking water and also addresses any potential health risks to public radiation exposure. Hence, this contributes to the establishment of appropriate and effective regulatory practices for protecting the public from radiation exposure and also facilitating smooth cross-border trade relations.

2. Methodology

2.1 Sample collection and preparation

A total of 21 brands (2 litres each brand) representing about 70% of several brands of water on the market approved by the Uganda National Bureau of Standards (UNBS) from all geographical regions of Uganda. The samples were taken to the laboratory and 1 liter of each water brand was transferred to the Marinelli beaker and acidified with four drops of 0.1M HNO₃ to prevent aggregation of radionuclides within the beaker walls. The Marinelli beakers were hermetically sealed and labeled with unique identification numbers from SAP-062 to SAP-082. The prepared samples were stored in a cool dry place for a minimum period of 21 days, to attain a secular equilibrium between the Uranium and thorium decay daughter elements^[15].

2.2 Measurement of radioactivity concentration

The activity concentration for each sample was measured using a P-type HPGe coaxial detector (Model: GC3018 and Serial no.: b 20041) with an energy resolution of 1.9 keV at a 1.33 MeV γ -ray line of Co-60 and a relative efficiency

of 50%.

The detector was connected to the interface module and an MCA for advanced digital signal processing configured by Genie 2000 software for acquiring and analyzing spectra from MCA, and reporting^[16]. The Marinelli beaker had the same geometry used during efficiency and energy calibration of the detector with a standard source, type: MBSS2, certificate no.: 1035-SE-40840-22 and serial no.: 241022-1871039. Each sample was placed on the detector, read for 12 hours, and its spectrum was acquired and saved. The activity (A) in Bq/l and the activity uncertainty of the radionuclides in each sample were then obtained using the Genie 2000 software. The activity and its uncertainty are the weighted means for several activities of different peaks selected based on significant yield percentages representing daughter elements for each radionuclide.

2.3 Radiological risk assessment

The radiological risks were assessed by estimating the annual effective dose using Equation (1) and excess lifetime cancer risk (*Lcr*) using Equation (2) for different age groups.

$$A = Ac * EDF * Q \quad (1)$$

Where *A* is an annual effective dose, *Ac* is the activity concentration for each radionuclide, *EDF* is the committed effective dose in General Safety Requirements Part 3^[17] and *Q* is the consumption rate.

$$Lcr = Radionuclide\ intake\ (Bq) * mortality\ riskcoefficient(Bq^{-1}) \quad (2)$$

The mortality risk coefficients for ingestion of food and tap water are published in^[18].

3. Results and discussion

3.1 Radioactivity analysis in bottled drinking water

The activity concentrations for each radionuclide are shown in **Figure 1**. The activity concentrations for K-40 and Th-232 show generally a uniform distribution except for the case of the activity concentration of Th-232 in SAP-081. The activity concentration of Ra-226, however, shows a normal distribution skewed to the left. In comparison, there

is a minimal variation between the concentrations of each radionuclide between tap water and bottled drinking water samples as shown in **Figure 1**. This variation might be due to the different water sources of origin or extraction for example most of the bottled drinking mineral water is extracted from underground geological structures while most tap water used in Kampala is treated water extracted from Lake Victoria. The statistical description of the activity concentration for each radionuclide is shown in **Table 1**. The standard deviation values were generally <2 . This implies a minimal variability from the mean in the activity concentration of the radionuclides in the different drinking water samples^[19]. This could be attributed to the different geological origins of groundwater, for example boreholes, rocks, underground water beds, wells and lakes from which the bottled drinking water is extracted and processed. This is based on the natural distribution and occurrence patterns of minerals from sediment particle surfaces and rocks that have dissolved into water over a longer lifetime. The standard error values are very small tending to zero which further indicates a minimal discrepancy between the measured radioactivity and the mean values as shown in **Table 1**.

