

Article

Assessment of Radioactivity Concentrations and Associated Radiological Health Risk in Natural Spring Mineral Bottled Drinking Water from South Africa

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Abstract: Naturally occurring radionuclides are ubiquitous at various levels of concentration, while exposure to ionizing radiation by humans is of global concern. Radiological health risk assessment due to the consumption of natural spring mineral water is critical for ensuring public health and safety. This study aims at investigating the radioactivity concentration levels of natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K and the associated radiological health risk in commercial natural spring bottled water in South Africa. A total of 21 of the most-consumed bottled drinking water brands from grocery stores, were analysed using the HPGe gamma detector. The results indicate that the range of radioactivity concentrations is from 1.060 ± 0.067 to $2.571 \pm 0.143 \text{ BqL}^{-1}$, with a mean of $1.766 \pm 0.399 \text{ BqL}^{-1}$ for ^{226}Ra ; 1.736 ± 0.112 to $7.807 \pm 0.099 \text{ BqL}^{-1}$, with a mean of $3.688 \pm 1.371 \text{ BqL}^{-1}$ for ^{232}Th and 149.000 ± 38.480 to $242.900 \pm 59.700 \text{ BqL}^{-1}$ with a mean of $220.229 \pm 22.297 \text{ BqL}^{-1}$ for ^{40}K . The potential radiological health risks evaluated show mean values for Ra_{eq} , D_{Ab} , AEID and AGED as $23.976 \pm 0.446 \text{ BqL}^{-1}$, $12.232 \pm 1.445 \text{ nGyh}^{-1}$, $0.060 \pm 0.007 \text{ mSvy}^{-1}$ and $0.090 \pm 0.027 \text{ mSvy}^{-1}$, respectively. The radiation dose based on age group is in the order of infants (≤ 1 year) > teenagers (12–17 years) > children (1–12 years) > adults (> 17 years). The activity concentrations of radionuclides in bottled water are ranked in the order of $^{40}\text{K} > ^{232}\text{Th} > ^{226}\text{Ra}$, with ^{232}Th contributing the highest radiation dose, consistent with findings reported in previous studies. The findings reveal that the activity concentration levels and estimated radiological health risks are within the permissible limits set by UNSCEAR guidelines. Therefore, the consumption of bottled water is radiologically safe. However, the findings also suggest that 12 out of 1000 individuals may suffer cancer fatality, while 6 out of 1 million individuals may experience hereditary effects over their lifetime from the consumption of bottled water. Regular monitoring and stringent regulatory controls are recommended to ensure the radiological safety of bottled drinking water in South Africa.

Keywords: effective dose; gamma spectrometry; natural spring mineral bottled drinking water; radioactivity; radiological health risk; South Africa



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1. Introduction

The availability and accessibility of safe drinking water is a global critical public health issue, especially in regions where natural and anthropogenic factors may contribute to the quality of water [1–3]. In the republic of South Africa, natural spring mineral bottled drinking water is a popular alternative to tap and other sources of drinking water, as

obtainable globally, and is often perceived to be safer and of higher quality [4–7]. But with abundant mineral deposits giving rise to mining, agricultural and industrial activities, groundwater contamination may result from the drainage, leaching and seepage of mine water and other chemicals [8–11]. With the rising global population, sanitation and global warming effect, the demand for and consumption of bottled water has been expected to increase in the coming years [12–14]. However, the radioactivity concentrations of naturally occurring radionuclides in commercial bottled water constitutes potential radiological health risks to consumers if not regularly checked and regulated, especially with the increased demand for drinking water [1,15,16]. This follows from the South African regulatory provision on bottled water, quoted as “No bottled water shall contain substances or emit radioactivity in quantities that may be injurious to the health of the consumer” [17].

The natural radionuclides like uranium-238 and thorium-232, with their progenies, as well as potassium-40, are ubiquitous in the earth and can be present in drinking water sources, as well [5]. These radionuclides enter water supplies through geological formations and human activities, potentially leading to long-term exposure when ingested. The continuous consumption of water with elevated levels of radionuclides may increase the risk of adverse health effects, including cancer and other radiation-induced diseases [18,19].

Studies on the radioactivity concentration levels of naturally occurring radionuclides in bottled water have been reported [16,20–22], using various analytical procedures with varied activity concentrations. Similar studies with different methodologies have also been reported by [23–25] on drinking water in the same study area. However, a careful search of the literature showed that such work has not been reported in South African bottled drinking water; hence, the aim of this baseline study is to assess the radioactivity concentration levels of the radionuclides in the natural spring mineral bottled commercial drinking water brands mostly consumed in South Africa. Utilising the High-Purity Germanium (HPGe) gamma spectrometry technique, the levels of radioactive contaminants in the sampled bottled water have been measured. By finding the levels of naturally occurring radioactive contaminants, the associated radiological health risks because of the ingestion of bottled drinking water were estimated and compared with national and international safety standards as well as other published related data. The findings from this research will provide a valuable understanding of the safety of consuming natural spring mineral bottled drinking water in South Africa and inform regulatory bodies to ensure public health protection.

The main aim and objectives of this study are to assess the activity concentrations and associated radiological health risks such as the effective dose, gonadal dose and lifetime cancer risks, among others, in the natural spring mineral bottled drinking water from South Africa. Hence, in this article, the methodology used for sampling and analysis will be discussed and the results of radioactivity concentration levels and analysis of potential health risks based on the national and international guidelines and standards will be presented. The sole objective is to contribute to the ongoing efforts in safeguarding public health through highlighting the importance of monitoring and regulating the levels of natural radionuclides, ^{226}Ra , ^{232}Th and ^{40}K , in drinking water, for sustainable growth and development, aligning with goal 6 of the Sustainable Development Goals.

2. Materials and Methods

The twenty-one different brands of water samples analysed in this study are made up of the most available and topmost consumed bottled natural spring mineral commercial drinking water, collected from grocery stores after an online mini survey of the literature, in South Africa [13]. The samples were prepared according to the specified standards which the South African Water Quality Guidelines [5] and the World Health Organization [1]

provide. The bottled water samples were prepared for analysis by pouring 0.5 L into a cylindrical polyethylene vial (made in South Africa) based on the gamma detector geometry and then sealed using black polyethylene adhesive tape, to make it airtight, and stored for a period of thirty days. The essence is for the naturally occurring radionuclides ^{238}U (^{226}Ra) and ^{232}Th , and their progenies, to reach secular equilibrium, as shown in Figure 1.



Figure 1. The experimental setup shows pictures of the collected and prepared samples as well as the HPGe gamma spectrometer for the natural spring mineral bottled drinking water analysis.

The equipment used in this study for data acquisition and spectrum analysis, as shown in Figure 1, is the Canberra well-type (HPGe) gamma detector, GCW-2021 model, made in the USA. It is connected to a multi-channel analyser (MCA) of the DSA-1000 model incorporated with software for analysis known as Genie 2000, version 3.3. The spectrometer has a 2.1 keV energy resolution with a ^{60}Co gamma energy line of 1.33 MeV FWHM, 20% relative efficiency. A multinuclide Europium-152 standard source(s), which can emit gamma rays of a wide energy range between 46.54 keV and 1836 keV, was used in the calibration of the detector, in terms of the energy (keV) and efficiency (%), with the curve shown in Figure 2. The measurement involves placing the water samples into a cylindrical enclosure made of lead with low background activity, 11.50 cm thick on the detector, and counted for 12 h. Prior to the determination of the activity, there was a background subtraction from the gamma ray spectrum. The analysis was performed at the analytical laboratory of the Centre for Applied Radiation Science and Technology (CARST), North-West University (Mahikeng Campus).

The equipment validation was achieved using the International Atomic Energy Agency (IAEA)'s standardised reference materials (manufactured by Eckert & Ziegler Isotope products, Valencia, CA 91355, USA), ^{152}Eu , ^{133}Ba and IAEA-RGU-1, to ascertain the level of accuracy of the spectrometer. This was performed by determining the activity of the sources at production, from their respective certificates and then at present after decay, with the Rad-Pro decay calculator online, and comparing with the activity measured by the detector. The results showed agreement with the calculated activity of ^{152}Eu shown to be 25.447 kBq, while the measured activity was 26.027 kBq, with a percentage error of 2.281%. The laboratory has recently participated in the IAEA interlaboratory proficiency exercise in 2024.

