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# Peer review history for:

Omogunloye OY, Chetty N, Ilori AO. Radiological risk assessment of cement used in contemporary South African buildings. S Afr J Sci. 2025;121(3/4), Art. #16778. <u>https://doi.org/10.17159/sajs.2025/16778</u>

#### HOW TO CITE:

Radiological risk assessment of cement used in contemporary South African buildings [peer review history]. S Afr J Sci. 2025;121(3/4), Art. #16778. <u>https://doi.org/10.17159/sajs.2025/16778/peerreview</u>

Reviewer 1: Round 1 Date completed: 13 December 2024 Recommendation: Accept / Revisions required / Resubmit for review / Resubmit elsewhere / Decline / See comments Conflicts of interest: None

Does the manuscript fall within the scope of SAJS?

Yes/No

pecialists
Page 1 of 10

# Is appropriate and adequate reference made to other work in the field?

# Yes/No

Is it stated that ethical approval was granted by an institutional ethics committee for studies involving human subjects and non-human vertebrates?

# Yes/No/Not applicable

If accepted, would you recommend that the article receives priority publication?

# Yes/No

Are you willing to review a revision of this manuscript?

# Yes/No

With regard to our policy on '<u>Publishing peer review reports</u>', do you give us permission to publish your anonymised peer review report alongside the authors' response, as a supplementary file to the published article? Publication is voluntary and only with permission from both yourself and the author.

# Yes/No

# Comments to the Author:

The manuscript was well written. Effect the minor corrections highlighted on the manuscript.

# [See Appendix 1 for Reviewer 1's comments made directly on the manuscript]

# Author response to Reviewer 1: Round 1

# Introduction

- 1. Line 29 "non-radioactive was deleted"
- 2. Line 31 "reference added"
- 3. Line 43 "reference added"
- 4. Line 45 "reference added"

# **Collection and Preparation of Samples**

5. Line 74 "permeability was changed to escape"

# **Results and discussion**

- Lines 162 & 163 was rephrased as "Alpine 1 had 226Ra activity concentration value of 54.00 ± 4.80 Bq.kg-1, which is slightly higher than the World average value.
- 7. Lines 166 169 was rephrased as "The average activity concentrations of 40K 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-1, slightly lower than the world average."
- 8. Lines 196 "UNSCEAR values of 226Ra, 232Th, 40K has been added to Table 2"

# Reviewer 2: Round 1

# Date completed: 25 June 2024

**Recommendation:** Accept / Revisions required / **Resubmit for review** / Resubmit elsewhere / Decline / See comments

Conflicts of interest: None

Does the manuscript fall within the scope of SAJS?
Yes/No
Is the manuscript written in a style suitable for a non-specialist and is it of wider interest than to specialists
alone?
Yes/No
Does the manuscript contain sufficient novel and significant information to justify publication?
Yes/No
Do the Title and Abstract clearly and accurately reflect the content of the manuscript?
Yes/No

Is the research problem significant and concisely stated?

Yes/No

Are the methods described comprehensively?

Yes/No

Is the statistical treatment appropriate?

Yes/No/Not applicable/Not qualified to judge

# Are the interpretations and conclusions justified by the research results?

Yes/Partly/No

Please rate the manuscript on overall contribution to the field

Excellent/Good/Average/Below average/Poor

Please rate the manuscript on language, grammar and tone

Excellent/Good/Average/Below average/Poor

Is the manuscript succinct and free of repetition and redundancies?

Yes/No

Are the results and discussion confined to relevance to the objective(s)?

Yes/**No** 

The number of tables in the manuscript is

Too few/Adequate/Too many/Not applicable

The number of figures in the manuscript is

Too few/Adequate/Too many/Not applicable

Is the supplementary material relevant and separated appropriately from the main document?

Yes/No/Not applicable

Please rate the manuscript on overall quality

Excellent/Good/Average/Below average/Poor

Is appropriate and adequate reference made to other work in the field?

Yes/No

Is it stated that ethical approval was granted by an institutional ethics committee for studies involving human subjects and non-human vertebrates?

Yes/No/Not applicable

If accepted, would you recommend that the article receives priority publication?

Yes/No

Are you willing to review a revision of this manuscript?

Yes/No

With regard to our policy on '<u>Publishing peer review reports</u>', do you give us permission to publish your anonymised peer review report alongside the authors' response, as a supplementary file to the published article? Publication is voluntary and only with permission from both yourself and the author.

Yes/No

# Comments to the Author:

The conceptual frame work for the research work is obsolete. The work is in the domain of existing exposure as recommended by ICRP publication 103.

This work falls in the Exposure due to commodities that incorporate radionuclides deriving from residual radioactive material: food, feed, drinking water, construction materials.

The applicable radiation protection principles are:

- Establish a Radiation Protection Strategy (Justification and Optimisation)
- Use of reference levels for the optimisation of protection

A reference level is the level of dose, activity concentration, or risk above which it is not appropriate to plan not to allow exposures to occur and below which optimization of protection and safety would be continue to be implemented.

Recommended range of applicable reference levels are 1-20 mSv depending upon the type of the existing

# exposure situation.

Low risk is within the range 1 in 10,000 to 1 in 1,000 Moderate risk is within the range 1 in 1,000 to 1 in 500 High risk 1 in 500 to 1 in 100

Authors should bring this work withing the context of IAEA BSS on Existing Exposure Situation and applicable National Regulations.

Is the risk level assessed low, moderate, acceptable or high? It there the need to establish a National Radiation Protection strategy to reduce risk by adopting dose reduction strategies?

The life expectancy for South Africa must be used for the risk estimation. The value is certainly not 70 years. The South African value must be used for the calculations.

