AUTHORS:
Olusegun Y. Omogunloye
Naven Chetty
Abiola O. Ilori

AFFILIATION:
School of Chemistry and Physics,
University of KwaZulu-Natal,
Pietermaritzburg, South Africa

CORRESPONDENCE TO:
Naven Chetty

EMAIL:
chettyn3@ukzn.ac.za

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Radiological risk assessment of cement used in contemporary South African buildings

Using a calibrated NaI(Tl) and a well-shielded detector connected to a computer-resident quantum multichannel analyser, the radionuclide contents of primordial radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were evaluated in commonly used cement brands in South Africa, and the associated radiation risk parameters were calculated. The reported activity concentrations varied from 8.00 ± 2.83 to 45.00 ± 2.79 Bq/kg, 12.00 ± 0.90 to 32.00 ± 0.51 Bq/kg and 454.00 ± 0.56 to 1765.00 ± 0.93 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The absorbed gamma dose rate, annual effective dose equivalent, annual gonadal dose equivalent, excess lifetime cancer risk, gamma index and alpha index were utilised as the radiological health impact metrics to evaluate the potential radiation risks. The determined radiological health impact parameter results were below the relevant radiation safety authorities' recommended reference levels for building materials. Therefore, the use of cement products as building materials presents no significant risk in the study areas.

Significance:

The results of this study contribute to understanding the radiological risks associated with different brands of commonly used cement in South Africa and provide fundamental data for the activity concentrations of primordial radionuclides ^{226}Ra, ^{232}Th and ^{40}K. These studies have been instructive from a building materials radiation safety viewpoint and reveal that the assessed risk parameters fall within the recommended safety limits. This is significant from the perspective of environmental health and radiological protection concerning the safe use of the studied cement. The methodology and findings can also inform similar studies in other regions, enhancing global awareness of material safety standards.

Introduction

It is indeed of great significance to have a better knowledge of the risk associated with the radiation emitted from dwellings due to the various building materials that contain radionuclides of different types, which constitute radiation exposure to the resident.[1] The radiation exposure takes place daily, and the capability of radionuclides to quickly find their way into the air makes them be transferred into human environments.[2,3] Most humans spend approximately 80% of their lifetime indoors, so assessing the radionuclide contents in cement used as building materials and related radiological health hazards for humans is essential.[4] Naturally occurring radionuclides of primordial origin in types of cement used as building materials are responsible for irradiation in dwellings.[5] The radiations come from potassium-40 and gamma radiation from the uranium and thorium families.[6] Gamma radiation exposure causes external exposure when it is directly absorbed.[7] Internal exposure, however, is brought on by radium-226 and thorium-232, as well as their daughter nuclides, including radon-222 and thoron-220, and their progenies.[8] Varying degrees of radiation exposure in man have been reported to lead to deterministic and stochastic effects, including cancer and genetic defects such as chromosome aberrations and mutation.[9,10]

The South African population has increased annually by about 2% since 1980.[11] In the same vein, South Africa's sales of types of cement have progressively increased from 7 million tonnes in 1980 to 11 million tonnes in 2010.[12] There is a clear indication of a corresponding increase in demand for housing as a basic human need due to population increase. Therefore, to cater to the ever-increasing demand for housing by the populace, increased demands for construction materials are inevitable. In both rural and urban areas of South Africa, cement is one of the most essential building materials used to construct homes and other structures.[13] It is used in concrete and block production, flooring and covering of the building floors and walls.[13] Thus, cement has played and will keep on playing significant roles in meeting South Africa's developmental agenda because buildings with cement as an essential component are virtually everywhere.[14] Buildings must contain cement because of its many beneficial properties, such as its 'bond'-like function, its role in filling the spaces between fine and coarse aggregates and its hydration reaction properties that allow buildings to gain strength continuously[15], and until now, no suitable materials with better or similar qualities have been discovered as an alternative to cement in buildings.