In this study, the various gamma lines of the radionuclides together with their daughters were used to determine the activity concentrations values by computing their weighted mean values with their respective uncertainties. The gamma line peak energy used for ^{238}U (^{226}Ra) progenies were ^{214}Pb (295.21 keV, P_γ of 19.2% and 351.95 keV, P_γ of 37.20%) and ^{214}Bi (609.31 keV, P_γ of 46.30% and 1120.29 keV, P_γ of 15.10%). For ^{232}Th , the gamma

photopeak energy lines of the daughters, ^{212}Pb (238.63 keV, P_γ of 44.60% and 727.17 keV, P_γ of 11.80%), ^{212}Bi (727.17 keV, P_γ of 11.80%) and ^{228}Ac (911.60 keV, P_γ of 27.70% and 969.11 keV, P_γ of 16.60%), were used, while for ^{40}K , the single gamma energy line at 1460.81 keV, P_γ of 10.67% was used.

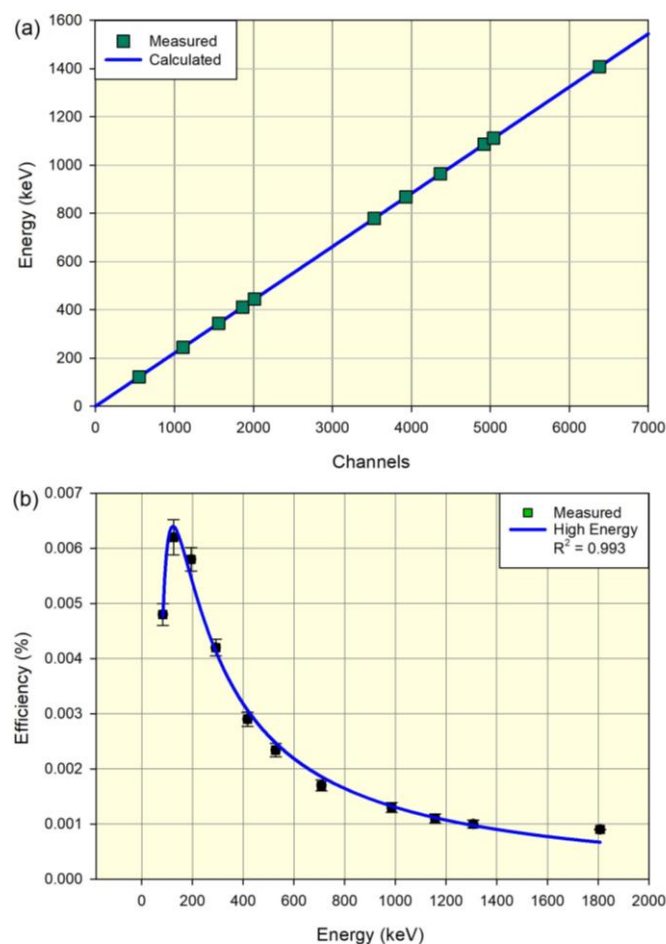


Figure 2. Calibration curves (a) Energy-Channels (b) Efficiency-Energy Curves, of the HPGe gamma detector at CARST-NWU South Africa.

The radioactivity concentration A (BqL^{-1}) was computed utilizing Equation (1), which was preceded by the subtraction of the decay corrections [26].

$$A(\text{BqL}^{-1}) = \frac{N_a}{P_\gamma \times \varepsilon_\gamma \times t \times V} \quad (1)$$

where N_a represents the net peak area corresponding to a specific energy line in the spectrum. P_γ denotes the probability of emitting gamma radiation at that particular energy photopeak, while ε_γ refers to the efficiency of detecting a radionuclide. The term t stands for the total counting time in seconds (s), and V represents the sample volume in litres (L).

The radium equivalent (Ra_{eq}) is a parameter that characterises the gamma ray output from a sample and its associated radiological hazard. It serves as the key guideline for regulating radiation safety standards. Ra_{eq} is calculated based on the assumption that specific amounts of radiation from ^{226}Ra (370 BqL^{-1}), ^{232}Th (259 BqL^{-1}) and ^{40}K (4810 BqL^{-1}) produce equal gamma radiation dose rates, as given by Equation (2) [27–30].

$$\text{Ra}_{\text{eq}}(\text{BqL}^{-1}) = \frac{A_{\text{Ra}}}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810} \times 370 \quad (2)$$

where A_{Ra} , A_{Th} and A_K represent the average radioactivity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively.

The dose rate (D) refers to the amount of radiation dose absorbed per hour from terrestrial or naturally occurring radioactive materials (NORMs) emitting gamma radiation in the air or surrounding material. The absorbed dose rate is calculated using Equation (5), as outlined in references [30,31].

$$D(\text{nGy h}^{-1}) = 0.462A_{Ra} + 0.621A_{Th} + 0.0417A_K \quad (3)$$

where A_{Ra} , A_{Th} and A_K represent the mean radioactivity concentrations of the radionuclides ^{226}Ra , ^{232}Th and ^{40}K , while the coefficients 0.462, 0.621 and 0.0417 nSv/h/Bq/L are the dose conversion factors.

The total annual effective ingestion dose (TAEID) (mSvy^{-1}) because of various radionuclides in the drinking water was estimated for the members of the public for the age categories of infants (≤ 1 year), children (1–2 years), teenagers (12–17 years) and adults (>17 years). It refers to the radiation dose absorbed or received annually from the ingestion of bottled water and computed using Equations (4) and (5), based on the data from Tables 1 and 2 [1,18,32].

$$\text{AEID}(\text{mSvy}^{-1}) = A_c \times I \times F_{dc} \quad (4)$$

$$\text{TAEID}(\text{mSvy}^{-1}) = \sum (A_c \times I \times F_{dc}) \quad (5)$$

where AEID (mSvy^{-1}) represents the annual effective ingestion dose because of the radionuclide. A_c (BqL^{-1}) stands for the radioactivity concentration of the radionuclides present in bottled drinking water sample, while I denotes the drinking water annual intake rate shown on the 5th row of Table 1, and F_{dc} represents the effective dose coefficient for the relevant radionuclide, for the various age groups as provided by references [30,32].

Table 1. The coefficients for effective dose (SvBq^{-1}) due to natural radionuclide ingestion for age of 70 years by members of the public [1,5,18,32].

Radionuclide	Infants (≤ 1 Year)	Children (1–12 Years)	Teenagers (12–17 Years)	Adults (>17 Years)
^{226}Ra	4.2×10^{-6}	6.2×10^{-7}	1.5×10^{-6}	2.8×10^{-7}
^{232}Th	3.0×10^{-5}	3.4×10^{-6}	5.3×10^{-6}	6.9×10^{-7}
^{40}K	6.2×10^{-8}	2.1×10^{-8}	7.6×10^{-9}	6.2×10^{-7}
Water intake, (I) (Lyear^{-1})	182.625	365.250	730.500	730.500

Due to the sensitivity of the gonads (testes and ovaries) and bone surface with active bone marrow organs to ionising radiation, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has identified them as organs of interest because if they are exposed to radiation, reproductive health could be affected. Hence, the index, annual gonadal equivalent dose (AGED) has been set to serve as the measure of ionising radiation that the gonads received for one year. The AGED (mSvy^{-1}) because of radionuclides ^{226}Ra , ^{232}Th and ^{40}K can be determined by Equation (6).

$$\text{AGED}(\text{Svy}^{-1}) = 3.090 A_{Ra} + 4.180 A_{Th} + 0.314 A_K \quad (6)$$

where A_{Ra} , A_{Th} and A_K represent the activity concentrations for the radionuclides ^{226}Ra , ^{232}Th and ^{40}K (BqL^{-1}), and the coefficients 3.090, 4.180 and 0.314 are the dose conversion factors.

The external and internal hazard indices (H_{ex} and H_{in}) are used to assess the risk from external and internal exposure to radon, a carcinogenic substance, and its short-lived progeny. Both indices are calculated using Equations (7) and (8), and for radiation risk to be considered negligible, each index must be less than one [27,31,33,34].

$$H_{\text{in}} = \frac{A_{\text{Ra}}}{185} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810} \leq 1 \quad (7)$$

$$H_{\text{ex}} = \frac{A_{\text{Ra}}}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810} \leq 1 \quad (8)$$

where A_{Ra} , A_{Th} and A_{K} denote the radioactivity concentration for the natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K , in (BqL^{-1}), while the values at the denominator represent the conversion factors.