# Reviewer's Report

# **General Comments**

The conceptual frame work for the research work is obsolete. The work is in the domain existing exposure as recommended by ICRP publication 103.

An *existing exposure situation* is a situation of exposure which already exists when a decision on the need for control needs to be taken. Existing exposure situations apply to:

1. Exposure due to contamination of areas by residual radioactive material from:

- Past activities never subject to regulatory control or where the control was not in accordance with the IAEA BSS
- A past nuclear or radiological emergency (the emergency declared ended)

2. *Exposure due to commodities that incorporate radionuclides deriving from residual radioactive material:* food, feed, drinking water, construction materials.

3. Exposure due to natural sources

- Radon in workplaces and dwellings and other buildings with high occupancy factors
- Radionuclides in commodities: food, feed, drinking water, agricultural products, construction materials, and in the environment
- Other materials with low activity concentrations of certain radionuclides
- Exposure to aircrew and space crew to cosmic radiation

The applicable radiation protection principles are:

- Establish a Radiation Protection Strategy (Justification and Optimisation)
- Use of reference levels for the optimisation of protection

A reference level is the level of dose, activity concentration, or risk above which it is not appropriate to plan not to allow exposures to occur and below which optimisation of protection and safety would be continue to be implemented.

Recommended range of applicable reference levels are 1-20 mSv depending upon the type of the existing exposure situation.

Low risk is within the range 1 in 10,000 to 1 in 1,000 Moderate risk is within the range 1 in 1,000 to 1 in 500 2 High risk 1 in 500 to 1 in 100

# IAEA BSS Requirement for the Government

*Requirement 47: Responsibilities of the government specific to existing exposure situations* The government shall ensure that existing exposure situations that have been identified are evaluated to determine which occupational exposures and public exposures are of concern from the point of view of radiation protection.

The government shall ensure that, when an existing exposure situation is identified, responsibilities for protection and safety are assigned and appropriate reference levels are established.

The regulatory body or other relevant authority assigned to establish a protection strategy for an existing exposure situation shall ensure that it defines: (a) The objectives to be achieved by means of the protection strategy; (b) Appropriate reference levels.

# Specific Comments

# Abstract

Line 14 permissible limits is not applicable to existing exposure situations. "Recommended and Permissible limits" must be change to "recommended reference levels".

The abstract must contain some conclusions on risk assessment conducted whether, low, moderate, acceptable or high.

Line 15 Recommended revision of the sentence. "Therefore, the use of the cements products as building materials presents no significant risk"

# Introduction

Line 22 "that the residents irradiate "be revised as "which constitute radiation exposure to the resident". The radiation exposure takes place daily ".....

Page 2 line 29 "non- radioactive" must be deleted.

Page 2 line 34 the obsolete term non-deterministic must be replaced by "stochastic"

Page 3 lines 54 -62 The recommended re-wording the sentences are as follows:

"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 the optimisation of protection of the public in using and managing cement as building materials as required by IAEA GSR Part 3. Therefore, this study seeks to evaluate the associated radiological health and safety risks associated with radiation exposure to due radioactivity concentrations of the primordial radionuclides (226Ra, 232Th, and 40K) in cement commonly used for buildings in South Africa." 3

A paragraph should be developed to link this work with the conceptual framework of existing exposure situation in line with ICRP Publication 103,2007 and IAEA GSR Part 3,2014.

# 2.Materials and Methods

Sampling and Sample size determination

Authors must provide the sampling method used and how they arrived at the sample size used for research work. How representative was the sampling size for population at risk. The statistical power of the sampling is weak. Risk assessment is a population-related concept.

# 2.1Collection and Preparation of Samples

Page 4 line 4 Justify how the sample size is representative for the population at risk. What is the population of the end—users of the cement products. The records of the three companies can give you some information on this data.

# 2.2 Gamma spectrometry Analysis

A precise account of the energy and efficiency calibration done must be given. What is technical basis for the choice of a counting time of 10hrs. How the minimum detection limit (MD) was estimated must be indicted in the text This determines the appropriate counting time to be used.

# 2.3 Radiological Health Hazards indices assessment

# 2.3.1 Absorbed dose rate

Page 5 line 112 insert "for indoor and outdoor" before respectively.

# 2.3.2 Annual Effective Dose equivalent

Page 5 line 114 and page 6 lines 116- 120. Annual effective dose equivalent (AEDE) is an old radiation protection quantity now replaced by "annual effective dose (E)."

Pages 5 and 6 lines 114-120

Explanation for the symbols CRa-226. CTh-232 and CK-40 must be given in the text.

AEDE in; AEDE out and AEDE tot are old radiation quantities. The current radiation quality is annual effective (E).

Page 6 line 123 give the reference from which the dose conversion factors were taken.

# 2.3.3 Annual Gonadal dose equivalent 4

Page 6 line 128 Explanation for the symbols CRa-226. CTh-232 and CK-40 must be given in the text. What the values 3.09 ,4.18 and 0.0314 mean must be provided in the text.

# 2.3.4 Gamma Index and Alpha Index

Page 6 lines 133 and 136 Explanation of the symbols CRa-226, CTh-232 and CK-40 must be given in the text. What the values 300 ,200, 3000 and 200 mean in equations 5a and 5b must be given in the text.

# 2.3.5 Excess Lifetime Cancer risk

Page 7 line 139 replace below by "6a,6b and 6c" Page 7 line 143 replace non-deterministic by "stochastic " The life expectancy for South Africa must be used for the risk estimation. The value is certainly not 70 years.

# **Results and Discussions**

Page 11 line 207 instead of allowed limit use "reference level"

Page 11 line 233 instead of mortal use "fatal"

Pages 11 line 225 and page 12 lines 226-227. The sentence is beyond the scope of the work done. It is strongly recommended that you delete this sentence.