The research on primordial radionuclide concentrations in cement used as building materials in numerous countries throughout the world has garnered much attention over the years.[16-18] The determination of radioactivity concentrations in cement is essential to assess the possible radiological health hazards to residents and to develop radiation protection strategies and reference levels for optimising the protection of the public when using and managing cement as a building material, as required by the International Atomic Energy Agency's (IAEA) General Safety Requirements (GSR) Part 3.[19] The study emphasises the importance of investigating radiation exposure from building materials containing radionuclides to understand and mitigate associated risks, aligning with the IAEA GSR Part 3 frameworks and the International Commission on Radiological Protection (ICRP) Publication 103.[19,20] Therefore, we sought to evaluate the radiological health and safety risks associated with radiation exposure due to the radioactivity concentrations of the primordial radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in cement commonly used for buildings in South Africa.

Materials and methods

Collection and preparation of samples

Samples of cement were obtained from multiple suppliers of building materials in Pietermaritzburg, South Africa. The sampling was purposive, focusing on commonly used brands in the area that represent a significant portion of the market. Specifically, cement samples were collected from suppliers stocking products from the Pretoria Portland Cement Company (PPC), Natal Portland Cement Company (NPC) and Dangote Cement South Africa (Sephaku). These brands were selected based on their prevalence and representation in the local construction industry. The sample size was determined based on practical considerations and standard practices in environmental and materials science research.[2,18] The collected samples were air-dried and sieved with a 2-mm mesh for homogeneity. An amount of 200 g of each sample was placed in Marinelli bottles with the same shape as the reference material for gamma spectrometric analysis. The bottles were well-labelled and sealed tightly with tape to prevent radon escape. There were a total of seven samples divided among three popular cement brands.

Gamma spectrometric analysis

The radioactivity concentrations of the naturally occurring radionuclides in the studied samples were measured with a thallium-doped sodium iodide (NaI(Tl)) gamma-ray spectrometric system at the Radiation Physics Research Laboratory of the University of Medical Sciences, Ondo, Ondo State, Nigeria. A multichannel computer-resident quantum analyser (MCA2100R) and a well-shielded detector were attached to the system. Spectral analysis was done using Palmtop MCA computer gamma analysis software (Model: MCA8k-01, Serial: 0202).

The reference standard source for the detector efficiency calibration was the Analytical Quality Control Service (AQCS, USA), which validated the activities of the radionuclides of interest.

The efficiency calibration was conducted by acquiring a calibration standard spectrum until the total absorption peak count rate was determined with a statistical uncertainty of less than 1% at a 95% confidence level. For the calculation of photo peaks, the net count rate was established to evaluate the output for all the energies used during the measurement. The output was then correlated with the count rate and the standard source using Equation 1[21]:

$$E_\gamma = \frac{N_E}{S_c \times \gamma_E \times t_c} \qquad \text{Equation 1}$$

where N_E is the full energy peak net count, E_γ is the energy probability of gamma photons, γ_E is the probability of gamma emission, S_c is the activity of the standard source, while the counting time is t_c.

Gamma rays emitted from the standard reference source were measured, and the resulting spectrum was used to create the efficiency curve. Subsequently, power fitting was applied to optimise the R^2-value, as depicted in Figure 1.

The samples being counted had the same geometry as the standard references. The gamma transition energies of 1764.5, 2614 and 1640.8 keV were used to estimate the sample's radioactivity levels for ^{226}Ra, ^{232}Th and ^{40}K. Each sample was counted for 36 000 s (10 h). The counting time of 10 h (36 000 s) is selected to achieve high statistical accuracy by accumulating a large number of photon counts. This extended duration helps minimise statistical uncertainties in the measured gamma spectra, ensuring precise determination of radionuclide concentrations in the cement samples.[22]

