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Natural Radioactivity and Associated Radiological Hazard in Ceramic and Porcelain Sanitary Ware Products Sold in Kenya

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Abstract

The common materials used in the production of sanitary wares for bathrooms, sinks, and toilets are ceramic and porcelain. The materials for the production of these products include a mixture of zinc oxide, feldspar, kaolin, and zircon, which contribute to natural radioactivity. We have determined natural radioactivity levels (^{226}Ra, ^{232}Th, and ^{40}K) in sanitary ware products sold in Kenya and investigated the radio-logical hazards associated with the use of the sanitary ware products. Using a thallium-doped sodium iodide detector, NaI (Ti), the distribution of the terrestrial radioisotopes ^{226}Ra, ^{232}Th, and ^{40}K for 8 different brands of ceramic and porcelain sanitary wares that are widely used domestically was determined. The mean concentration based on production material for ceramics was 24.75 ± 1.2425 (Bq/Kg), 66.4 ± 3.3175 (Bq/Kg), and 444.5 ± 22.235 (Bq/Kg) for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The mean concentration for porcelain was 14 ± 0.72 (Bq/Kg), 83.175 ± 4.16 (Bq/Kg), and 401.5 ± 20.135 (Bq/Kg) for ^{226}Ra, ^{232}Th, and ^{40}K, respectively**.** The activity concentration was lower than the average limits of 50 and 500 (Bq/Kg), respectively, except for values of ^{232}Th for both ceramic and porcelain, which had a slightly higher average compared to the world average limits of 50Bq/kg. The average of radiological parameters (Ra_{eq}, lγ, H_{ex}, H_{in}, ADR, $AEDE_{in}$, $AEDE_{out}$, and ELCR) was calculated as 158.63 ± 7.95 (Bq/kg), 1.113 ± 0.05, 0.425 ± 0.02, 0.375 ± 0.02, 72.5 ± 3.65 (nGy/h), 0.179 ± 0.009, 0.125 ± 0.003, and 0.942, respectively. The radio-logical parameters were found to have lower values than the limits recommended by international bodies, except for ELCR, $_{\text{which}}$ had slightly higher values. The values were compared with the prescribed limits set by commis-

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sions and organizations concerned with radiation protection (the WHO, ICRP, UNSCEAR, and EC) to ensure the safe use of the sanitary ware products investigated. The results showed that materials made of ceramic and porcelain as production material for sanitary wares are safe to be used in building and construction, but the manufacturers of the products should check the levels of ^{232}Th to ensure they are within the acceptable levels.

Subject Areas

Environmental Radioactivity, Radiation Physics

Keywords

Natural Radioactivity, Ceramic and Porcelain Sanitary Wares, Radiological Hazard Indices

1. Introduction

Modern buildings are fitted with sanitary ware in bathrooms, sinks, and toilets. The common materials used in the production of sanitary wares are ceramic and porcelain. The materials for the production of these products include a mixture of zinc oxide, feldspar, kaolin, and zircon as an opacifying constituent [1]. The glaze zircon shows an elevated concentration of natural radioactivity significantly higher than the permissible limits [2]. High or elevated levels of radioactivity in construction materials increase internal and external exposure [3]. The sanitary ware market in the world has a trading volume of approximately 9.1 billion Euros [4], and radioactivity levels of sanitary ware are crucial in assessing the radiological risks owing to radiation exposures and developing guidelines for these materials. The sanitary ware market is expected to witness a compounded growth and high usage, attributed to the changing lifestyles of people regarding home decoration products. Kenya has approximately 12.1 million households according to the Kenya National Bureau of Statistics and faces an annual housing demand of about 250,000 units, far exceeding current supply; this growing deficit has significant implications for sanitation infrastructure needs as each new housing unit requires associated sanitation facilities [5] [6]. This makes sanitary wares to be used at an increased rate by Kenyans, and hence the need to determine natural radioactivity associated with the use of sanitary ware as decoration in buildings. The fact that sanitary ware elements are fundamental building materials, an investigation of their radio-logical impact on members of the public is necessary, with emphasis on gamma radiation and inhalation of radon released from the product as the main exposure pathway of the radionuclides [7] [8]. Epidemiological studies substantially link raised levels of radon exposure in houses to an increased risk of lung cancer [9]. Research on ceramic tiles for flooring or walling has been done and documented [10], but little has been done on sanitary ware based on production materials (ceramic and porcelain). Therefore, the general objective of this study is to comparatively determine natural radioactivity levels (^{226}Ra, ^{232}Th, and ^{40}K) in sanitary ware products based on the material of production (ceramic and porcelain) sold in Kenya. The research also investigated the radio-logical hazards associated with the use of sanitary ware products.