"The values of the risk obtained are within the acceptable risk range" is the recommended replacement. Low risk is within the range 1in 10,000 to 1in 1,000

Moderate risk is within the range 1in 1,000 to 1 in 500

High risk 1 in 500 to 1in 100

# 4.Conclusion

The conclusions must capture conclusions from the findings of the objectives of the study.

Page 12 lines 229-233 must deleted since it not a conclusion from the work.

Page 12 Line 239 complete by inserting 2000, "UNSCEAR 2000 Report "

Page 12 line 246 instead permissible limit use "reference level "

Page 248 instead of recommended limit use "reference level"

Page 13 lines 249 to 252. 40K cannot be regulated according to IAEA GSR Part 3.

It will be better to conclude on your finding on the risk assessment conducted. 5

Is the risk level assessed low, moderate, acceptable or high? It there the need to establish a National Radiation Protection strategy to reduce risk by adopting dose reduction strategies? It is recommended that you reword your conclusion captured on page 13 lines 249 to 252 accordingly.

# References

Reference 10 issue number missing Reference 16 page numbers incomplete Reference 19 volume and issue numbers missing Reference 21 volume and issue numbers missing Reference 30 Issue number missing Reference 34 Issue number missing Reference 36 page numbers incomplete

#### Author response to Reviewer 2: Round 1

# Abstract

- 1. Line 14: Recommended and Permissible limits changed to "recommended reference levels".
- 2. Line 15 changed to "Therefore, the use of cement products as building materials presents no significant risk in the study areas".

#### Introduction

- 3. Line 22 "that the residents irradiate revised as "which constitute radiation exposure to the resident. The radiation exposure takes place daily....."
- 4. Line 29 "non- radioactive" deleted.
- 5. Line 34 non-deterministic replaced by "stochastic"
- 6. Lines 54 -62 sentences are re-worded as follows:

"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 optimizing the protection of the public when using and managing cement as building material, as required by IAEA GSR Part 3. Therefore, this study seeks to evaluate the radiological health and safety risks associated with radiation exposure due to the radioactivity concentrations of the primordial radionuclides (<sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K) in cement commonly used for buildings in South Africa."

#### 2.1 Collection and Preparation of Samples

7. Lines 65 - 70 "the sample collection procedure was rewritten".

#### 2.2 Gamma Spectrometric Analysis

- 8. Lines 83 85 "A precise account of the energy and efficiency calibration has been added"
- 9. Line 87 "The technical basis for the choice of a counting time of 10hrs has been added"
- 10. Lines 96 & 97 "Statement on how minimum detection limit (MD) was estimated was added"

#### 2.3 Radiological Health Hazards indices assessment

#### 2.3.1 Absorbed dose rate

11. Line 112 "for indoor and outdoor" inserted before respectively.

#### 2.3.2 Annual Effective Dose equivalent

- 12. Annual effective dose equivalent (AEDE) replaced by "annual effective dose (E)." throughout the manuscript.
- 13. Explanation of the symbols  $C_{Ra-226}$ ,  $C_{Th-232}$  and  $C_{K-40}$  given in the text.
- 14. Line 123, the reference from which the dose conversion factors were taken given.

#### 2.3.3 Annual Gonadal dose equivalent

15. Line 128, Explanation for the symbols  $C_{Ra-226}$ ,  $C_{Th-232}$  and  $C_{K-40}$  given in the text. Meaning of values 3.09 ,4.18 and 0.0314 provided in the text.

#### 2.3.4 Gamma Index and Alpha Index

16. Lines 133 & 136; Explanation of the symbols C<sub>Ra-226</sub>, C<sub>Th-232</sub> and C<sub>K-40</sub> given in the text. Meaning of values 300, 200, 3000 and 200 in equations 5a and 5b given in the text.

# 2.3.5 Excess Lifetime Cancer risk

- 17. Line 139 "below replaced by "6a, 6b and 6c"
- 18. Line 143 "non-deterministic replaced by stochastic"

# **Results and Discussions**

- 19. Line 207 "allowed limit replaced by reference level"
- 20. Line 233 "mortal replaced by fatal"
- 21. Lines 225 227 "sentences deleted and replaced by The values of the risk obtained for this study are within the acceptable risk limits.."

# 4. Conclusion

The conclusions must capture conclusions from the findings of the objectives of the study.

Page 12 lines 229-233; deleted.

Page 12 Line 239; 2000 inserted, "UNSCEAR 2000 Report"

Page 12 line 246; permissible limit replaced by "reference level"

Page 248; recommended limit replaced by "reference level"

# References

Reference 10; issue number inserted

Reference 16; page numbers inserted

Reference 19; volume and issue numbers inserted

Reference 21; volume and issue numbers inserted

Reference 30; Issue number inserted

Reference 34; Issue number inserted

Reference 36; page numbers completed

Reference 46; Issue number inserted

Reference 47; page numbers inserted

Reference 48; page numbers inserted

Reviewer 2: Round 2 Date completed: 02 August 2024 Recommendation: Accept / Revisions required / Resubmit for review / Resubmit elsewhere / Decline / See comments Conflicts of interest: None

Does the manuscript fall within the scope of SAJS?