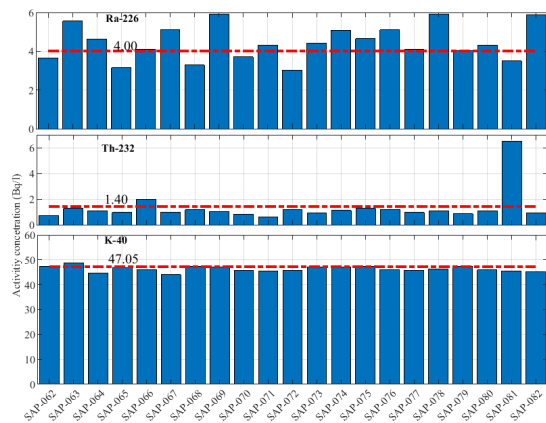


Figure 1. A bar graph showing the variation of activity concentration of the radionuclides in the bottled drinking water samples in comparison with tap water is shown by a dotted line.

The activity concentration of Ra-226 was within a range of 3.0 to 6.0 Bq/l with a mean of 4.45 ± 0.20 Bq/l which was higher than the mean radioactivity level of Ra-226 in tap water and above the WHO guidance level of 1.0 Bq/l^[11]. However, the results are comparable to Ra-226 levels from previous studies in other countries like Malaysia, Spain, Turkey, and Finland as shown in **Table 2**.

The concentration of Th-232 was within the range of

0.63 to 2.00 Bq/l except SAP-081 whose activity concentration is about 6 times greater than the WHO guidance level of 1.0 Bq/l. The mean activity concentration of Th-232 was 1.32 ± 0.27 which is slightly above the WHO guidance level of 1.0 Bq/l^[11]. Compared with tap water, the mean activity concentration in bottled drinking water was higher than that of tap water and higher than the previously reported Th-232 concentration levels of 0.005 Bq/l in Pakistan^[23], 0.19 Bq/l in Bangladesh^[24], and 0.007 Bq/l in Hong Kong^[25].

The mean concentration level of K-40 was 46.0 ± 0.24 Bq/l which was above the mean concentration of tap water. In comparison with the previously reported concentration levels in other countries, the mean concentration of K-40 is 0.14 Bq/l in Pakistan^[23], 4.16 Bq/l in Bangladesh^[24], 0.11 Bq/l in Hong Kong^[25], and 3.8–14.6 Bq/l^[7]. Note that, there is no established international guideline for K-40 concentration levels in drinking water since human metabolic processes control its concentration^[11, 12]. In this regard, the next analysis is silent on the contribution of Potassium to annual effective dose and excess lifetime cancer risk.

Our study established that no artificial radionuclides were detected in the water samples. The mean activity concentrations for each natural radionuclide in the bottled drinking water samples generally exceeded the guidance level recommended by WHO and IAEA^[11]. However, this may not imply unsafe bottled drinking water according to IAEA^[11].

3.2 Estimation of annual effective doses

The study considered different age groups representing the whole population of Kampala City to estimate the annual effective dose resulting from the uptake of each radionuclide in drinking water to an individual using Equation (1). The different age groups considered were 1–5 years, 5–10 years, 10–18 years, and over 18 years. The consumption rate for each age group is 328, 440, 512, and 730 liters of drinking water per year^[11, 23]. The effective dose coefficients used in the study were committed effective doses provided in the General Safety Requirements Part 3^[17]. The calculated age-dependent annual effective dose from each radionuclide for each age group is shown in **Table 3**. Generally, the dose contributed by Ra-226 only already exceeded the WHO recommended limits of the annual effective dose of 0.1 mSv/y while the dose contributed by Th-232 was less than the WHO recommended limits of the annual effective dose

Table 1. A table showing the statistical description of the activity concentration of radionuclides in the analyzed bottled drinking water samples.