Table 2. Results of the radioactivity concentrations (BqL^{-1}) for radioisotopes ^{226}Ra , ^{232}Th and ^{40}K and the total annual effective ingestion dose (TAEID) (mSv^{-1}) according to groupings by age for the natural spring mineral bottled water from South Africa.

Sample ID	Radioactivity Concentrations (BqL^{-1})			Total Annual Effective Ingestion Dose (TAEID) (mSv^{-1})			
	^{226}Ra	^{232}Th	^{40}K	Infants (≤ 1 Year) ($\times 10^{-2}$)	Children (1–12 Years) ($\times 10^{-3}$)	Teenagers (12–17 Years) ($\times 10^{-2}$)	Adults (>17 Years) ($\times 10^{-3}$)
W-1	2.016 ± 0.131	4.164 ± 0.308	228.500 ± 57.100	2.713	7.380	1.960	3.546
W-2	2.571 ± 0.075	4.156 ± 0.108	225.000 ± 56.700	2.752	7.469	2.016	3.639
W-3	2.082 ± 0.128	4.506 ± 0.293	236.900 ± 60.930	2.916	7.884	2.104	3.770
W-4	1.684 ± 0.121	3.931 ± 0.303	237.800 ± 61.800	2.567	7.087	1.838	3.403
W-5	1.911 ± 0.126	5.090 ± 0.097	242.900 ± 59.700	3.228	8.617	2.315	4.057
W-6	2.171 ± 0.128	3.253 ± 0.125	231.000 ± 57.690	2.230	6.303	1.626	3.129
W-7	2.405 ± 0.129	4.644 ± 0.305	228.700 ± 56.700	3.010	8.066	2.188	3.869
W-8	1.553 ± 0.143	7.807 ± 0.099	202.100 ± 10.120	4.639	11.597	3.305	5.168
W-9	1.408 ± 0.106	3.615 ± 0.117	192.200 ± 23.020	2.319	6.282	1.661	2.981
W-10	1.391 ± 0.106	3.915 ± 0.276	207.400 ± 51.530	2.499	6.768	1.783	3.197
W-11	1.060 ± 0.069	1.982 ± 0.124	206.000 ± 51.890	1.410	4.281	0.998	2.149
W-12	1.611 ± 0.110	4.412 ± 0.280	205.500 ± 51.240	2.788	7.420	1.999	3.484
W-13	1.988 ± 0.126	3.787 ± 0.111	238.000 ± 58.870	2.515	6.979	1.816	3.393
W-14	1.482 ± 0.104	1.736 ± 0.112	198.200 ± 49.250	1.303	4.012	0.945	2.076
W-15	2.021 ± 0.126	4.440 ± 0.321	227.600 ± 56.920	2.863	7.717	2.067	3.682
W-16	1.192 ± 0.067	1.973 ± 0.135	229.100 ± 57.160	1.443	4.477	1.022	2.276
W-17	1.196 ± 0.078	2.290 ± 0.199	149.000 ± 38.480	1.526	4.258	1.100	2.074
W-18	1.867 ± 0.124	2.899 ± 0.100	235.000 ± 58.360	2.015	5.825	1.457	2.907
W-19	1.647 ± 0.129	3.059 ± 0.138	233.100 ± 57.870	2.081	5.959	1.494	2.934
W-20	2.039 ± 0.127	2.078 ± 0.134	238.000 ± 36.730	1.583	4.868	1.160	2.542
W-21	1.797 ± 0.120	3.721 ± 0.313	232.800 ± 57.490	2.456	6.813	1.767	3.297
Range	1.060–2.571	1.736–7.807	149.000–242.900	1.303–4.639	4.012–11.597	0.945–3.305	2.074–5.168
Mean	1.766 ± 0.399	3.688 ± 1.371	220.229 ± 22.297	2.422 ± 0.768	6.669 ± 1.771	1.745 ± 0.549	3.218 ± 0.749
[5]	0–0.42	0–0.228	-	0.3	0.3	0.3	0.3
Worldwide	35	30	400	0.2–1	0.2–1	0.2–1	0.1–1

The alpha and gamma level indices (I_{α} and I_{γ}) are used to estimate the risk associated with the annual effective ingested doses from alpha and gamma radiation exposure from these radionuclides. The values of I_{α} and I_{γ} are calculated using Equations (9) and (10). If $I_{\gamma} \leq 2$, the annual effective ingested dose is increased by $0.3 \mu\text{Sv}$, while for $2 \leq (I_{\gamma}) \leq 6$, the dose increases by $1.0 \mu\text{Sv}$. Therefore, I_{γ} can also be used as an indicator for material identification [27,34].

$$I_{\gamma} = \frac{A_{\text{Ra}}}{300} + \frac{A_{\text{Th}}}{200} + \frac{A_{\text{K}}}{3000} \quad (9)$$

$$I_{\alpha} = \frac{A_{\text{Ra}}}{200} \quad (10)$$

A_{Ra} , A_{Th} and A_{K} (BqL^{-1}) represent the radioactivity concentrations of the natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K .

Based on the International Commission on Radiological Protection (ICRP) as well as the South African Water Quality Guideline [5] recommendations covering adult members of the public, the ionizing radiation of stochastic effects from the consumption of bottled water sources were estimated using Equations (11)–(14). Additionally, other health risk parameters because of exposure to radionuclides ^{226}Ra , ^{232}Th and ^{40}K , yielding a low dose of radiation considered to be a chronic risk and having effects on the somatic as well as hereditary organs, have also been estimated using the lifetime fatal cancer risk and hereditary effects coefficient [35,36].

$$\text{FCR} = \text{TAEID} \times \text{CRF}_c \quad (11)$$

$$\text{LFCR} = \text{TAEID} \times A_g \times \text{CRF}_c \quad (12)$$

$$\text{SHE} = \text{TAEID} \times \text{HEF}_c \quad (13)$$

$$\text{ELH} = \text{TAEID} \times A_g \times \text{HEF}_c \quad (14)$$

where FCR represents the fatal cancer risk, TAEID denotes the total annual effective ingestion dose and CRF_c represents the lifetime fatal cancer risk factor ($5 \times 10^{-2} \text{ Sv}^{-1}$). The lifetime fatal cancer risk to adults, A_g , is the adult lifetime age of 70 years [37] of continual exposure to small amounts of ionizing radiation by the population, SHE is the severe hereditary effect, HEF_c is the hereditary effect conversion factor and ELH is the estimated lifetime hereditary effect for the adult group.

3. Results

3.1. Radioactivity Concentration of Radionuclides

The radioactivity concentrations (BqL^{-1}) with their respective uncertainties ($\pm$ standard deviation) for the natural radioisotopes ^{226}Ra , ^{232}Th and ^{40}K found in the natural spring bottled water measured are presented in Table 2, columns 2 to 4. Additionally, the total annual effective ingestion dose (TAEID) (mSvy^{-1}) according to groupings by age as infants (≤ 1 year), children (1–12 years), teenagers (12–17 years) and adults (>17 years) because of the ingestion of radionuclides through natural spring mineral bottled drinking water from South Africa, is given in the same table but in columns 5 to 8, computed using the information in Table 1. The mean radioactivity concentrations of ^{226}Ra varied from $1.060 \pm 0.067 \text{ BqL}^{-1}$ in sample W-11 to $2.571 \pm 0.143 \text{ BqL}^{-1}$ from sample W-2, having average of $1.766 \pm 0.399 \text{ BqL}^{-1}$; ^{232}Th varied from $1.736 \pm 0.112 \text{ BqL}^{-1}$ in sample W-14 to $7.807 \pm 0.099 \text{ BqL}^{-1}$ in W-8, with a mean value of $3.688 \pm 1.371 \text{ BqL}^{-1}$ and ^{40}K ranged from $149.000 \pm 38.480 \text{ BqL}^{-1}$ in W-17 to $242.900 \pm 59.700 \text{ BqL}^{-1}$ in W-5 with a mean of $220.229 \pm 22.297 \text{ BqL}^{-1}$. The TAEID for the infants (≤ 1 year) varied from 1.303 to $4.639 \times 10^{-2} \text{ mSvy}^{-1}$, having an average of $(2.422 \pm 0.768) \times 10^{-2} \text{ mSvy}^{-1}$, children (1–12 years) ranged between 4.012 and $11.597 \times 10^{-3} \text{ mSvy}^{-1}$, with a mean of $(6.669 \pm 1.771) \times 10^{-3} \text{ mSvy}^{-1}$, teenagers (12–17 years) between 0.945 and $3.305 \times 10^{-2} \text{ mSvy}^{-1}$, having an average of $(1.745 \pm 0.549) \times 10^{-2} \text{ mSvy}^{-1}$, while for adults (>17 years), it ranged from 2.074 to $5.168 \times 10^{-3} \text{ mSvy}^{-1}$, having an average of $(3.218 \pm 0.749) \times 10^{-3} \text{ mSvy}^{-1}$. The distribution of the radioactivity concentrations of naturally occurring radioisotopes are also presented in Figure 3. Both the radioactivity concentrations of the naturally occurring radioisotopes and the total annual effective ingestion dose are variable across the samples and comparable to similar studies in the literature [31,38].