Yes/No

•
Is the manuscript written in a style suitable for a non-specialist and is it of wider interest than to specialists
alone?
Yes/No
Does the manuscript contain sufficient novel and significant information to justify publication?
Yes/No
Do the Title and Abstract clearly and accurately reflect the content of the manuscript?
Yes/No
Is the research problem significant and concisely stated?
Yes/No
Are the methods described comprehensively?
Yes/No
Is the statistical treatment appropriate?
Yes/No/Not applicable/Not qualified to judge
Are the interpretations and conclusions justified by the research results?
Yes/Partly/No
Please rate the manuscript on overall contribution to the field
Excellent/ <b>Good</b> /Average/Below average/Poor
Please rate the manuscript on language, grammar and tone

Excellent/Good/Average/Below average/Poor

# Is the manuscript succinct and free of repetition and redundancies?

Yes/No

#### Are the results and discussion confined to relevance to the objective(s)?

Yes/No

The number of tables in the manuscript is

Too few/Adequate/Too many/Not applicable

The number of figures in the manuscript is

Too few/**Adequate**/Too many/Not applicable

Is the supplementary material relevant and separated appropriately from the main document?

#### Yes/No/Not applicable

Please rate the manuscript on overall quality

Excellent/Good/Average/Below average/Poor

Is appropriate and adequate reference made to other work in the field?

#### Yes/No

Is it stated that ethical approval was granted by an institutional ethics committee for studies involving human subjects and non-human vertebrates?

# Yes/No/Not applicable

If accepted, would you recommend that the article receives priority publication?

#### Yes/No

Are you willing to review a revision of this manuscript?

Yes/No

With regard to our policy on '<u>Publishing peer review reports</u>', do you give us permission to publish your anonymised peer review report alongside the authors' response, as a supplementary file to the published article? Publication is voluntary and only with permission from both yourself and the author.

#### Yes/No

#### **Comments to the Author:**

# General Comments

The current form of the manuscript is acceptable for publication. Minor corrections detected must be effected.

#### Specific Comments Abstract

OK.

UK.

Introduction

ОК

#### 2. Materials and Methods Sampling and Sample size deter

Sampling and Sample size determination OK

# 2.1Collection and Preparation of Samples

Page 3 line 62 a reference must be cited to support the statement made.

# 2.2 Gamma spectrometry Analysis

Page 3line 72 Model and Serial Numbers must be provided for PalmTop MCA analyser system

# **2.3 Radiological Health Hazards indices assessment** OK

# 2.3.2 Annual Effective Dose equivalent

ОК

**2.3.3 Annual Gonadal dose equivalent** OK

2.3.4 Gamma Index and Alpha Index

ОК

# 2.3.5 Excess Lifetime Cancer risk

The lifetime duration for South Africa is 65.10 years. The values estimated are higher since 70 years was used.

**Results and Discussions** 

ОК

# 4.Conclusion

OK.

# Reference

Include the relevant reference for the Statement "The sample size was determined based upon practical consideration and standard practice in environmental and material science research"

# Author response to Reviewer 2: Round 2

References have been added to Page 3, line 62, to support the statement.
 The model and serial numbers of the Palmtop MCA analyzer system used have been added to the manuscript.
 The lifetime duration for South Africa (65.10 years) have been used in the calculation of Excess Lifetime Cancer Risks and new values changes effected in Table 3 and the manuscript.

# Radiological Risk Assessment of Cement Used in Contemporary South African Buildings

#### 3 Abstract

4 Using a calibrated NaI(TI) and a well-shielded detector connected to a computer-resident 5 quantum multichannel analyzer, the radionuclide contents of primordial radionuclides (<sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K) were evaluated in commonly used cement brands in South Africa, 6 7 and the associated radiation risk parameters were calculated in this study. The reported activity concentrations varied from  $(8.00 \pm 2.83 \text{ to } 45.00 \pm 2.79 \text{ Bg.kg}^{-1})$ ,  $(12.00 \pm 0.90 \text{ to } 10.00 \text{$ 8  $32.00 \pm 0.51$  Bq.kg<sup>-1</sup>) and (454.00  $\pm 0.56$  to 1765.00  $\pm 0.93$  Bq.kg<sup>-1</sup>), for <sup>226</sup>Ra, <sup>232</sup>Th, and 9 10 <sup>40</sup>K, respectively. The absorbed gamma dose rate, annual effective dose equivalent, 11 annual gonadal dose equivalent, excess lifetime cancer risk, gamma index, and alpha 12 index have been utilized as the radiological health impact metrics to evaluate the 13 potential radiation risks. The determined radiological health impact parameters results 14 were below relevant radiation safety authorities' recommended and permissible limits for 15 building materials. Therefore, using cement samples as construction materials in the 16 study areas is radiologically safe.

17 Keywords: activity concentrations, radiological hazards, cement, radionuclides, excess
18 lifetime cancer risk.

19 **1.** 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 that the residents irradiat This irradiation takes place daily, and the capability of radionuclides to quickly find their way into the air makes

them be transferred into human environments.<sup>1,2</sup> Most humans spend approximately 80% 24 of their lifetime indoors, so assessing the radionuclides contents in cement used as 25 building materials and related radiological health hazards on humans is essential.<sup>3</sup> 26 27 Naturally occurring radionuclides of primordial origin in types of cement used as building materials are responsible for irradiation in dwellings.<sup>4</sup> The radiations come from 28 non-radioactive Potassium-45 and gamma radiation from the Uranium and Thorium 29 family.<sup>5</sup> Gamma radiation exposure causes external exposure when it is directly 30 absorbed. Internal exposure, however, is brought on by Radium-226 and Thorium-232, as 31 32 well as their daughter nuclides, including Radon-222 and Thoron-220, and their progenies.<sup>6</sup> Varying degrees of radiation exposure in man have been reported to lead to 33 34 deterministic and non-deterministic effects, including cancer and genetic defects like chromosome aberrations and mutation.<sup>7,8</sup> 35