2. Materials and Methods

2.1. Collection of Samples

Ceramic and porcelain pieces of closet and sink samples as end-user products for building and sanitation elements were purchased from different local hardware stores around Kenyan towns. The collected samples were labeled according to the material of production. Each sample collected represented a pooled composite from several items of each production material. The ceramic samples were manufactured in China, and porcelain was manufactured in India. The sampling unit included retail hardware shops, building material depots, sanitary ware showrooms, wholesale distributors, and construction supply outlets.

2.2. Preparation of Samples

The sanitary ware collected from the large distributing hardware stores was broken into small sizes with a sledgehammer. Then the small pieces were reduced further by using a grinding mill for triturating. The ground samples were then sieved through a fine 0.6 mm mesh to obtain a homogenized particle size, and each sample had a mass of about 300 g. They were finally filled in uncontaminated sealed empty uniform-sized cylindrical standard 1000millimeter plastic containers of uniform size to avoid escape of gaseous radon ^{222}Rn at airtight conditions. They were stored for four weeks (28 Days) before analysis at airtight conditions to obtain secular equilibrium between radium and its short-lived decay products. The sanitary ware samples were labeled and taken for measurement, where the levels of radiation present were determined.

2.3. Determination of Radioactivity

The radioactivity levels (^{226}Ra, ^{232}Th, and ^{40}K) content and risk assessment were determined for porcelain and ceramic production material samples. A gamma spectrometry system model 802-4 thallium-doped sodium iodide detector NaI (Tl) crystal with a resolution range from 7.5 to 8.5% at a 662 keV peak of Cs-137, connected to a personal computer analyzer using Win TMCA32 software, was employed for these measurements. The detector system comprises built-in electronic modules plugged into a PC via a USB link. The system also includes an Oxford PCA-P multichannel analyzer (MCA) card and its software for spectral data acquisition and analysis. The PCA-P comprises a high voltage supply, a charge sensitive pre-amplifier, a shaping amplifier, an 80 MHz Wilkinson analog to digital converter (ADC) with a multichannel analyzer (MCA).

The gamma spectrometer detector was calibrated before it was used for analysis. The energy peaks of 662 keV of ^{137}Cs, 1170 keV and 1330 keV of ^{60}Co were used to calibrate the spectrometer [11]. The background radioactivity distribution in the environment around the detector was determined by counting plastic containers filled with distilled water in the same manner and in the same geometry as the samples. The background measurements were repeated at regular intervals for quality control with the assistance of three reference materials (RGU-1, RGTH-1, and RGK-1) obtained from the International Atomic Energy Agency (IAEA). The background radiation was subtracted from each of the recorded spectra [12]. Standard IAEA-certified samples of ^{238}U, ^{232}Th, and ^{40}K were analyzed under identical experimental conditions to validate analytical procedures. The peaks corresponding to 1460 keV (^{40}K) for ^{40}K, 1765 keV (Bi-214) for ^{238}U, and 2615 keV (Tl-208) for ^{232}Th were considered in arriving at the activity levels. The energy and efficiency calibrations were performed using standard gamma-emitting radioactive sources. Quality assurance procedures included regular background measurements, detector stability measurements, detector stability monitoring, and maintenance of constant counting geometry.