The estimated contribution of each analysed natural radionuclide to the total annual effective ingestion dose for different age groups in natural spring mineral bottled water is presented in Figure 4. The results show that ^{232}Th accounted for the highest percentage

of the dose, contributing 83.44% for infants (≤ 1 year), followed by 81.89% for teenagers (12–17 years), 68.68% for children (1–12 years) and 57.78% for adults (>17 years). ^{40}K contributed an average percentage to the dose of 31.00% for adults (>17 years), followed by 25.33% for children (1–12 years), 10.30% for infants (≤ 1 year) and 7.01% for teenagers (12–17 years). ^{226}Ra contributed the lowest percentage to the dose of 11.23% for adults (>17 years), followed by 11.10% for teenagers (12–17 years), 6.26% for infants (≤ 1 year) and 6.00% children (1–12 years).

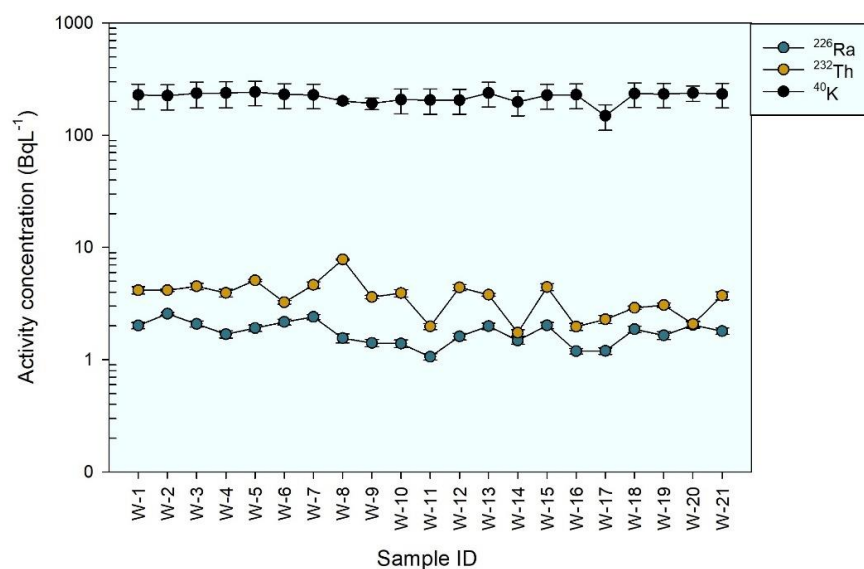


Figure 3. The radioactivity concentration (BqL^{-1}) distribution for the naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K present in commercial bottled water of South Africa.

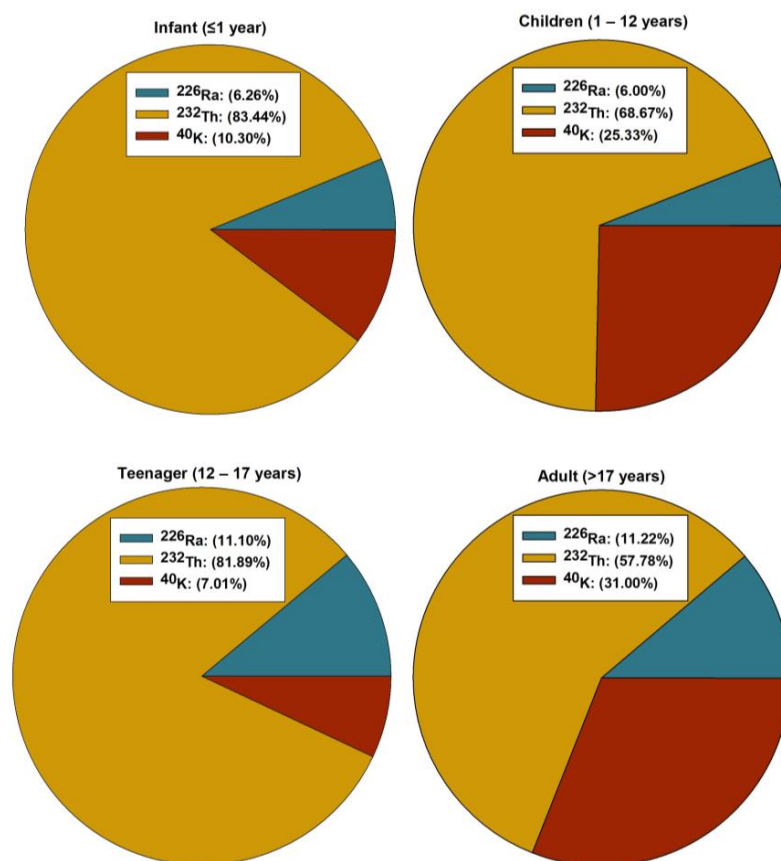


Figure 4. Contribution of each of the analysed natural radioisotopes ^{226}Ra , ^{232}Th and ^{40}K to the total annual effective ingestion dose for the groupings by age, for bottled water from South Africa.

3.2. Radium Equivalent and Absorbed Dose

The parameter radium equivalent (Ra_{eq}) (BqL^{-1}), which is the index for the combined radioactivity concentrations for the naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K within the natural spring mineral bottled drinking water, was evaluated using Equation (2), and the result is presented in Figure 5. The result of Ra_{eq} showed a maximum activity value of $28.252 \pm 0.613 BqL^{-1}$ and a minimum activity value of $15.929 \pm 0.268 BqL^{-1}$, which are the respective radium equivalent activity values of sample W-8 and sample W-17, having a mean of $23.976 \pm 0.446 BqL^{-1}$. The parameter absorbed dose rate (D_{Ab}) ($nGyh^{-1}$), was also estimated utilizing Equation (3), and the result is presented in Figure 5. The results show that the maximum value of the absorbed radiation dose rate is $14.092 \pm 0.436 nGh^{-1}$ from sample W-5, while the minimum value is $8.152 \pm 0.207 nGyh^{-1}$ from sample W-17, with a mean of $12.232 \pm 1.445 nGyh^{-1}$.

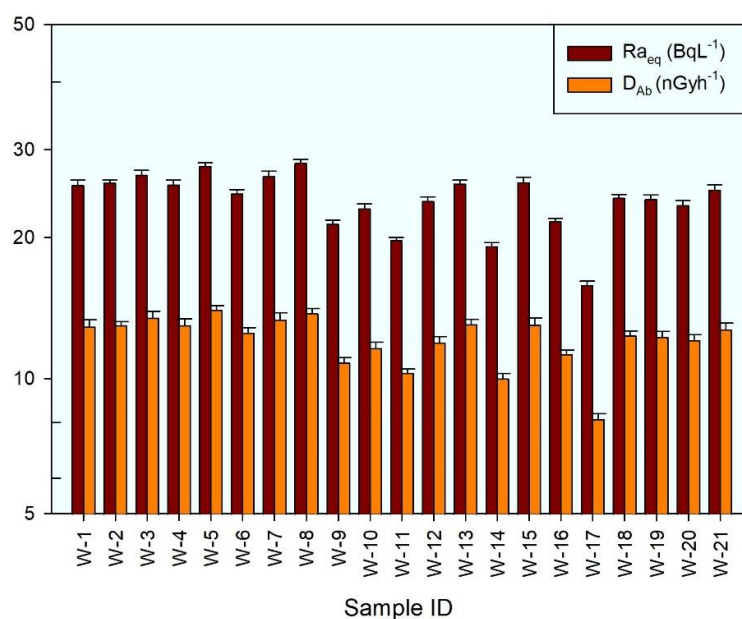


Figure 5. Result of the radium equivalent (Ra_{eq}) (BqL^{-1}) and absorbed dose (D_{ab}) ($nGyh^{-1}$) for the water samples based on the radionuclides ^{226}Ra , ^{232}Th and ^{40}K .