The South African population has increased annually by about 2% since 1980.9 In the 36 37 same vein, South Africa's sales of types of cement have progressively increased from seven million tonnes in 1980 to eleven million tonnes in 2010.<sup>10</sup> There is a clear 38 39 indication of a corresponding increase in demand for housing as a basic human need due to population increase. Therefore, to cater for the ever-increasing demand for housing by 40 41 the populace, increased demands in construction materials are inevitable. In both rural and urban areas of South Africe ment is among the most essential building materials 42 43 used to construct homes and other structures. It uses in construction is inevitable because it is used in concrete and block production = oring, and covering of the building floors 44 45 and walls. Thus, cement has played and will keep on playing significant roles in meeting 46 South Africa's developmental agenda because buildings with cement as an essential 47 component are virtually everywhere.<sup>11</sup> Buildings must contain cement because of its 48 many beneficial properties, such as its 'bond'-like function, its role in filling the spaces 49 between fine and coarse aggregates, and its hydration reaction properties that allow 50 buildings to be gaining strength continuously <sup>12</sup> and till now no suitable materials with 51 better or similar qualities had been discovered as an alternative to cement in buildings.

52 The research on primordial radionuclide concentrations in cement used as building 53 materials in numerous countries throughout the world has garnered much attention over the years.<sup>13-15</sup> Understanding the concept of radioactivity concentrations in cement is 54 55 significant in evaluating the possible radiological health hazards to residents and 56 developing reference standards and guidelines for using and managing cement as building materials.<sup>16</sup> However, further information regarding primordial radionuclide 57 58 radiation concentrations in cement and other building materials from South Africa has to be reported. Therefore, this study seeks to evaluate the associated radiological health 59 60 concerns on the population of the study region by measuring the radioactivity concentrations of the primordial radionuclides (226Ra, 232Th, and 40K) in cement 61 62 commonly used for buildings in South Africa.

63

# 64 2. Materials and Methods

**Collection and Preparation of Samples** 

# **65 2.1**

In Pietermaritzburg, South Africa, samples of cement from commonly used brands were
obtained from suppliers of building materials and labeled appropriately. The Pretoria
Portland Cement Company (PPC), Natal Portland Cement Company (NPC), and Dangote
Cement South Africa (Sephaku) brands of cement were the most commonly used ones in

the research area. The collected samples were air-dried and sieved with a 2 mm mesh for homogeneity. Two hundred (200) grams of each sample were placed in Marinelli bottles with the same shape as the reference material for gamma spectrometric analysis. The bottles were well-labeled and sealed tightly with tapes to prevent radon permet with There were a total of seven samples divided among three popular cement brands.

75 2.2

# 2 Gamma Spectrometric Analysis.

The radioactivity concentrations of the naturally occurring radionuclides in the studied samples were measured with thallium-doped sodium iodide (NaI(TI)) gamma-ray spectrometry system at the Radiation Physics Research Laboratory of the [anonymised]. A multichannel computer-resident quantum analyzer (MCA2100R) and a well-shielded detector were attached to the system. Spectral analysis was done using Palmtop MCA computer gamma analysis software.

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 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 <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K. Each sample is counted for 36 000 seconds (10 hours).

Equation 1 was used to calculate the radioactivity concentration of the radionuclide from
 a measurement of the detector's efficiency <sup>17-19</sup>:

90 
$$C_{sp} = \frac{N_{sam}}{P_{E}.\epsilon.T_{c}.M}$$
(1)

91 where  $\varepsilon$  is the detection system's overall counting efficiency,  $C_{sp}$  is the activity 92 concentration of the radionuclides of interest in Bq.kg<sup>-1</sup>, N<sub>sam</sub> 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 Tc is the sample counting time. The Data Analytical tool in Microsoft Excel 2010 running on Windows 10 was used for the statistical analysis. The gamma spectrometry system's minimal detection limits (MDL) for the radionuclides  $^{226}$ Ra,  $^{232}$ Th, and  $^{40}$ K were 0.69, 0.78, and 2.35 Bq.kg<sup>-1</sup>, respectively.

# 98 2.3 Radiological Health Hazards Indices Assessment.

99 In cement samples from the study area, activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K 100 have been studied for potential radiological risks that could impact human health due to 101 radiation exposure. The radiological health impact metrics examined are the absorbed 102 gamma dose rate, annual effective dose equivalent, annual gonadal dose equivalent, 103 excess lifetime cancer risk, gamma index, and alpha index.

# 104 2.3.1 Absorbed Gamma Dose Rate

105 The following equations were applied to the measured activity concentrations to calculate 106 the indoor and outdoor absorbed gamma dose rates ( $D_{in}$  and  $D_{out}$ ) produced by gamma 107 radiation caused by <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K at a height of 1 m above the ground <sup>20</sup>:

108 
$$D_{in} (nGyh^{-1}) = 0.92 C_{Ra-226} + 1.1 C_{Th-232} + 0.081C_{K-40}$$
 (2a)

109 
$$D_{out} (nGyh^{-1}) = 0.462 C_{Ra-226} + 0.0604 C_{Th-232} + 0.0417 C_{K-40}$$
 (2b)

110 where the conversion factors for the doses associated with the radioactive concentrations

- of 226Ra, 232Th, and 40K for materials used as building materials are 0.92, 1.1, 0.081,
- 112 0.462, 0.0604, and 0.0417 in nGyh<sup>-1</sup>/Bq.kg<sup>-1</sup>, respectively.
- 113 2.3.2 Annual Effective Dose Equivalent
- 114 The annual effective dose equivalent (AEDE) has been calculated using the formulae (3a 115 - 3c)<sup>20,21</sup>:

116 AEDE<sub>in</sub> (
$$\mu$$
Svy<sup>-1</sup>) = 4.91 x D<sub>in</sub> (nGyh<sup>-1</sup>) (3a)

117 
$$AEDE_{out}(\mu Svy^{-1}) = 1.23 \text{ x } D_{out}(nGyh^{-1})$$
 (3b)

118 AEDE<sub>tot</sub> (
$$\mu$$
Svy<sup>-1</sup>) = AEDE<sub>in</sub> + AEDE<sub>out</sub> (3c)

The annual effective dose equivalents for indoor, outdoor, and total exposure are  $AEDE_{in}$ , AEDE<sub>out</sub>, and  $AEDE_{tot}$ , respectively. The annual effective dose equivalent (AEDE) 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 Sv.Gy<sup>-1</sup> from the air-absorbed dose rate to an effective dose.