Equation 2 was used to calculate the radioactivity concentration of the radionuclide from a measurement of the detector's efficiency[23-25]:

$$C_{sp} = \frac{N_{sam}}{P_E \cdot \varepsilon \cdot T_c \cdot M} \qquad \text{Equation 2}$$

where ε is the detection system's overall counting efficiency, C_{sp} is the activity concentration of the radionuclides of interest in Bq/kg, N_{sam} is the radionuclide's net count in the sample, P_E is the probability of gamma ray emission (gamma yield), M is the sample's mass (in kg) and T_c is the sample counting time. The Data Analytical tool in Microsoft Excel 2010 running on Windows 10 was used for statistical analysis. To determine the minimum detection limit (MDL) for radionuclides ^{226}Ra, ^{232}Th and ^{40}K using gamma spectrometry, the background counts and the system's detection efficiency were evaluated. This calculation yields the smallest detectable radioactivity level above the background noise. The gamma spectrometric system's MDLs for ^{226}Ra, ^{232}Th and ^{40}K were 0.69, 0.78 and 2.35 Bq/kg, respectively. The MDL values provided signify the system's sensitivity to detect these radionuclides at extremely low levels, reflecting its capability under defined measurement conditions.

Radiological health hazard indices assessment

In cement samples from the study area, activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K have been studied for potential radiological risks that could impact human health due to radiation exposure. The radiological health impact metrics examined are the absorbed gamma dose rate, annual effective dose equivalent, annual gonadal dose equivalent (AGDE), excess lifetime cancer risk (ELCR), gamma index and alpha index.

Absorbed gamma dose rate

The following equations were applied to the measured activity concentrations to calculate the indoor and outdoor absorbed gamma dose rates (D_{in} and D_{out}) produced by gamma radiation caused by ^{226}Ra, ^{232}Th and ^{40}K at a height of 1 m above the ground[26]:

$$D_{in}(\text{nGyh}^{-1}) = 0.92\,C_{Ra\text{-}226} + 1.1\,C_{Th\text{-}232} + 0.081\,C_{K\text{-}40} \qquad \text{Equation 2a}$$

$$D_{out}(\text{nGyh}^{-1}) = 0.462\,C_{Ra\text{-}226} + 0.0604\,C_{Th\text{-}232} + 0.0417\,C_{K\text{-}40} \qquad \text{Equation 2b}$$

where the conversion factors for the doses associated with the radioactive concentrations of ^{226}Ra, ^{232}Th and ^{40}K for materials used as

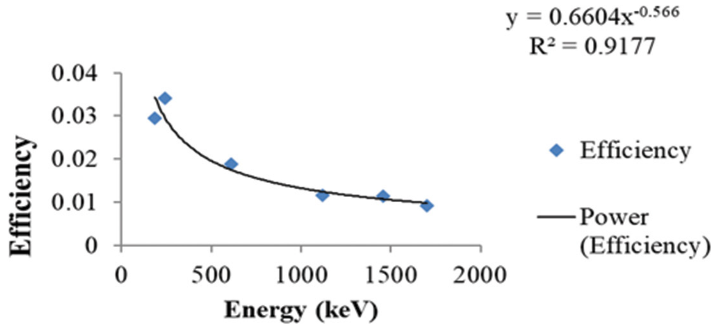

Figure 1: Efficiency calibration curve showing the detection efficiency of the detector.

building materials are 0.92, 1.1, 0.081, 0.462, 0.0604 and 0.0417 in $nGyh^{-1}/Bq/kg$ for indoor and outdoor, respectively.

Annual effective dose

The annual effective dose (E) has been calculated using Equation 3a–3c[26,27]:

$$E_{in}(\mu Svy^{-1}) = 4.91 \times D_{in}(nGyh^{-1}) \qquad \text{Equation 3a}$$

$$E_{out}(\mu Svy^{-1}) = 1.23 \times D_{out}(nGyh^{-1}) \qquad \text{Equation 3b}$$

$$E_{tot}(\mu Svy^{-1}) = E_{in} + E_{out} \qquad \text{Equation 3c}$$

The annual effective doses for indoor, outdoor and total exposure are E_{in}, E_{out} and E_{tot}, respectively. The annual effective dose (E) was estimated using the following factors: the number of hours in a year (8610), the percentage of time spent indoors and outdoors (0.8 and 0.2) and the dose conversion factor of 0.7 $Sv.Gy^{-1}$,[26] from the air-absorbed dose rate to an effective dose.