3.3. Annual Gonadal Equivalent Dose (AGED) and Hazard Indices (H_{ex} , H_{in} , I_{γ} and I_{α})

The estimated results of the parameters annual gonadal equivalent dose (AGED), internal and external hazard indices (H_{in} and H_{ex}) and representative alpha and gamma indices (I_{α} and I_{γ}) are presented in Figure 6a. The presented results indicate that the annual effective ingestion dose (AEID) varies between $0.040 \pm 0.038 mSvy^{-1}$ and $0.069 \pm 0.054 mSvy^{-1}$ in samples W-17 and W-5, having a mean of $0.060 \pm 0.007 mSvy^{-1}$. AGED has a maximum figure of radiation dose from drinking water as $0.103 \pm 0.037 mSvy^{-1}$ and a minimum value of $0.060 \pm 0.017 mSvy^{-1}$ in samples W-5 and W-17, respectively, with a mean of $0.090 \pm 0.027 mSvy^{-1}$. The computed averages of the radiological health risk indices H_{ex} , H_{in} , I_{γ} and I_{α} are 0.065 ± 0.008 , 0.070 ± 0.009 , 0.098 ± 0.012 and 0.009 ± 0.002 , respectively, as shown in Figure 6a.

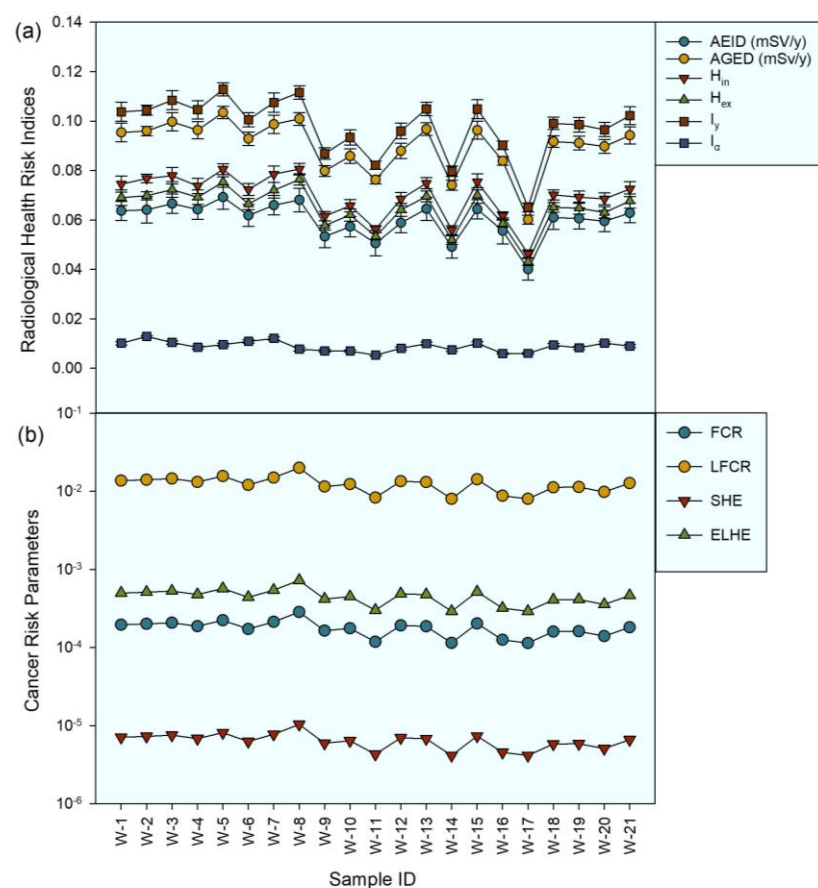


Figure 6. Estimated results for the radiological health risk indices and cancer risks and hereditary effects for the public because of the intake of natural spring mineral bottled water from South Africa. Panel (a) shows the radiological health risk parameters and panel (b) shows the cancer risk and hereditary effect parameters.

3.4. Cancer Risks and Hereditary Effects

Figure 6b presents the computed results for the parameters, cancer risks and hereditary effects, because of ingesting natural spring bottled drinking water containing naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K . The fatal cancer risk (FCR) varies between 1.141×10^{-4} and 2.842×10^{-4} , having an average of $1.769 \times 10^{-4} \pm 4.119 \times 10^{-5}$. The lifetime fatality cancer risk (LFCR), taking into cognizance the reference human age of 70 years, varies between 7.983×10^{-3} and 19.897×10^{-3} , having an average of $12.389 \times 10^{-3} \pm 2.883 \times 10^{-3}$. The severe hereditary effect (SHE) to adults per year range between 4.147×10^{-6} and 10.336×10^{-6} , having a mean value of $6.436 \times 10^{-6} \pm 1.498 \times 10^{-6}$, while the estimated lifetime hereditary effect (ELHE) varies from 2.903×10^{-4} to 7.235×10^{-4} with a mean of $4.505 \times 10^{-4} \pm 1.049 \times 10^{-4}$. These results are lower but comparable to those reported by [36].

3.5. Statistical Analysis

The descriptive statistical analysis (minimum, maximum, mean, standard deviation (STD DV), range, variance, skewness and kurtosis) of the determined radioactivity concentrations levels of the naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K together with the estimated radiological health risk indices in the natural spring bottled water are shown in Table 3, while the frequency distribution with the Gaussian distribution curve is presented in Figure 7. The statistical analysis was performed using the following statistical packages, MS Excel, SigmaPlot version 15.0 and IBM SPSS Statistics 29.0.0.0.

Table 3. Descriptive statistics for the radionuclides and radiological health risk parameters.

Parameter	No	Minimum	Maximum	Mean	Stdev	Range	Variance	Skewness	Kurtosis
^{226}Ra	21	1.060 ± 0.067	2.571 ± 0.143	1.766	0.398	1.511	0.1598	0.07099	−0.47215
^{232}Th	21	1.736 ± 0.097	7.807 ± 0.321	3.688	1.371	6.071	1.879	1.069	2.922
^{40}K	21	149.0 ± 10.120	242.9 ± 50.931	220.229	22.297	93.9	497.161	−1.816	4.029
Ra_{eq}	21	15.929 ± 0.268	28.252 ± 0.613	23.976	3.033	12.323	9.201	−1.056	1.144
TAEID Infants	21	0.013	0.046	0.024	0.008	0.033	5.894×10^{-5}	0.874	2.243
TAEID Children	21	0.004	0.012	0.007	0.002	0.008	3.137×10^{-6}	0.698	1.726
TAEID Teenagers	21	0.009	0.033	0.017	0.005	0.024	3.017×10^{-6}	0.799	1.992
TAEID Adults	21	0.002	0.005	0.003	0.001	0.003	5.609×10^{-7}	0.430	1.012