124 2.3.3 Annual Gonadal Dose Equivalent

125 The exceptionally high radio-sensitivity of the human gonads, bone marrow, and bone 126 surface cells makes them organs of interest.<sup>22</sup> To calculate the annual gonadal dose 127 equivalent (AGDE), Equation 4 was utilized <sup>22</sup>:

128 AGDE (
$$\mu$$
Svy<sup>-1</sup>) = 3.09C<sub>Ra-226</sub> + 4.18C<sub>Th-232</sub> + 0.0314C<sub>K-40</sub> 4

130 To determine if the cement had complied with the radiological safety criteria for 131 construction materials, the gamma index (I) was calculated using the following equation 132  $^{23}$ :

133 
$$I_{\gamma} = \frac{C_{226}Ra}{300} + \frac{C_{232}Th}{200} + \frac{C_{40}K}{3000}$$
 5a

134 The alpha index (
$$I_{\alpha}$$
), which symbolizes the surplus alpha radiation brought on by  
135 breathing in radon-222 from the cement, was calculated using the equation below.

136 
$$I_{\alpha} = \frac{C_{226}Ra}{200}$$
 5b

137 2.3.5 Excess Lifetime Cancer Risk

138	The	annual	effective	dose	equivalent	(AEDE)	values	computed	as	specified	in	the
139	form	nulae bel	ow were u	used to	o calculate tl	ne excess	lifetime	cancer risk	s (I	ELCR) <sup>20,24</sup>	<sup>4</sup> :	

140 $ELCR_{in} = AEDE_{in} \times D_{l} \times R_{f}$	140	$ELCR_{in} = AEDE_{in} \ge D_1 \ge R_f$	68
---	-----	---	----

141  $ELCR_{out} = AEDE_{out} \times D_I \times R_f$  6b

142 
$$ELCR_{tot} = ELCR_{in} + ELCR_{out}$$
 6c

- 143  $R_f$  and  $D_l$  are the fatal cancer risk factors for non-deterministic effect (estimated 0.05 Sv<sup>-1</sup>
- 144 for the general public) and lifetime duration (70 years), respectively.

# 145 **3** Results and discussion

146 3.1 Natural radioactivity concentrations

As shown in Table 1, the measured activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in
cement samples are unevenly distributed.

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

Sample Id.	<sup>226</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>232</sup> Th (Bq.kg <sup>-1</sup> )	<sup>40</sup> K (Bq.kg <sup>-1</sup> )
NPC 1	$22.00 \pm 2.51$	$20.00 \pm 3.59$	$1662.00 \pm 1.15$
NPC 2	$15.00 \pm 2.64$	$32.00 \pm 0.51$	$569.00\pm0.72$
NPC 3	$8.00 \pm 2.83$	$12.00\pm0.90$	$1765.00 \pm 0.93$
Mean	$15.00 \pm 2.66$	$21.33 \pm 1.66$	$1332.00 \pm 0.94$
Alpine 1	$54.00\pm4.80$	$29.00\pm0.42$	$454.00\pm0.56$
Alpine 2	$16.00 \pm 4.70$	$25.00\pm0.24$	$1285.00 \pm 1.73$
Alpine 3	$35.00 \pm 2.41$	$17.00\pm0.48$	$1195.00 \pm 1.33$
Mean	$35.00 \pm 3.97$	$23.67\pm0.38$	$978.00 \pm 1.21$
Dangote	$45.00\pm2.79$	$15.00\pm1.28$	$1249.00 \pm 0.63$

150

The average activity concentration of <sup>226</sup>Ra in the cement samples was 27.857 Bq.kg<sup>-1</sup>, ranging from 8.00  $\pm$  2.83 to 45.00  $\pm$  2.79 Bq.kg<sup>-1</sup>. Dangote cement (Sephaku) had the highest value of <sup>226</sup>Ra, while Natal Portland cement (NPC 3) had the lowest value. With an average value of 21.43 Bq.kg<sup>-1</sup>, the measured activity concentrations of <sup>232</sup>Th in the cement samples ranged from 12.00  $\pm$  0.90 to 32.00  $\pm$  0.51 Bq.kg<sup>-1</sup>. The Natal Portland cement samples (NPC 2 and NPC 3) contained the highest and lowest results for <sup>232</sup>Th,