Statistic description	Activity of radionuclides (Bq/l)		
	Ra-226	Th-232	K-40
Average	4.45	1.33	46.33
Median	4.32	1.08	46.00
Standard error	0.20	0.27	0.24
Standard deviation	0.91	1.22	1.11
Skewness	0.19	3.93	-0.04
Kurtosis	2.00	17.25	2.78
Minimum	3.01	0.63	44.00
Maximum	5.93	6.52	48.70
Mean radioactivity levels of tap water	4.00	1.40	47.05

Table 2. A table showing the activity concentration of Ra-226 in drinking water done in previous studies in other Countries.

Country	Concentration of Ra-226 (Bq/l)	References
Finland	0.01–49	[20]
Germany	0.001–1.8	[21]
Spain	0.02–4.0	[6]
Malaysia	1.46–3.30	[22]
Turkey	3.8–14.6	[5]

of 0.1 mSv/y from the results in **Table 3**. Taking the initial assumption of neglecting the K-40 contribution, the summation of dose contributed by Th-232 and Ra-226 exceeded the WHO guidance limit of the annual effective dose of 0.1 mSv/y for all age groups. The radionuclides identified in **Table 3** once ingested, are transported along the blood streams and accumulate in other tissues which maybe damaged^[26]. A few epidemiological studies detail the evidence of possible radiation effects from radionuclides in drinking water for example cancer incidence^[27], Leukemia incidence and mortality^[28], cases of tuber/renal damage^[29,30], and other case studies detailed by^[31]. Therefore, the results exceeding the WHO guidance limit of Annual Effective Dose may pose an increased risk of possible radiological exposure to the public though does not imply unsafe drinking water^[11].

3.3 Radiological risk assessment from bottled water intake

For risk assessment purposes, an average city dweller in Kampala uses at least 2 to 3 brands of water in a year in

addition to tap water which maybe boiled or non-boiled. All these water brands contribute to the assumed consumption rate used to calculate annual effective doses given in **Table 3**. The preference and the rate of consumption of water depend mainly on four factors.

- i. The perception of the quality of the water brand
- ii. The availability of the water brand
- iii. The price of the water and

Based on the findings of^[32,33], about 20% of people in Kampala drink non-branded water sources like boiled and non-boiled tap water, borehole water sources, and others. Based on the above assumptions, three water brands were considered for the calculation of the total annual ingestion dose based on the above factors. SAP-063 is preferred by the high-quality end people in offices, residences, and any official gatherings and functions due to its assumed quality and availability, and sustainable market demands for over 20 years in Uganda branding it as the best quality water in Uganda^[34]. Hence this contributes about 45% to the consumption rate. SAP-071 is preferred by the low-quality end

Table 3. A table showing the activity concentration of each radionuclide and their contribution to the annual effective dose in the sampled bottled water.