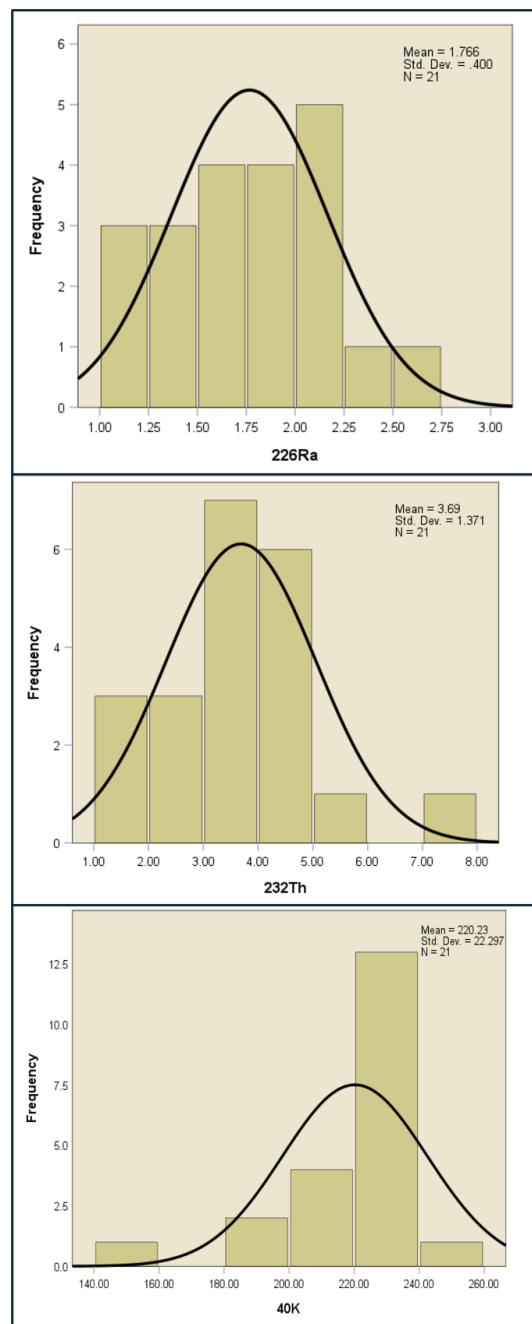
**Figure 7.** Frequency distributions with Gaussian distribution curve of the radioactivity concentrations for the radioisotopes ^{226}Ra , ^{232}Th and ^{40}K present in bottled water.

Table 4 presents the computed Pearson's correlation coefficients for the relationship between the radioactivity concentrations for naturally occurring radioisotopes ^{226}Ra , ^{232}Th

and ^{40}K , and the estimated radiological health risk indices. The purpose is to ascertain the order of mutual relationships between the pairs of the variables determined. It also tests the inter-relationship between the different models used in the estimation of the radiological health risks, as well as the cancer risk and hereditary effects. The correlation could be positive or negative, whereas the level of significance depends on whether the p -Value is < 0.050 . Inter-relationships between naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K showed a weak positive correlation (p -Values in parentheses) of 0.331 (0.142), 0.598 (0.004) and 0.137 (0.554), implying that although the pairs increase together, there is no significant relationship between them, with p -Values > 0.050 except for ^{232}Th . This explains why both ^{226}Ra and ^{232}Th are from different naturally occurring radionuclide decay series, while ^{40}K is a non-decay series radioisotope. However, a strong positive correlation was observed between the three natural radionuclides and the radium equivalent index of 0.684, 0.767 and 0.733, with their respective p -Values < 0.050 , evidence of significant correlation between the pairs, implying that the radionuclides all contributed to the index, proportionately. In a similar vein, a strong positive correlation was observed among the three naturally occurring radioisotopes and the corresponding radiological health risk variables, in the range of 0.881 to 1.000, except for ^{226}Ra and ^{40}K which are low, having uniform values of 0.327 and 0.496, which reflect their individual contributions to the parameters as shown also in Figure 4.

Table 4. The Pearson's correlation coefficient analysis for the radioisotopes ^{226}Ra , ^{232}Th and ^{40}K and radiological health risks.

Parameter	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	TAEID Infants (≤ 1 Year)	TAEID Children (1–12 Years)	TAEID Teenagers (12–17 Years)	TAEID Adults (> 17 Years)	FCR	LFCR	SHE	ELH
^{226}Ra	1	0.331	0.598	0.684	0.389	0.428	0.414	0.496	0.496	0.496	0.496	0.496
^{232}Th		1	0.137	0.767	0.998	0.991	0.996	0.977	0.977	0.977	0.977	0.977
^{40}K			1	0.733	0.194	0.259	0.203	0.327	0.327	0.327	0.327	0.327
Ra_{eq}				1	0.805	0.843	0.812	0.881	0.881	0.881	0.881	0.881
TAEID Infants (≤ 1 year)					1	0.998	1.000	0.989	0.989	0.989	0.989	0.989
TAEID Children (1–12 years)						1	0.998	0.996	0.996	0.996	0.996	0.996
TAEID Teenagers (12–17 years)							1	0.991	0.991	0.991	0.991	0.991
TAEID Adults (> 17 years)								1	1.000	1.000	1.000	1.000
FCR									1	1.000	1.000	1.000
LFCR										1	1.000	1.000
SHE											1	1.000
ELH												1

The assessment of physicochemical parameters, trace elements and heavy metal concentration levels due to the consumption of natural spring mineral bottled water from South Africa and the associated carcinogenic and non-carcinogenic risks have been reported [13].

The statistical analysis of the relationship between the naturally occurring radioisotope (^{226}Ra , ^{232}Th and ^{40}K) radioactivity concentrations and potential hydrogen (pH) together with the total dissolved solids (TDS) (ppm) has been performed. The purpose is to evaluate the natural spring bottled water geochemical properties' correlation with the natural radionuclides, and the results are presented in Figures 8a–c and 9a–c, respectively.

A very low positive correlation was observed between the natural radionuclides' radioactivity concentrations for ^{226}Ra and ^{232}Th and pH for the natural spring bottled water, which was not significant, with the regression curve fit indicating that both variables

increased together, while no correlation was found between ^{40}K and pH. The results of these trends have been reported by [21,38]. The general trend observed concerning correlation of radioactivity concentrations for natural radioisotopes and TDS, where the correlation coefficients of the radionuclides ^{226}Ra , ^{232}Th , and ^{40}K have values of 0.2110, 0.0019 and 0.0136, respectively, showed that ^{226}Ra seemed to present more relations than others due to its ability to readily dissolve in water rich in minerals.

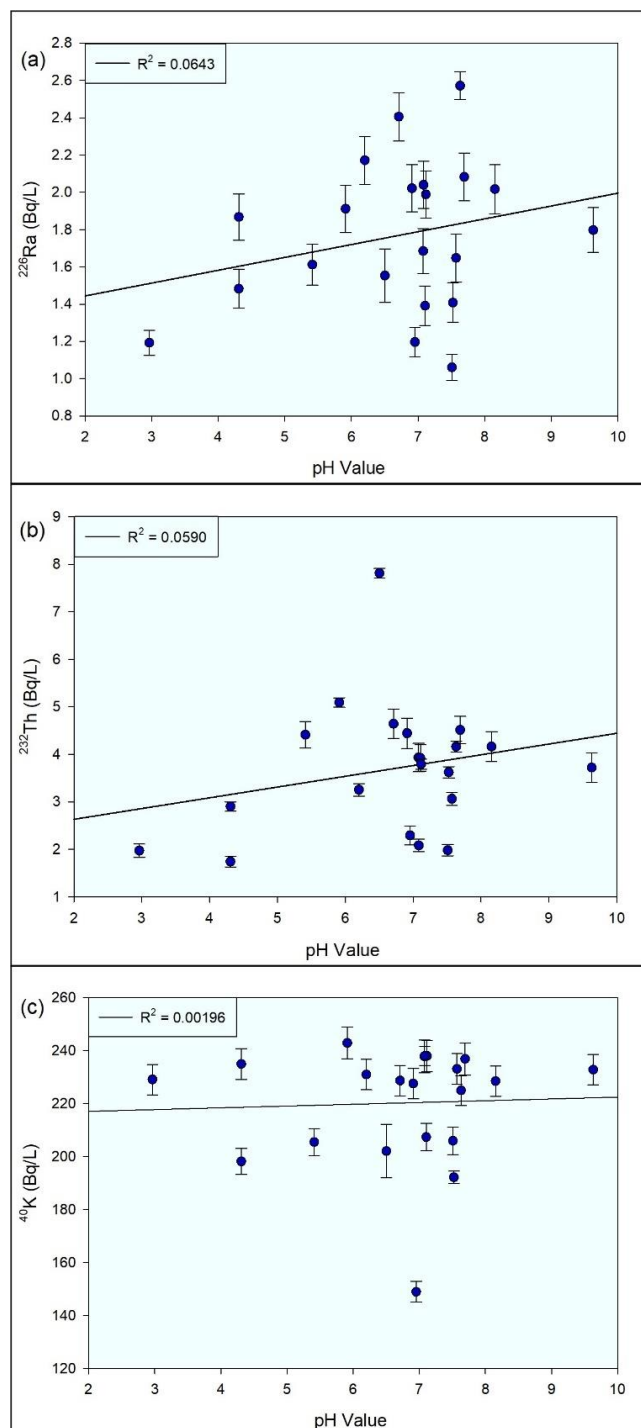


Figure 8. The relationship between the natural radionuclide radioactivity concentrations (BqL^{-1}) and the pH for (a) ^{226}Ra , (b) ^{232}Th and (c) ^{40}K for the bottled drinking water.