Annual gonadal dose equivalent

The exceptionally high radiosensitivity of the human gonads, bone marrow and bone surface cells makes them organs of interest.[28] To calculate the AGDE, Equation 4 was utilised[28]:

$$AGDE\ (\mu Svy^{-1}) = 3.09C_{Ra\text{-}226} + 4.18C_{Th\text{-}232} + 0.0314C_{K\text{-}40} \qquad \text{Equation 4}$$

where $C_{Ra\text{-}226}$, $C_{Th\text{-}232}$ and $C_{K\text{-}40}$ are ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in Bq/kg, respectively. Values 3.09, 4.18 and 0.0314 are the conversion factors for the doses associated with the radioactive concentrations of ^{226}Ra, ^{232}Th and ^{40}K in materials used as building materials, respectively.

Gamma index and alpha index

To determine if the cement had complied with the radiological safety criteria for construction materials, the gamma index (I_γ) was calculated using the following equation[29]:

$$I_\gamma = {}^{C_{226}Ra}\!/_{300} + {}^{C_{232}Th}\!/_{200} + {}^{C_{40}K}\!/_{3000} \qquad \text{Equation 5a}$$

The alpha index (I_α), which symbolises the surplus alpha radiation brought on by breathing in radon-222 from the cement, was calculated using Equation 5b.

$$I_\alpha = {}^{C_{226}Ra}\!/_{200} \qquad \text{Equation 5b}$$

Table 1: The reported ^{226}Ra, ^{232}Th and ^{40}K levels in the cement samples

Sample ID	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
NPC 1	22.00 ± 2.51	20.00 ± 3.59	1662.00 ± 1.15
NPC 2	15.00 ± 2.64	32.00 ± 0.51	569.00 ± 0.72
NPC 3	8.00 ± 2.83	12.00 ± 0.90	1765.00 ± 0.93
Mean	15.00 ± 2.66	21.33 ± 1.66	1332.00 ± 0.94
Alpine 1	54.00 ± 4.80	29.00 ± 0.42	454.00 ± 0.56
Alpine 2	16.00 ± 4.70	25.00 ± 0.24	1285.00 ± 1.73
Alpine 3	35.00 ± 2.41	17.00 ± 0.48	1195.00 ± 1.33
Mean	35.00 ± 3.97	23.67 ± 0.38	978.00 ± 1.21
Dangote	45.00 ± 2.79	15.00 ± 1.28	1249.00 ± 0.63

where $C_{Ra\text{-}226}$, $C_{Th\text{-}232}$ and $C_{K\text{-}40}$ are ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in Bq/kg, respectively. 300, 200 and 3000 are the dose conversion factors associated with the radioactive concentrations of ^{226}Ra, ^{232}Th and ^{40}K for materials used as building materials for gamma index, while 200 is the dose conversion factor associated with the radioactive concentrations of ^{226}Ra in building materials for alpha index.

Excess lifetime cancer risk

The annual effective dose (E) values computed as specified in Equation 6a–6c were used to calculate the ELCRs[26,30]:

$$ELCR_{in} = E_{in} \times D_l \times R_f \qquad \text{Equation 6a}$$

$$ELCR_{out} = E_{out} \times D_l \times R_f \qquad \text{Equation 6b}$$

$$ELCR_{tot} = ELCR_{in} + ELCR_{out} \qquad \text{Equation 6c}$$

R_f and D_l are the fatal cancer risk factors for stochastic effect (estimated 0.05 Sv^{-1} for the general public) and lifetime duration (65.10 years for South Africa), respectively.

Results and discussion

Natural radioactivity concentrations

As shown in Table 1, the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in cement samples are unevenly distributed.