respectively. With an average value of 1168.43 Bq.kg<sup>-1</sup>, the recorded activity 157 158 concentrations of  ${}^{40}$ K in the cement samples ranged from  $454.00 \pm 0.56$  to  $1765.00 \pm 0.93$ Bq.kg<sup>-1</sup>. Pretoria Portland cement (Alpine 1) had the lowest value, whereas Natal 159 160 Portland cement (NPC 3) had the highest value for <sup>40</sup>K respectively. The World average values of radionuclides (<sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K) in building materials are 50, 50, and 500 161 Bq.kg<sup>-1</sup>, respectively.<sup>22</sup> Except for Alpine 1, which had activity concentration values of 162  $54.00 \pm 4.80$  Bq.kg<sup>-1</sup>, which is slightly higher than the World average value for  $\frac{226}{Ra}$ . 163 The measured values and averages of the activity concentrations for <sup>226</sup>Ra and <sup>232</sup>Th in 164 165 virtually all of the examined cement samples were found to be lower than the world average values. Also, except for Alpine 1, which had an active concentrations value of 166  $454.00 \pm 0.56$  Bq.kg<sup>-1</sup>, which is slightly lower than the World average value for <sup>40</sup>K, the 167 measured values and average of the activity concentrations for <sup>40</sup>K in almost all of the 168 169 examined cement samples were found to be higher than the world average values. In general, the mean activity concentrations of <sup>40</sup>K were the highest in all the cement 170 samples compared to the other two naturally occurring radionuclides (<sup>226</sup>Ra and <sup>232</sup>Th), 171 172 respectively. This is typical and expected from any geologically derived materials because potash feldspar minerals are relatively enriched in the natural environment.<sup>25</sup> The 173 174 concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in cement samples from the study locations are 175 depicted in Figure 1.



Figure 1: Activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K activity in the cement samples
 under study.

- 181 The results of earlier research from various parts of the world were also compared with 182 the calculated average values of the activity concentrations of naturally occurring 183 radionuclides in the studied cement samples. Table 2 displays the comparison.

- -

194 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	<sup>226</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>232</sup> Th (Bq.kg <sup>-1</sup> )	<sup>40</sup> K (Bq.kg <sup>-1</sup> )	Reference
NPC	South Africa	$15.00\pm2.66$	$21.33 \pm 1.66$	$1332.00 \pm 0.94$	Present study
Alpine	South Africa	$35.00\pm3.97$	$23.67\pm0.38$	$978.00 \pm 1.21$	Present study
Dangote	South Africa	$45.00\pm2.79$	$15.00 \pm 1.28$	$1249.00 \pm 0.63$	Present study
Cement Sample	Albania	$179.70\pm8.90$	$55.00 \pm 5.80$	$17.00 \pm 3.30$	26
Cement Sample	Algeria	$41.00\pm7.00$	$27.00\pm3.00$	$422.00\pm3.00$	27
Cement Sample	Bangladesh	61.00	65.00	952.00	28
Cement Sample	Cameroon	$27.00\pm4.00$	$15.00 \pm 1.00$	$277.00 \pm 117.00$	29
Cement Sample	China	$118.70\pm14.20$	$36.10 \pm 17.80$	$444.50 \pm 163.10$	30
Cement Sample	China	59.00	39.00	181.00	31
Cement Sample	Egypt	$36.00 \pm 4.00$	$43.00 \pm 2.00$	$82.00 \pm 4.00$	32
Cement Sample	Egypt	134.00	88.00	416.00	33
Cement Sample	Ghana	$35.94 \pm 0.78$	$25.44 \pm 0.80$	$233.00 \pm 3.95$	34
Cement Sample	India	26.00	29.00	260.00	35
Cement Sample	Iraq	$24.25 \pm 1.45$	$25.41 \pm 1.65$	$93.17 \pm 7.30$	36
Cement Sample	Laos	$41.12 \pm 2.44$	$16.60 \pm 2.37$	$141.48 \pm 4.50$	37
Cement Sample	Malaysia	$29.00 \pm 7.00$	$31.00 \pm 9.00$	$205.00 \pm 71.00$	38
Cement Sample	Morocco	$31.00 \pm 5.00$	$19.00 \pm 3.00$	$238.00 \pm 13.00$	39
Cement Sample	Nigeria	20.00	8.00	51	40
Cement Sample	Pakistan	$25.00 \pm 10.00$	$37.00 \pm 9.00$	$245.00 \pm 95.00$	41
Cement Sample	Serbia	37.00	15.00	43.00	42
Cement Sample	Senegal	$112.69 \pm 26.02$	$13.12 \pm 1.88$	$73.35 \pm 18.12$	43
Cement Sample	Turkey	$34.00 \pm 4.00$	$15.00 \pm 2.00$	$220.00 \pm 13.00$	44
Cement Sample	Turkey	26.00	10.00	130.00	45
196					

197 3.2 Radiological Hazard Indices.

198 Table 3 displays the findings of the assessed radiological health hazard parameters. The 199 table shows that the assessed indoor and outdoor absorbed gamma dose rates (Din and Dout) varied from 95.089 to 176.862 nGyh<sup>-1</sup> and 49.985 to 91.549 nGyh<sup>-1</sup>. All of the 200 cement samples' indoor absorbed gamma dose rates above the population-weighted 201 average of 84 nGyh<sup>-1</sup>.<sup>20</sup> The annual effective dose equivalents for indoor, outdoor, and 202 total exposure ranged from 466.887 to 868.392  $\mu$ Svy<sup>-1</sup>, 61.482 to 112.606  $\mu$ Svy<sup>-1</sup> and 203 528.369 to 980.998 µSvy<sup>-1</sup>, respectively. In Natal Portland cement (NPC 1 and NPC 2), 204 205 the highest and lowest values of the indoor and outdoor absorbed gamma dose rate and 206 the indoor, outdoor, and total annual effective dose equivalent were recorded. All of the

207	samples' annual effective dose equivalent values were below the allowed limit of 1000
208	$\mu$ Svy <sup>-1</sup> . <sup>20</sup> The annual gonadal dose equivalent (AGDE) range was 130.301 to 302.336
209	$\mu$ Svy <sup>-1</sup> . While Natal Portland cement (NPC 3) had the lowest value, Pretoria Portland
210	cement (Alpine 1) had the highest value of AGDE. Except for Alpine 1, whose yearly
211	gonadal dose equivalent value was slightly higher at 302.336 $\mu$ Svy <sup>-1</sup> . All of the samples'
212	recorded values were below the 300 $\mu Svy^{\text{-}l_{\star}}$ world average. The alpha index (I_{\alpha}) ranged
213	from 0.040 to 0.270, and the gamma index $(I_{\gamma})$ ranged from 0.040 to 0.727, respectively.