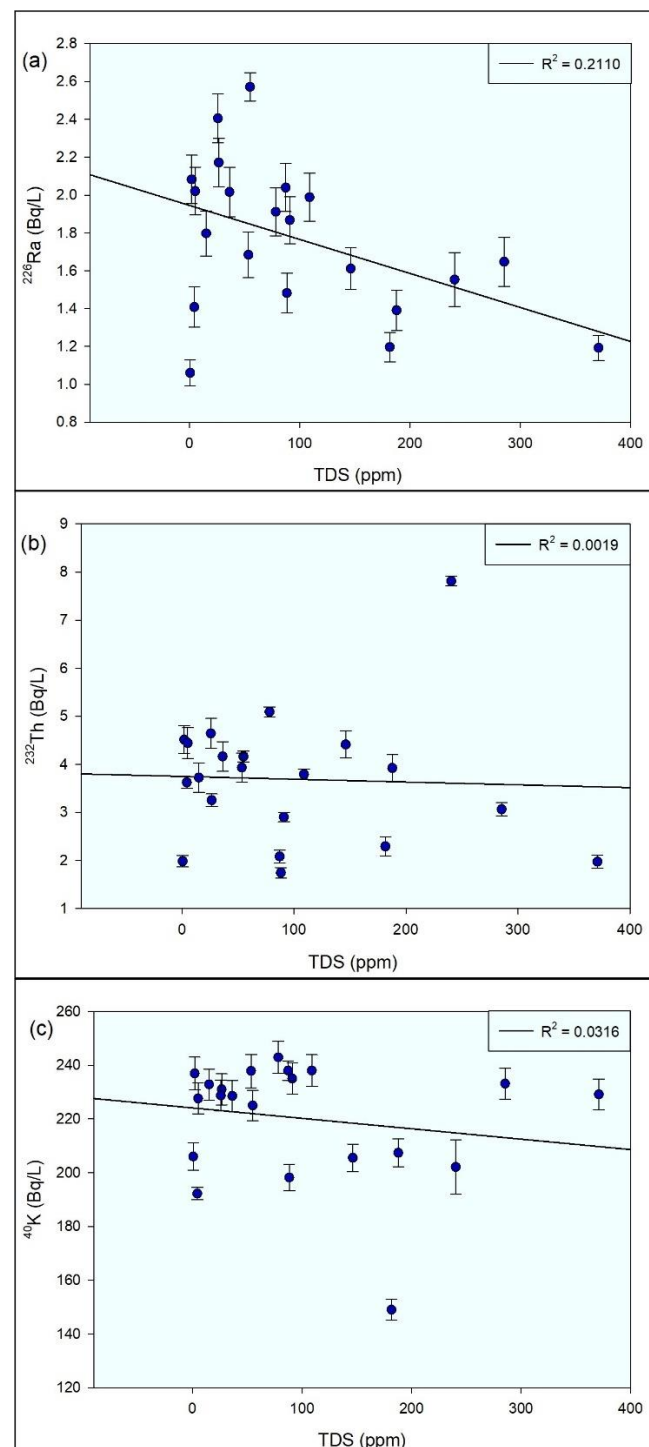


Figure 9. The relationship between total dissolved solids (TDS) (ppm) and radionuclide radioactivity concentrations (BqL^{-1}) for (a) ^{226}Ra , (b) ^{232}Th and (c) ^{40}K for the natural spring bottled water.

Table 5 presents the comparison of radioactivity concentrations for naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K (BqL^{-1}) and the associated radiological health risk variable, total annual effective ingestion dose (TAEID) (mSvy^{-1}), in this study and other nations of the world. The radioactivity concentrations in bottled water of African countries including Morocco, Egypt and Nigeria ranged between 0.00194 and 11.100 BqL^{-1} , for ^{226}Ra , 0.00246 and 4.600 BqL^{-1} for ^{232}Th and 0.237 and 28.500 BqL^{-1} for ^{40}K , whereas the TAEID values varied from $0.991 \times 10^{-3} \text{ mSvy}^{-1}$ (for Morocco) to 3.300 mSvy^{-1} (for Nigeria) [18,20]. The value for the radioactivity concentrations for South Africa from this study is found

to be within the range of those reported from other African countries, except for ^{40}K , whose values are much greater. The radioactivity concentrations in bottled drinking water of countries other than Africa ranged between 0.105×10^{-3} and 4.900 BqL^{-1} for ^{226}Ra , 0.016×10^{-3} and 1.600 BqL^{-1} for ^{232}Th and 0.157 and 10.500 BqL^{-1} for ^{40}K , whereas the TAEID values varied from 0.0016 mSvy^{-1} (for Croatia) to 0.500 mSvy^{-1} (for Spain) [16,22]. It is clearly observable that the radioactivity concentrations for natural radioisotopes in this work compare well with those reported by [31,38].

Table 5. Comparison of mean and/or range of radioactivity concentrations for natural radioisotopes ^{226}Ra , ^{232}Th and ^{40}K (BqL^{-1}) as well as total annual effective ingestion doses (TAEIDs) (mSvy^{-1}) of various origins.

Origin	^{226}Ra (BqL^{-1})	^{232}Th (BqL^{-1})	^{40}K (BqL^{-1})	TAEID (mSvy^{-1})	Reference
South Africa	1.766 ± 0.399	3.688 ± 1.371	220.229 ± 22.297	0.01275 ± 0.004	Present study
Iraq	1.19 ± 0.5	0.96 ± 0.2	10.5 ± 0.5	0.015 ± 0.0025	[31]
Morocco	0.00194	0.00246	0.2366	0.991×10^{-3}	[20]
Egypt	1.6–11.1	0.21–0.97	9.1–23.0	0.67	[38]
USA	0.052	0.052	-	0.027	[39]
Croatia	0.0422 ± 0.0032	0.0219 ± 0.0035	-	0.0016–0.0115 (NSW); 0.0109–0.0183 (MW)	[16]
Nigeria	4.3 ± 0.8	4.6 ± 0.5	28.5 ± 3.0	3.3	[18]
Spain	0.02–4.0	-	-	0.5	[22,40]
Pakistan	0.0113 ± 0.0023	0.0052 ± 0.0004	0.140 ± 0.0306	0.0041	[21]
Sweden	4.9	1.6	-	0.45	[41]
Serbia	<0.070	<0.050	<0.250	0.0092	[22]
Poland	0.0008–0.437	-	0.157–10.064	-	[22]
France	0.007–0.70	-	-	-	[22]
Germany	0.00085–0.35	-	-	-	[22]
Hungary	0.0043–0.910	-	-	-	[22]
Saudi Arabia	$(0.105\text{--}0.568) \times 10^{-3}$	$(0.016\text{--}0.382) \times 10^{-3}$	18.84×10^{-3}	-	[20]
Malaysia	$(0.7\text{--}7.03) \times 10^{-3}$	$(0.55\text{--}8.64) \times 10^{-3}$	$(22\text{--}53) \times 10^{-3}$	-	[20]
SAWQG	0–0.42	0–0.228	-	0.3	[5]
Worldwide	35	30	400	0.2–1	[30]

4. Discussion

The naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K abound everywhere in the natural environment at various concentration levels, including in foodstuff and water, and are ingested by humans, releasing a radiation dose to vital organs and contributing to the risk of developing various cancers [3,19,42]. This study discusses activity concentrations for naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K (BqL^{-1}) and the related radiological health risks as a result of ingesting mineral spring bottled water. From the findings, all the radionuclides being studied were detected at various concentrations within natural spring bottled drinking water. The radioactivity concentration of ^{40}K was found to be greater than that of ^{226}Ra and ^{232}Th , showing a pattern of activity concentrations in the order of $^{40}\text{K} > ^{232}\text{Th} > ^{226}\text{Ra}$, comparable to that reported by [18,20]. Potassium-40, an alpha emitter, has been described as being found everywhere, both in all water supplies and in all living organisms, because it is a very useful intracellular mineral and is homeostatically controlled [3,5,30]. The mean activity concentration levels exceed the recommended guideline limit for drinking water by the South African Water Quality Guideline but are way less than the worldwide average value [5,20,30]. The reason could be due to the diverse geology of South Africa, which is rich in NORMs such as uranium and thorium, anthropogenic factors, including mining activities, and the industrial processing of bottled drinking water. However, the estimation of TAEID presents overall results that are way below the recommended dose limit of 0.1 mSvy^{-1} for all the age groups assessed. The assessment of the effective ingestion dose showed that the age groups each receive doses in the descending order of infants (≤ 1 year) > teenagers (12–17 year) > children (1–12 year) > adults (>17). It shows that ^{232}Th or its progeny ^{228}Ra which is radiotoxic usually con-

tributes more proportionally to the radiation dose in water and especially is highest in the infant age group. It therefore requires remediation action, especially because infants are more sensitive and vulnerable to radiation exposure. This result is also way below the ambient and cosmic (above sea level) levels of background radiation reported for South Africa and the global average of $1.5\text{--}3.0\text{ mSv y}^{-1}$ and 0.3 mSv y^{-1} , respectively. It implies that the mineral spring bottled water from South Africa is safe for drinking according to global guideline limits for radionuclide contamination [30]. It will not pose any significant radiological health risk to consumers in the interim.