A time of 30000 seconds for acquiring data for each sample and distilled water for determining background radiation was set. The reference sample was also analyzed. From the counting spectra, the activity concentration of ^{238}U, ^{232}Th, and ^{40}K was determined. The activity concentrations of radio nuclides in the samples were calculated using Equation (1).

$$A_C = \frac{C}{\varepsilon . m\, p_y . t} \tag{1}$$

where C is the count (area), ε is the detector efficiency, p_y is the transition probability, m is the mass in (kg), and t is the time taken in seconds (s).

2.4. Assessment of the Radiological Hazards

To evaluate the radiological effects of ceramic and porcelain sanitary ware products, the elements containing different levels of ^{226}Ra, ^{232}Th and ^{40}K, one of the determined values used was the (Ra$_{eq}$) value which is a combined sum of activities of ^{226}Ra, ^{232}Th and ^{40}K, based othe estimation that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4180 Bq/kg of ^{40}K give the same dose rate. The radium equivalent was calculated using Equation (2).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k \tag{2}$$

where A_{Ra}, A_{Th} and A_k were activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively [13].

The external radiation exposure due to ^{226}Ra, ^{232}Th, and ^{40}K (H_{ex}) was calculated by Equation (3).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{3}$$

where A_{Ra}, A_{Th} and A_k were the values of activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The values should be less than unity for safety [14]. The internal hazard index due to exposure to radon and its short-lived products is also hazardous to the respiratory organs. The internal exposure to radon and its progeny products is quantified by the internal hazard index (H_{in}), which is determined by Equation (4), and it should also be less than unity.

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{4}$$

The representative level index (I_γ) used to assess the levels of how much they were exposed to the level of gamma radiation hazards related radio nuclides was obtained by Equation (5).

$$I\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \tag{5}$$

where A_{Ra}, A_{Th} and A_k were activities of (Bq/kg) of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The absorbed dose rate (D) in nGy/in air at 1 m above the floor because of natural radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using Equation (6).

$$D = 0.427 A_{Ra} + 0.662 A_{Th} + 0.0432 A_K \tag{6}$$

where A_{Ra}, A_{Th} and A_k were activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in Bq/kg, respectively [15].

The annual effective dose rate (AEDR), which is the value of dose taken into the human body, was obtained by Equation (7) to show the health effect of the absorbed dose rates.

$$\text{AEDR}\left(\text{mSv}/y\right) = \text{D} \times \text{DCF} \times \text{IOF} \times \text{T} \tag{7}$$

where D is the dosage in air, DCF is the dose conversion factor of (0.7 Sv/Gy), IOF is the indoor occupancy factor given by (0.8), and T is the annual exposure time that's estimated at (8760 h/y) [15].

The excess lifetime cancer risk (ELCR) indicates the rate at which one can get cancer when they are exposed to a certain level, and it was calculated using Equation (8).

$$\text{ELCR} = \text{AED} \times \text{LS} \times \text{RF} \tag{8}$$

where LS is the life we live of (70 years), and RF is the risk factor (Sv-1) of 0.05 for the community as stochastic effects [16].

3. Results and Discussion

3.1. Specific Activities

The mean concentration based on production material as shown in **Table 1** for ceramics was 24.75 ± 1.2425 (Bq/Kg), 66.4 ± 3.3175 (Bq/Kg) and 444.5 ± 22.235 (Bq/Kg) for ^{238}U, ^{232}Th, and ^{40}K, respectively while for porcelain it was 14 ± 0.72 (Bq/Kg), 83.175 ± 4.16 (Bq/Kg) and 401.5 ± 20.135 (Bq/Kg) for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The world's respective average values are 50, 50, and 500 Bq/kg [1].

Table 1. Natural radioactivity concentration of sanitary wares based on production material.