The average activity concentration of ^{226}Ra in the cement samples was 27.857 Bq/kg, ranging from 8.00 ± 2.83 to 45.00 ± 2.79 Bq/kg. Dangote Cement (Sephaku) had the highest value of ^{226}Ra, while NPC 3 had the lowest value. With an average value of 21.43 Bq/kg, the measured activity concentrations of ^{232}Th in the cement samples ranged from 12.00 ± 0.90 to 32.00 ± 0.51 Bq/kg. NPC 2 and NPC 3 contained the highest and lowest results for ^{232}Th, respectively. With an average value of 1168.43 Bq/kg, the recorded activity concentrations of ^{40}K in the cement samples ranged from 454.00 ± 0.56 to 1765.00 ± 0.93 Bq/kg. Pretoria Portland cement (Alpine 1) had the lowest value, whereas NPC 3 had the highest value for ^{40}K, respectively. The world average values of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building materials are 50, 50 and 500 Bq/kg, respectively.[28] Alpine 1 had ^{226}Ra activity concentration value of 54.00 ± 4.80 Bq/kg, which is slightly higher than the world average value. The measured values and averages of the activity concentrations for ^{226}Ra and ^{232}Th in virtually all of the examined cement samples were found to be lower than the world average values. The average activity concentrations of ^{40}K in all the examined cement samples were found to be higher than the world average values, except for Alpine 1, which had an activity concentration value of 454.00 ± 0.56 Bq/kg, slightly lower than the world average. In general, the mean activity concentrations of ^{40}K were the highest in all the cement samples compared to the other two naturally occurring radionuclides (^{226}Ra and ^{232}Th), respectively. This is typical and expected from any geologically derived materials because potash feldspar minerals are relatively enriched in the natural environment.[31] The concentrations of ^{226}Ra, ^{232}Th and ^{40}K in cement samples from the study locations are depicted in Figure 2.

The results of earlier research from various parts of the world were also compared with the calculated average values of the activity concentrations of naturally occurring radionuclides in the studied cement samples. Table 2 displays the comparison.

Radiological hazard indices

Table 3 displays the findings of the assessed radiological health hazard parameters. The table shows that the assessed indoor and outdoor absorbed gamma dose rates (D_{in} and D_{out}) varied from 95.089 to 176.862 $nGyh^{-1}$ and 49.985 to 91.549 $nGyh^{-1}$. All of the cement samples' indoor absorbed gamma dose rates were above the population-weighted average of 84 $nGyh^{-1}$.[26] The annual effective dose for indoor, outdoor

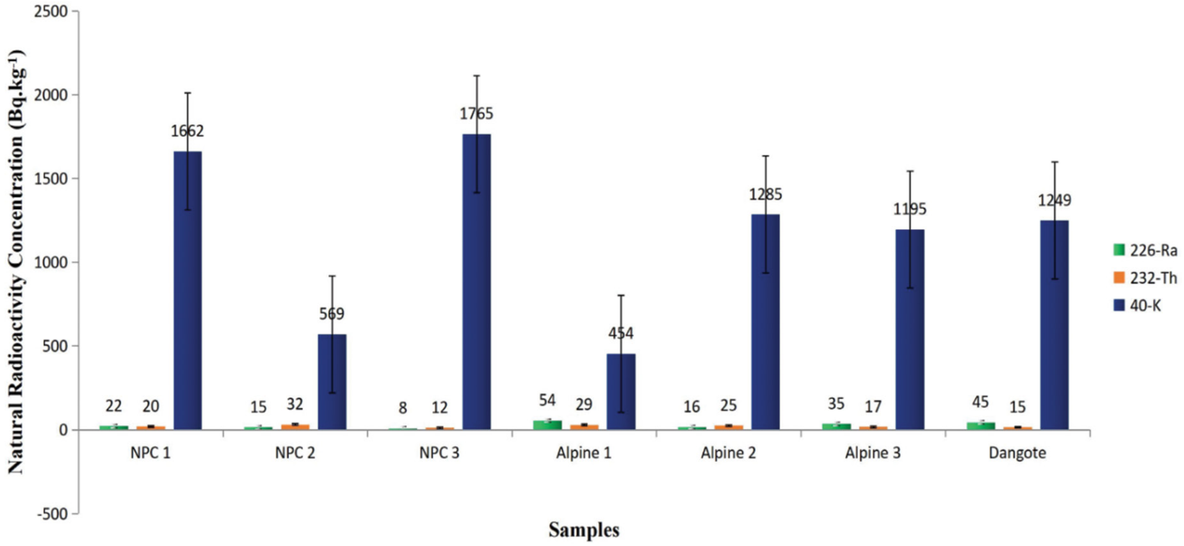

Figure 2: Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K activity in the cement samples under study.