Radiological health risk models usually estimate the impact of naturally occurring radioisotope radioactivity concentrations on human organs due to ingestion of foodstuff and water. The parameters utilized in this study, which include the radium equivalent, absorbed dose, annual gonadal equivalent dose, hazard indices and representative alpha and gamma indices, presented values that are within the acceptable limits, with the following implications. The Ra_{eq} findings will provide a base guideline which will be used to regulate safety standards regarding the consumption of bottled water [29]. The absorbed dose gives an idea of the risk of exposure as the water is consumed, also bearing in mind the stochastic effect from the accumulation of small doses over prolonged periods of consumption. The gonads and bone surfaces, which are very sensitive to radiation dose, will receive very minimal AGED according to this study's findings. Meanwhile, the internal and external exposure to radon, which is short-lived and carcinogenic, as well as the representative alpha and gamma indices, from the bottled water is less than 1, and hence offers negligible levels of radiation risk.

The lifetime cancer risks and severe hereditary effects to adults due to the consumption of bottled water as estimated could be interpreted to mean that 12 persons out of 1000 may likely present some kind of cancer fatality, while 6 persons out of 1 million may present some kind of hereditary effect. However, the findings present values to be within the acceptable guidance limit set by the United States Environmental Agency (USEPA) of 1.0×10^{-6} to 1.0×10^{-4} , which is interpreted to mean 1 person in a million to 10,000 persons may present some kind of cancer fatality [35,36].

The statistical analysis of data in this study indicates that, for the 21 samples, the values of the standard deviation are less than the values of the corresponding means for the natural radioisotopes ^{226}Ra , ^{232}Th and ^{40}K as well as the radiological health risk parameters. This implies that the distribution of the data has a very high degree of uniformity, and it is comparable with the results reported by [34], but differs from [36]. The frequency distribution of the naturally occurring radioisotopes shows that ^{226}Ra and ^{232}Th and TAEID for the various age groups are skewed to the right or are slightly asymmetric (positively skewed), having a distribution tail pointing to the right with a value of skewness greater than 0, while ^{40}K and Ra_{eq} are skewed to the left or slightly asymmetric (negatively skewed), also having a skewness value less than 0, as shown in Table 3, column 9. The skewness of the data indicates their nature or extent of symmetry, with perfect symmetry at zero; hence, the frequency distribution of the radionuclides and the corresponding radiological health risk parameters in this study are only slightly asymmetric. Additionally, the kurtosis of the radionuclide concentrations presents a negative value for ^{226}Ra , while ^{232}Th , ^{40}K , Ra_{eq} and TAEID for various age groups showed positive values (Table 3, column 10). There is an indication that the frequency distribution curve of the radionuclides is less peaked than the normal curve, known as platykurtic, for ^{226}Ra , while being more peaked than the normal curve, called leptokurtic, for ^{232}Th , ^{40}K , Ra_{eq} and TAEID, for the various age groups. The behaviour of the statistical frequency distribution in this study could be because of the unequal distributions of naturally occurring radioisotopes within natural spring mineral drinking water, especially from bedrock source aquifers. The Pearson's

correlation coefficient used in this study presents a very low positive correlation between the naturally occurring radioisotopes' radioactivity concentrations for ^{226}Ra and ^{232}Th and pH for drinking water, which was not significant with the regression curve fit, indicating that both variables increased together, while no correlation was found between ^{40}K and pH. Findings with this kind of trend have been reported by [21,38]. The impact of pH on the variable characteristics of ^{238}U and ^{232}Th decay series and acid leaching could be responsible for the findings in this study. With an increase in pH, uranium and thorium, as well as other minerals, are usually leached in the surrounding rocks, and likewise in the sediment [43]. The general trend observed concerning the correlation of radioactivity concentrations of natural radioisotopes and the TDS, where the correlation coefficients of radioisotopes ^{226}Ra , ^{232}Th and ^{40}K present values of 0.2110, 0.0019 and 0.0136, respectively, showed that ^{226}Ra seemed to present more relations than others due to its ability to readily dissolve in water rich in minerals.

The findings of this study, when compared to those from other countries, suggest that the activity concentrations of naturally occurring radioisotopes ^{226}Ra , ^{232}Th and ^{40}K are similar to those reported in studies from other African countries [20,38], with values lower than [18]. However, this study's values are higher than those reported in non-African countries [16,22], yet all the values are within the permissible limit set by [5,30], implying that the bottled drinking water is radiologically reliable for consumption.

5. Conclusions

The assessment of radioactivity concentration levels in natural spring mineral bottled commercial drinking water in South Africa using HPGe gamma spectrometry, has provided significant insights into the radiological quality of these water sources. This study revealed varying levels of naturally occurring radionuclides across different brands, with mean values of $1.766 \pm 0.399 \text{ BqL}^{-1}$ for ^{226}Ra , $3.688 \pm 1.371 \text{ BqL}^{-1}$ for ^{232}Th and $220.229 \pm 22.297 \text{ BqL}^{-1}$ for ^{40}K . The related radiological health risks of these concentrations have been revealed to be less than the acceptable safety guideline limits proposed by UNSCEAR, indicating that the consumption of natural spring mineral bottled water in South Africa is radiologically harmless.

However, our findings also highlight the associated potential health risks for the prolonged consumption of bottled drinking water containing elevated radionuclide concentration levels. The effective ingestion dose from drinking water is received by different age groups in the following order: infants (≤ 1 year) > teenagers (12–17 years) > children (1–12 years) > adults (17 years). Among the radionuclides, ^{232}Th contributes the highest radiation dose, accounting for 83.44% in the infant group, while ^{226}Ra contributes the least, with 6.00% in the children group. ^{40}K falls in between, with contributions ranging from 7.01% in teenagers to 31.00% in adults.

Based on our findings, approximately 12 out of 1000 individuals may suffer a cancer fatality and 6 out of 1 million individuals may experience hereditary effects because of the consumption of natural spring bottled drinking water over their lifetime. While the immediate risk appears exceptionally low and provides overall safe water quality, the cumulative effects over the lifetime could pose significant health hazards. This emphasizes the need for public awareness and education on the importance of choosing safe drinking water.

The findings of this study align with those reported in the literature, showing lower levels than those observed in some African countries, but higher than those found in non-African countries. This highlights the importance of geological location variations in radionuclide concentrations. This study is limited to assessing the activity concentration levels and estimation of the radiological health risks in bottled drinking water. Further

research is recommended on how to mitigate activity levels and associated health risk in drinking water.

The results of this study call for a collaborative effort between regulatory agencies, water bottling companies and health organisations to strengthen the enforcement of guidelines that ensure safe and good quality bottled water. Regular testing and transparent reporting of radionuclide levels should be mandated to prevent possible radiological health risks.

In conclusion, this research contributes to a better understanding of the radiological quality of the natural spring bottled drinking water of South Africa and serves as a foundation for future studies and policy development aimed at ensuring safe drinking water for all, in alignment with the global Sustainable Development Goals. By addressing the identified radiological risks, proactive steps can be taken towards safeguarding public health and enhancing the overall quality of bottled drinking water available to consumers.

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