214	Table 3:	Calculated	radiological	health	hazard	indices.

Sample Id.	Din	Dout	AEDEin	<b>AEDE</b> <sub>out</sub>	AEDEtot	AGDE			<b>ELCR</b> <sub>in</sub>	ELCRout	ELCRtot
	(nGyh <sup>-1</sup> )	(nGyh <sup>-1</sup> )	(µSvy <sup>-1</sup> )	(µSvy <sup>-1</sup> )	(µSvy <sup>-1</sup> )	(µSvy <sup>-1</sup> )	Iγ	Iα	x 10 <sup>-3</sup>	x 10 <sup>-3</sup>	x 10 <sup>-3</sup>
NPC 1	176.862	91.549	868.392	112.606	980.998	203.767	0.727	0.110	3.343	0.434	3.777
NPC 2	95.089	49.985	466.887	61.482	528.369	197.977	0.400	0.075	1.798	0.237	2.034
NPC 3	163.525	84.545	802.908	103.990	906.898	130.301	0.675	0.040	3.091	0.400	3.492
Mean	145.159	75.360	712.729	92.692	805.422	177.348	0.601	0.075	2.744	0.357	3.101
Alpine 1	118.354	61.396	581.118	75.517	656.635	302.336	0.476	0.270	2.237	0.291	2.528
Alpine 2	146.305	76.077	718.358	93.574	811.932	194.289	0.607	0.080	2.766	0.360	3.126
Alpine 3	147.695	76.270	725.183	93.811	818.994	216.733	0.600	0.175	2.792	0.361	3.153
Mean	137.451	71.247	674.886	87.634	762.520	237.786	0.561	0.175	2.598	0.337	2.936
Dangote	159.069	81.933	781.029	100.778	881.807	240.969	0.641	0.225	3.007	0.388	3.395
215											

216 The gamma  $(I_{\gamma})$  and alpha  $(I_{\alpha})$  index values fell below the recommended upper limit of unity (1).<sup>46</sup> For indoor (ELCR<sub>in</sub>), outdoor (ELCR<sub>out</sub>), and total (ELCR<sub>tot</sub>) excess lifetime 217 218 cancer risk, respectively, the excess lifetime cancer risk values ranged from 1.798 x 10<sup>-3</sup> to  $3.343 \times 10^{-3}$ ,  $0.237 \times 10^{-3}$  to  $0.433 \times 10^{-3}$ , and  $2.034 \times 10^{-3}$  to  $3.777 \times 10^{-3}$ . The ELCR<sub>in</sub> 219 and ELCR<sub>tot</sub> reported in this study are higher than the world average values of  $0.29 \times 10^{-3}$ 220 and 1.45 x 10<sup>-3,</sup> reported by Mohammed & Ahmed.<sup>47</sup> The values of excess lifetime cancer 221 risk equivalent to 1000, 100, 10, and 1  $\mu$ Svy<sup>-1</sup> will increase the chance of developing 222 mortal cancer by 4%, 0.4%, 0.04%, and 0.004%, respectively.<sup>38,48</sup> The ELCR<sub>in</sub> and 223 ELCR<sub>tot</sub> reported in this study are higher than the world average values of  $0.29 \times 10^{-3}$  and 224 1.45 x 10<sup>-3,</sup> respectively.<sup>47</sup> Even though all excess lifetime cancer risk values were higher 225

than the world average, there is minimal possibility that lifetime cancer risks will increasedue to the low radionuclide concentrations reported for several cement samples.

#### **4.** Conclusion

229 A calibrated NaI(TI) and a well-shielded detector coupled to a computer-resident 230 quantum multichannel analyzer were used to measure the radionuclide contents of naturally occurring radionuclides (<sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K) and the associated radiological 231 232 hazards indices in commonly used cement brands in South Africa were estimated for this 233 study. According to the study, there were uneven distributions of the measured natural radioactivity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in the cement samples. In almost all 234 235 of the analyzed cement samples, the measured values and averages of the activity concentrations for <sup>226</sup>Ra and <sup>232</sup>Th were lower than the global average. In contrast, in 236 237 almost all of the analyzed cement samples, the observed values and average activity concentrations for <sup>40</sup>K were higher than the global average levels provided by 238 UNSCEAR. The findings show that <sup>40</sup>K is the radionuclide in the environment with the 239 240 highest measured radioactivity content. Radiological health impact measures, including 241 absorbed gamma dose rate, annual effective dose equivalent, annual gonadal dose 242 equivalent, excess lifetime cancer risk, gamma index, and alpha index, were established 243 to evaluate the potential radiation risks. The cement samples' indoor absorbed gamma 244 radiation rates were higher than the population-weighted global average of 84 nGyh<sup>-1</sup> 245 provided by UNSCEAR in 2000. The annual effective dose equivalent and annual 246 gonadal dose equivalent values for all the samples were lower than the permissible limit of 1000 µSvy<sup>-1</sup> and the world average value of 300 µSvy<sup>-1</sup>, respectively. The gamma 247 248 index  $(I_{\gamma})$  and alpha index  $(I_{\alpha})$  values were all below the recommended limit of unity.

Even if all of the determined extra lifetime cancer risks are higher than the global average value, there is very little chance that this will increase cancer risk in the long run. However, <sup>40</sup>K naturally occurring radioactivity content was higher than the global average, which could serve as a warning to the radiation safety authority.

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