Table 2: Comparing the average concentrations of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in cement samples to those discovered in other countries

Sample ID	Country	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Reference
NPC	South Africa	15.00 ± 2.66	21.33 ± 1.66	1332.00 ± 0.94	Present study
Alpine	South Africa	35.00 ± 3.97	23.67 ± 0.38	978.00 ± 1.21	Present study
Dangote	South Africa	45.00 ± 2.79	15.00 ± 1.28	1249.00 ± 0.63	Present study
Cement sample	Albania	179.70 ± 8.90	55.00 ± 5.80	17.00 ± 3.30	32
Cement sample	Algeria	41.00 ± 7.00	27.00 ± 3.00	422.00 ± 3.00	33
Cement sample	Bangladesh	61.00	65.00	952.00	34
Cement sample	Cameroon	27.00 ± 4.00	15.00 ± 1.00	277.00 ± 117.00	35
Cement sample	China	118.70 ± 14.20	36.10 ± 17.80	444.50 ± 163.10	36
Cement sample	China	59.00	39.00	181.00	37
Cement sample	Egypt	36.00 ± 4.00	43.00 ± 2.00	82.00 ± 4.00	38
Cement sample	Egypt	134.00	88.00	416.00	39
Cement sample	Ghana	35.94 ± 0.78	25.44 ± 0.80	233.00 ± 3.95	40
Cement sample	India	26.00	29.00	260.00	41
Cement sample	Iraq	24.25 ± 1.45	25.41 ± 1.65	93.17 ± 7.30	42
Cement sample	Laos	41.12 ± 2.44	16.60 ± 2.37	141.48 ± 4.50	43
Cement sample	Malaysia	29.00 ± 7.00	31.00 ± 9.00	205.00 ± 71.00	44
Cement sample	Morocco	31.00 ± 5.00	19.00 ± 3.00	238.00 ± 13.00	45
Cement sample	Nigeria	20.00	8.00	51	46
Cement sample	Pakistan	25.00 ± 10.00	37.00 ± 9.00	245.00 ± 95.00	47
Cement sample	Serbia	37.00	15.00	43.00	48
Cement sample	Senegal	112.69 ± 26.02	13.12 ± 1.88	73.35 ± 18.12	49
Cement sample	Turkey	34.00 ± 4.00	15.00 ± 2.00	220.00 ± 13.00	50
Cement sample	Turkey	26.00	10.00	130.00	51
Building materials	World average	50	50	500	28

Table 3: Calculated radiological health hazard indices

Sample ID	D_{in} ($nGyh^{-1}$)	D_{out} ($nGyh^{-1}$)	E_{in} (μSvy^{-1})	E_{out} (μSvy^{-1})	E_{tot} (μSvy^{-1})	AGDE (μSvy^{-1})	$I\gamma$	$I\alpha$	$ELCR_{in} \times 10^{-3}$	$ELCR_{out} \times 10^{-3}$	$ELCR_{tot} \times 10^{-3}$
NPC 1	176.862	91.549	868.392	112.606	980.998	203.767	0.727	0.110	2.827	0.367	3.193
NPC 2	95.089	49.985	466.887	61.482	528.369	197.977	0.400	0.075	1.520	0.200	1.720
NPC 3	163.525	84.545	802.908	103.990	906.898	130.301	0.675	0.040	2.613	0.338	2.952
Mean	145.159	75.360	712.729	92.692	805.422	177.348	0.601	0.075	2.320	0.302	2.622
Alpine 1	118.354	61.396	581.118	75.517	656.635	302.336	0.476	0.270	1.892	0.246	2.137
Alpine 2	146.305	76.077	718.358	93.574	811.932	194.289	0.607	0.080	2.338	0.305	2.643
Alpine 3	147.695	76.270	725.183	93.811	818.994	216.733	0.600	0.175	2.360	0.305	2.666
Mean	137.451	71.247	674.886	87.634	762.520	237.786	0.561	0.175	2.197	0.285	2.482
Dangote	159.069	81.933	781.029	100.778	881.807	240.969	0.641	0.225	2.542	0.328	2.870