Sample Code	Material	^{226}Ra (Bq/Kg)	^{232}Th (Bq/Kg)	^{40}K (Bq/Kg)
C_5	Ceramic	17 ± 0.86	81.7 ± 4.08	433 ± 21.65
C_6		22 ± 1.1	73 ± 3.65	406 ± 20.32
C_7		44 ± 2.2	73 ± 3.65	628 ± 31.42
C_8		16 ± 0.81	37.9 ± 1.89	311 ± 15.55
Mean		**24.8 ± 1.24**	**66.4 ± 3.32**	**444.5 ± 22.24**
C_9	Porcelain	17 ± 0.86	84.6 ± 4.23	275 ± 13.75
C_{10}		16 ± 0.81	64.2 ± 3.21	506 ± 25.32
C_{11}		11 ± 0.57	131.44 ± 6.57	672 ± 33.61
C_{12}		12 ± 0.63	52.5 ± 2.62	153 ± 7.66
Mean		**14 ± 0.72**	**83.18 ± 4.16**	**401.5 ± 20.14**

The Activity concentrations for the various samples are shown in Figure 1.

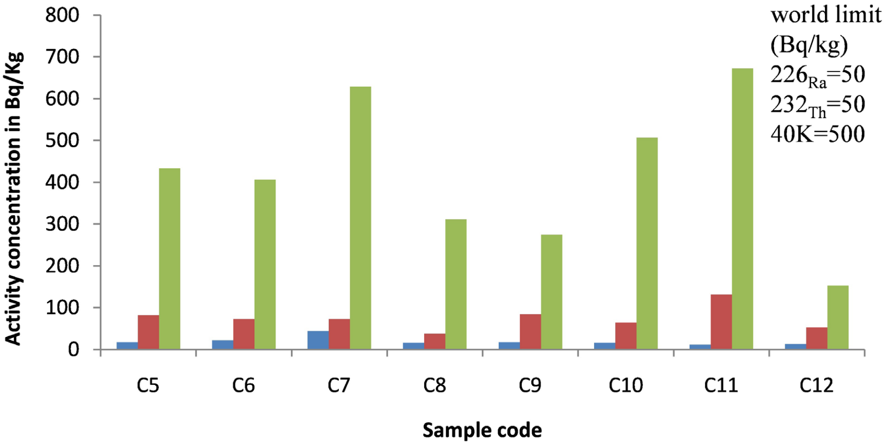

Figure 1. Activity concentration based on production material (ceramic and porcelain).

As shown in Table 2, the concentrations of ^{226}Ra and ^{40}K in both production materials are below the world average limit of 50 Bq/kg and 500 Bq/kg, while the concentration of ^{232}Th was found to be higher than the world average in both production materials, which may be attributed to the glaze zircon during the production stage.

Table 2. Average of activity concentrations and the world average.

Sample Type	Sample Number	^{226}Ra (Bq/Kg)	^{232}Th (Bq/Kg)	^{40}K (Bq/Kg)
Ceramic	(C_5 - C_8)	24.8 ± 1.24	66.4 ± 3.32	444.5 ± 22.24
Porcelain	(C_9 - C_{12})	14 ± 0.72	83.18 ± 4.16	401.5 ± 20.14
World Average		**50**	**50**	**500**

3.2. Radium Equivalent Activity (Ra_{eq})

Radium equivalent activity in all the samples is shown in Figure 2. All the samples for both production materials had lower values than the acceptable world limit of 370 Bq/kg [17]. This indicated that sanitary wares can be safely used as building and sanitation elements without creating any radiological risk to the building occupants.