Sample	Activity of radionuclides (Bq/l)			Annual effective dose due to Ra-226 (mSv/y)				Annual effective dose due to Th-232 (mSv/y)			
	Ra-226	Th-232	K-40	1-5 y	5-10 y	10-18 y	≥18y	1-5 y	5-10 y	10-18 y	≥18y
SAP-062	3.64	0.71	47.40	0.334	0.448	0.522	0.744	0.051	0.069	0.080	0.114
SAP-063	5.55	1.27	48.70	0.510	0.684	0.796	1.134	0.092	0.123	0.143	0.204
SAP-064	4.63	1.08	44.60	0.425	0.570	0.664	0.946	0.078	0.105	0.122	0.173
SAP-065	3.14	0.96	46.70	0.288	0.387	0.450	0.642	0.069	0.093	0.108	0.154
SAP-066	4.11	1.99	46.00	0.377	0.506	0.589	0.840	0.144	0.193	0.224	0.320
SAP-067	5.10	0.99	44.00	0.468	0.628	0.731	1.042	0.071	0.096	0.112	0.159
SAP-068	3.29	1.17	47.30	0.302	0.405	0.472	0.672	0.084	0.113	0.132	0.188
SAP-069	5.93	1.05	47.10	0.545	0.731	0.850	1.212	0.076	0.102	0.118	0.169
SAP-070	3.70	0.84	45.80	0.340	0.456	0.530	0.756	0.061	0.081	0.095	0.135
SAP-071	4.30	0.63	45.50	0.395	0.530	0.616	0.879	0.045	0.061	0.071	0.101
SAP-072	3.01	1.18	45.80	0.276	0.371	0.432	0.615	0.085	0.114	0.133	0.190
SAP-073	4.41	0.94	47.20	0.405	0.543	0.632	0.901	0.068	0.091	0.106	0.151
SAP-074	5.09	1.12	47.10	0.467	0.627	0.730	1.040	0.081	0.108	0.126	0.180
SAP-075	4.65	1.27	47.50	0.427	0.573	0.667	0.950	0.092	0.123	0.143	0.204
SAP-076	5.10	1.19	46.00	0.468	0.628	0.731	1.042	0.086	0.115	0.134	0.191
SAP-077	4.10	0.96	45.80	0.377	0.505	0.588	0.838	0.069	0.093	0.108	0.154
SAP-078	5.92	1.09	46.40	0.544	0.729	0.849	1.210	0.079	0.106	0.123	0.175
SAP-079	4.03	0.85	47.40	0.370	0.496	0.578	0.824	0.061	0.082	0.096	0.137
SAP-080	4.32	1.08	45.90	0.397	0.532	0.619	0.883	0.078	0.105	0.122	0.173
SAP-081	3.49	6.52	45.60	0.321	0.430	0.500	0.713	0.470	0.631	0.734	1.047
SAP-082	5.88	0.94	45.20	0.540	0.724	0.843	1.202	0.068	0.091	0.106	0.151
Mean	4.45	1.33	46.33	0.408	0.548	0.638	0.909	0.096	0.128	0.149	0.213

people due to its lowest price and is generally available in every shop outlet within the city, and hence contributes about 25% to the consumption rate. SAP-076 has high market demand for being used to refill water dispensers within Kampala city and its suburbs due to the assumed quality, price, and availability, and hence contributes 10% to the yearly consumption rate. Therefore, taking the above assumptions, a modification to Equation (1) to Equation (3) for the calculation of the total ingestion dose (mSv/y), where the subscript *i* represents the different radionuclides in each water sample numbering from 1 to *n* value that contributes to the consumption rate and *Q* represents the consumption rate contributed by each water brand.

$$Total\ ingestion\ dose\ (mSv/y) = \sum_{i=1}^n Ac_i * ADF_i * Q \tag{3}$$

Therefore, the total ingestion dose was calculated as the summation of the annual effective doses of at least 3 brands of water and the results are shown in **Table 4**.

The total ingestion dose for all age groups shown in **Table 4** exceeds the Individual Dose Criterion (IDC) of 0.1 mSv/y recommended by WHO and IAEA^[11]. This highlights a particular concern for the regulator and the public since it shows a possibility of radiation exposure due to elevated levels of natural radiation from the ingestion of bottled drinking water under the considered assumptions. Based on

these results, the national authorities may decide to implement some measures to reduce the radionuclide levels and or impose restrictions to limit the consumption of drinking water. However, for all cases of age groups in **Table 4**, the total ingestion dose does not exceed the dose criterion of 1 mSv/y recommended by GSR Part 3^[17] as a guidance level for initiating remedial measures to limit radioactivity.