and total exposure ranged from 466.887 to 868.392 μSvy^{-1}, 61.482 to 112.606 μSvy^{-1} and 528.369 to 980.998 μSvy^{-1}, respectively. In NPC 1 and NPC 2, the highest and lowest values of the indoor and outdoor absorbed gamma dose rate and the indoor, outdoor and total annual effective dose were recorded. All of the samples' annual effective dose values were below the reference level of 1000 μSvy^{-1}.[26] The AGDE ranged from 130.301 to 302.336 μSvy^{-1}. While NPC 3 had the lowest value, PPC (Alpine 1) had the highest value of AGDE. Except for Alpine 1, whose yearly gonadal dose equivalent value was slightly higher at 302.336 μSvy^{-1}. All of the samples' recorded values were below the 300 μSvy^{-1} world average. The alpha index (I_α) ranged from 0.040 to 0.270, and the gamma index (I_γ) ranged from 0.040 to 0.727.

The gamma (I_γ) and alpha (I_α) index values fell below the recommended upper limit of unity.[52] For indoor ($ELCR_{in}$), outdoor ($ELCR_{out}$) and total ($ELCR_{tot}$) ELCR, respectively, the ELCR values ranged from 1.520×10^{-3} to 2.827×10^{-3}, 0.200×10^{-3} to 0.367×10^{-3} and 1.720×10^{-3} to 3.193×10^{-3}. The $ELCR_{in}$ and $ELCR_{tot}$ reported in this study are higher than the world average values of 0.29×10^{-3} and 1.45×10^{-3} reported by Mohammed and Ahmed.[53] The values of ELCR equivalent to 1000, 100, 10 and 1 μSvy^{-1} will increase the chance of developing fatal cancer by 4%, 0.4%, 0.04% and 0.004%, respectively.[44,54] The values of the risk obtained for this study are within the acceptable risk limits.

Conclusion

According to the study, there were uneven distributions of the measured natural radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the cement samples. In almost all of the analysed cement samples, the measured values and averages of the activity concentrations for ^{226}Ra and ^{232}Th were lower than the global average. In contrast, in almost all of the analysed cement samples, the observed values and average activity concentrations for ^{40}K were higher than the global average levels provided by the UNSCEAR 2000 Report. The findings show that ^{40}K is the radionuclide in the environment with the highest measured radioactivity content. Radiological health impact measures, including absorbed gamma dose rate, annual effective dose, AGDE, ELCR, gamma index, and alpha index, were established to evaluate the potential radiation risks. The cement samples' indoor absorbed gamma radiation rates were higher than the population-weighted global average of 84 $nGyh^{-1}$ provided by the UNSCEAR in 2000. The annual effective dose and annual gonadal dose equivalent values for all the samples were lower than the reference level of 1000 μSvy^{-1} and the world average value of 300 μSvy^{-1}, respectively. The gamma index (I_γ) and alpha index (I_α) values were all below the reference level of unity. Even if all of the determined ELCRs are higher than the global average value, there is very little chance that this will increase cancer risk in the long run. However, ^{40}K naturally occurring radioactivity content was higher than the global average, which could serve as a warning to the radiation safety authority.

Data availability

The data supporting the results of this study are available upon request to the corresponding author.

Declarations

This study was carried out as part of O.Y.O.'s PhD research under the supervision of N.C. and co-supervision of A.O.I. at the University of KwaZulu-Natal, South Africa. We have no competing interests to declare. We have no AI or LLM use to declare.

Authors' contributions

O.Y.O.: Collected the samples and prepared them for gamma ray spectrometry. N.C.: Conceived the study and supervised and guided the process of the research. A.O.I.: Co-supervised the research and participated in the spectrometric analysis. All authors read and approved the final manuscript.

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