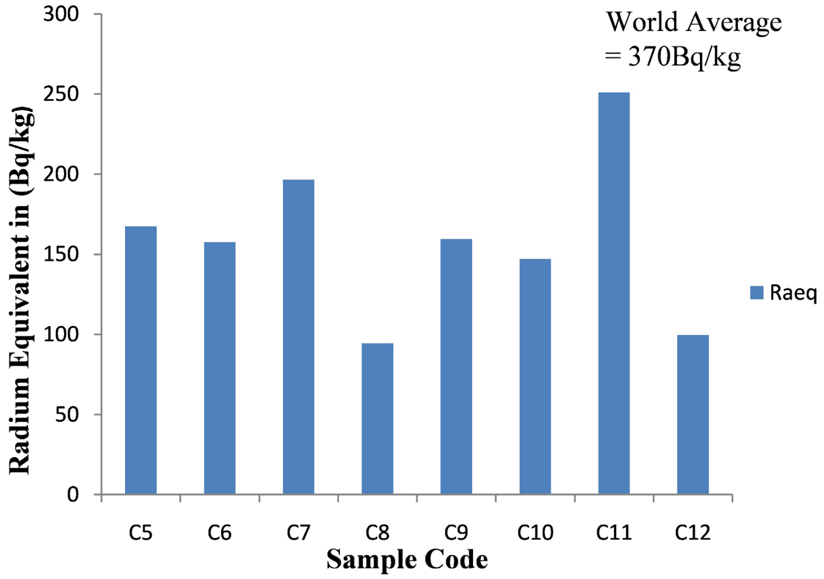

Figure 2. Radium equivalent activity for sanitary ware samples based on production material.

3.3. Hazard Indices

The Hazard indices for both Ceramic (C_5 - C_8) and porcelain (C_9 - C_{12}) are shorn in Table 3 and Figure 3.

Table 3. The radiological hazards for sanitary ware products are based on the production material.

Sample Material	ELCR × 10^{-4}	ADR (nGy/h)	I_γ	Ra_{eq} (Bq/kg)	H_{in}	H_{ex}	AED_{in} (mSv/y)	AED_{out} (mSv/y)
C_5	9.90	76 ± 3.84	1.2 ± 0.06	167 ± 8.37	0.4 ± 0.02	0.4 ± 0.02	0.2 ± 0.01	0.1 ± 0
C_6	9.309	72 ± 3.61	1.1 ± 0.05	157 ± 7.87	0.4 ± 0.02	0.4 ± 0.02	0.2 ± 0.01	0.1 ± 0
C_7	11.75	91 ± 4.56	1.4 ± 0.07	196 ± 9.83	0.5 ± 0.02	0.6 ± 0.03	0.3 ± 0.01	0.2 ± 0.01
C_8	5.66	43 ± 2.19	0.6 ± 0.03	94 ± 4.71	0.2 ± 0.01	0.2 ± 0.01	0.1 ± 0	0.1 ± 0
Mean	**9.16**	**70.5 ± 3.6**	**1.08 ± 0.05**	**153.5 ± 7.7**	**0.38 ± 0.02**	**0.4 ± 0.02**	**0.1 ± 0.01**	**0.1 ± 0.02**
C_9	9.26	71 ± 3.59	1.1 ± 0.05	159 ± 7.97	0.4 ± 0.02	0.4 ± 0.02	0.2 ± 0.01	0.1 ± 0
C_{10}	8.84	68 ± 3.43	1 ± 0.05	146 ± 7.34	0.3 ± 0.01	0.4 ± 0.02	0.2 ± 0.01	0.1 ± 0
C_{11}	14.89	115 ± 5.8	1.8 ± 0.09	251 ± 12.6	0.6 ± 0.03	0.7 ± 0.03	0.4 ± 0.02	0.2 ± 0.01
C_{12}	5.76	44 ± 2.23	0.7 ± 0.03	99 ± 4.98	0.2 ± 0.01	0.3 ± 0.01	0.1 ± 0	0.1 ± 0
Mean	**9.421**	**74.5 ± 3.8**	**1.15 ± 0.06**	**163.8 ± 8.2**	**0.38 ± 0.02**	**0.45 ± 0.02**	**0.2 ± 0.01**	**0.13 ± 0.01**