From the above results, a case of adults (≥18 years) who had the maximum annual effective dose of 0.970 mSv/y was considered to calculate the excess lifetime cancer risk, utilizing the mortality risk coefficients for radionuclide uptake in water published by^[18]. Therefore, the excess lifetime cancer risk (*Lcr*) is calculated using the United States Environmental Protection Agency approximation given in Equation (2), Section 2.3. Radionuclide intake is the product of radionuclide concentration, ingestion rate, ingested fraction from the contaminated source, exposure frequency, and exposure duration^[5]. The exposure duration of 68 years was assumed for a Ugandan adult based on a recent survey^[35], the ingestion rate of 2 l/day, the ingested fraction of 100% or 1, and the exposure frequency of 365 days/y were considered^[5]. Therefore, the calculated *Lcr* for each radionuclide is shown in **Table 5**.

The *Lcr* for Ra-226 ranged from 0.00167 – 0.00188 exceeding the acceptable level of 0.001 while that of Th-232 ranged from 0.00008 – 0.00011 which is below the accept-

Table 4. A table showing the total annual ingestion contributed from drinking water.

Sample name	Total ingestion dose (mSv/y)			
	1-5 y	5-10 y	10-18 y	≥18y
SAP-063	0.271	0.363	0.422	0.602
SAP-071	0.110	0.148	0.172	0.245
SAP-076	0.055	0.074	0.087	0.123
	0.436	0.585	0.681	0.970

Table 5. A table showing the Lcr for radiological risk assessment of bottled drinking water.

Radionuclide	Activity concentration (Bq/l)		Lcr
	Min	Max	
Ra-226	Min	4.682	0.00167
	Mean	4.98	0.00177
	Max	5.278	0.00188
Th-232	Min	0.866	0.00008
	Mean	1.03	0.00010
	Max	1.194	0.00011

able level of 0.001 [5, 7, 36]. This is in line with other previous studies for example, [5] calculated excess lifetime cancer risk of Ra-226, Ra-228, and Pb-210 as 8.04×10^{-5} , 1.3×10^{-3} , 1.1×10^{-2} , and 7.6×10^{-6} in Iran. Therefore, from this assessment, Ra-226 makes the most contribution to the total ingestion dose and is likely to cause increased chances of developing cancer to the public due to ingestion of bottled drinking water.

4. Conclusions

The activity concentrations of Ra-226, Th-232, and K-40 in 21 different brands of bottled drinking water consumed in Uganda were determined using gamma spectrometry. The results revealed that all the average activity concentrations for each radionuclide were generally above the recommended WHO guidance levels. The study neglected the contribution of K-40 to the dose since its concentration is controlled by human metabolic activities despite its activity concentration being greater than most of the previous studies. The activity concentration of Ra-226 contributed about 80% of the total annual effective dose resulting in excess lifetime cancer risk exceeding the recommended limit of 0.001. The annual effective dose defined as the total ingestion dose exceeded the WHO recommended IDC of 0.1 mSv/y. Given the high solubility and toxicity of Ra-226, its presence in high levels of radioactivity in drinking water is of great concern and re-

quires further investigation. The recommendation for further investigation should consider the guidelines recommended by the WHO to screen for gross alpha and beta activity concentration levels [9, 12].

Therefore, the data obtained contributes to the determination of baseline levels of natural radioactivity in bottled drinking water in Uganda. The study also provides basic information on radionuclide intake by the consumers and the concerned stakeholders for risk assessment from radioactivity in drinking water. However, throughout the study, there was a limitation of data on several aspects for example the statistics of consumption rates of the different water brands which may associate the results with some bias. However, these acted as assumptions for the study that need to be improved on in the subsequent studies. Additionally, the next studies need to investigate further the geographical origins of these water sources and also consider tap water in the calculation of the effective dose.

Author Contributions

Wellen Rukundo and Natharius Nimbashabira developed the concept, and the manuscript, and also team-led in conducting the research, Kagulire Daniel, and Dornum Katusiime and the whole team reviewed the manuscript, and Noah Deogratias Luwalira funded the research.

Conflict of interest

The authors declare that they have no competing financial and personal interests that could influence the work reported in this paper.

Data Availability Statement

The data that was used for this research is available and is part of the submission.

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