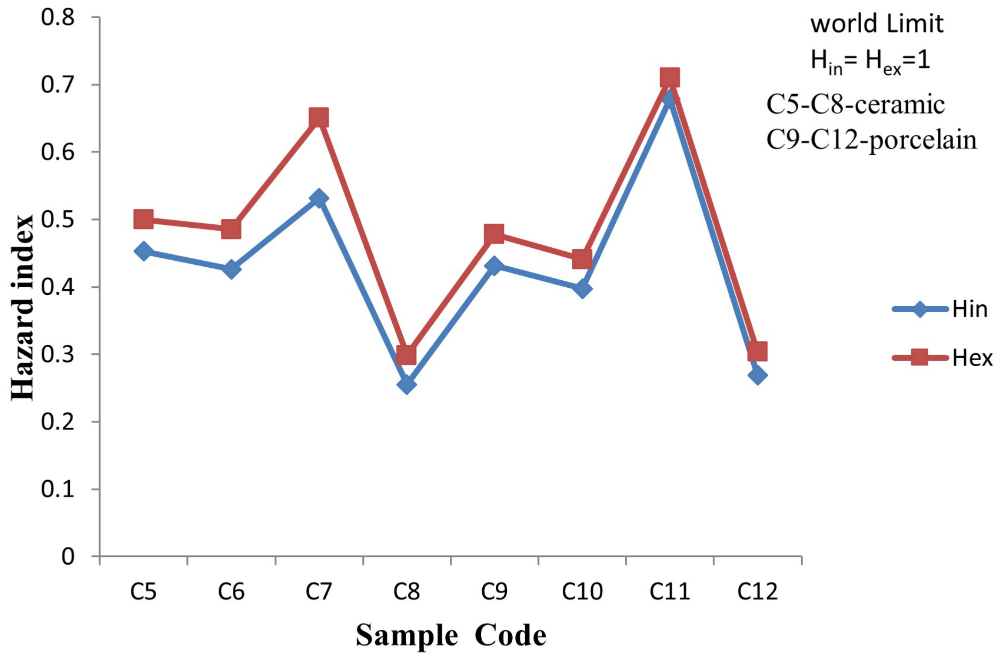

Figure 3. Comparison of external and internal hazard indices for sanitary ware products based on production materials.

As shown in **Table 4** and **Figure 3**, the internal and external hazard indices in both production materials (ceramic and porcelain) were less than the world average value of unity. This indicates that they can be safely used in construction without creating any radiological risk.

Table 4. A comparison of the mean values for each production material and the world average with acceptable limits.

Type	ELCR × 10^{-4}	ADR (nGy/h)	I_γ	Ra_{eq} (Bq/kg)	H_{in}	H_{ex}	AED_{in} (mSv/y)	AED_{out} (mSv/y)
Ceramic	9.16	70.5 ± 3.6	1.08 ± 0.05	153.5 ± 7.7	0.38 ± 0.02	0.4 ± 0.02	0.13 ± 0.01	0.1 ± 0
Porcelain	9.421	74.5 ± 3.8	1.15 ± 0.06	163.8 ± 8.2	0.375 ± 0.02	0.45 ± 0.02	0.23 ± 0.01	0.13 ± 0.01
World Average	**2.9 × 10^{-3}**	**84**	**6**	**370**	**1**	**1**	**1**	**1**

From the table above, all the radiological hazard indices were less than the world average values except for the excess lifetime cancer risk, whose mean value was slightly above the world average.

3.4. Absorbed Dose Rate (ADR)

The absorbed dose rate (ADR) is shown in **Figure 4**. The average values were lower than the average indoor gamma dose rate of (84 nG/y) [17] [18]. This implied that sanitary ware products from both production materials can be safely used for sanitation purposes without creating any radiological risk to the public exposed to the sanitary ware.

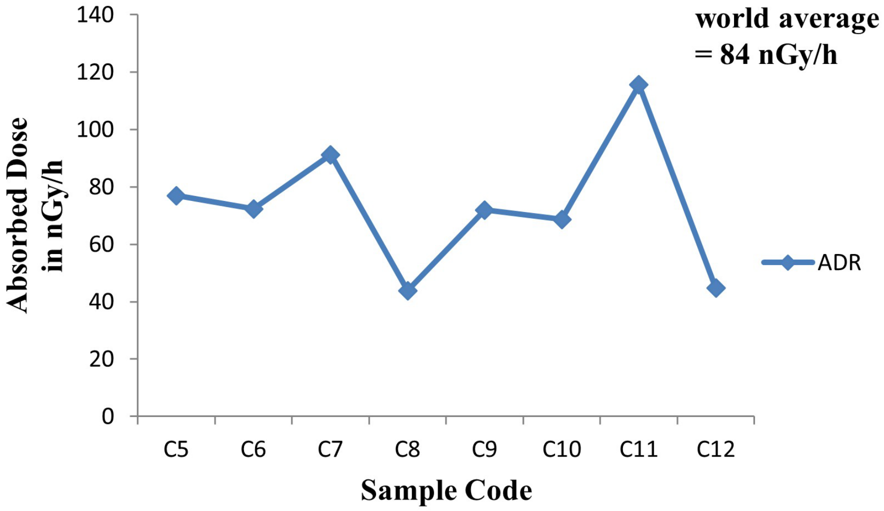

Figure 4. A graph of absorbed dose rate in (nGy/h) for sanitary ware products based on production material.

3.5. Representative Gamma Index

The values of the representative gamma index are shown in **Figure 5**. From the figure, all the values obtained for the two production materials were below the acceptable limit of 6 (4). **Figure 5** gives a representation of the gamma index for porcelain and ceramic production material.

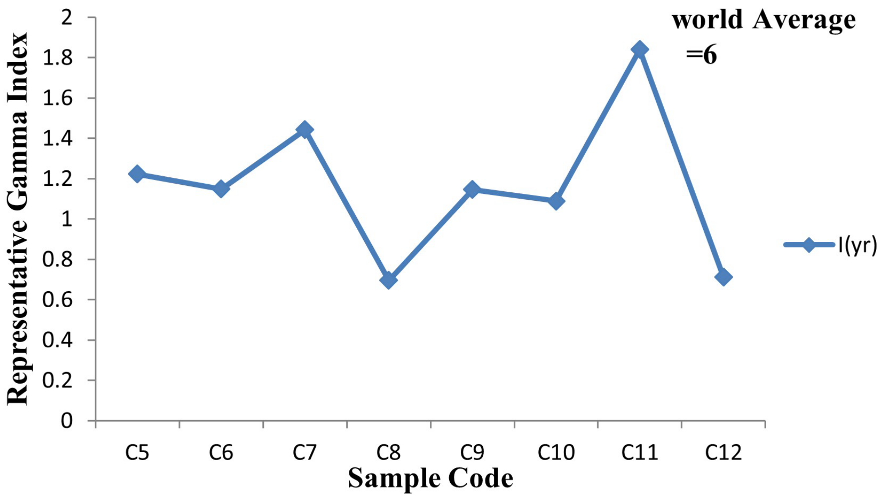

Figure 5. The representative gamma index for production material of sanitary ware products.

This implies that sanitary wares from both production materials pose no threat to human health when exposed to sanitary wares.

3.6. Annual Effective Dose Rate (AEDR)

The obtained values of annual effective dose rate are shown in **Figure 6**. Both the outdoor and indoor values were less than the world average limit of 1mSv/y limit set by the European Commission [19]. The calculated annual effective dose values showed that sanitary ware products for both production materials can be used for building and sanitation without creating any radiological risk to the exposed population.

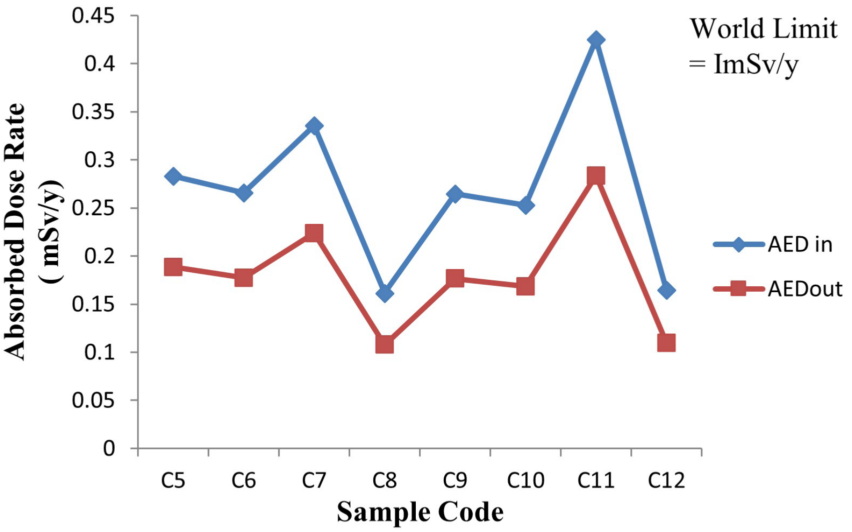

Figure 6. Annual effective dose rate for indoor and outdoor in mSv/y for production material.

3.7. Excess Lifetime Cancer Risk

The excess lifetime cancer risk values ranged from a maximum of 1.489×10^{-3} to a minimum of 5.66×10^{-4} with an average value of 9.421×10^{-4}. The values were higher than the world average value of 2.9×10^{-4} [20]. This implied sanitary ware could be safely used for hygiene purposes, but the exposure levels should be taken into consideration to minimize the exposure. This will aid in reducing chronic diseases that occur because of uncontrolled exposure to the sanitary ware products (**Figure 7**).

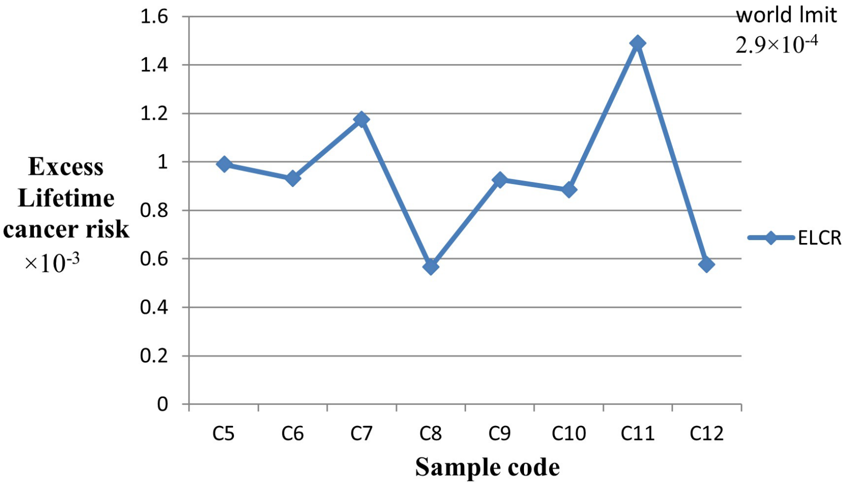

Figure 7. Excess lifetime cancer risk for sanitary ware based on production materials.

4. Conclusions and Recommendations

The natural radioactivity levels in ^{226}Ra, ^{232}Th, and ^{40}K and related radiological hazards in ceramic and porcelain sanitary wares were determined using a sodium iodide detector. A total of 8 samples were examined. Based on the results, except for ^{232}Th, all the other values (^{226}Ra and ^{40}K) show that ceramic and porcelain sanitary wares are safe to be used in building and construction. Despite the values for levels of radium and potassium being within the acceptable limits, the manufacturers of the sanitary wares should check on the ^{232}Th levels, which may be elevated due to the glazing element zircon at the manufacturing stage, to ensure they are below or within the acceptable values for the safety of the population exposed.

The elevated levels of ^{232}Th can be controlled by the manufacturers by screening raw materials for their activity concentration before production, and purchasing is done. This could save the users from high thorium-232 levels in the two materials of production. The study was limited to only two production materials (porcelain and ceramic). All the radiological hazard parameters (ADR, I_γ, Ra_{eq}, H_{in}, H_{ex}, AED_{in}, and AED_{out}) were within the acceptable limit recommended by international bodies except for ELCR$_{\text{, which}}$ had slightly higher values. The results of activity concentration and radiological hazard parameters indicated that sanitary wares are safe for use with care on production materials at the manufacturing stage to reduce ^{232}Th levels. Those involved in construction using sanitary wares can make an informed choice based on the activity concentrations.

In addition, there exists a limitation of a narrow sample base focusing only on two production materials and a lack of raw material composition data.

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Conflicts of Interest

The authors declare no conflict of interest regarding publication of this article.

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