Assessment of Natural Radioactivity and Chemical Composition of Cement produced in South Korea

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Abstract. The chemical composition of cement influences the physical and mechanical properties of concrete and the natural radionuclide levels. The cement composition depends on selecting of raw materials (lime-stone, fly ash, iron ore, and clay) and geological locations for cement production. The chemical composition and natural radioactivity levels of cements are important factors related to the strength of concrete and radiological risk to human life. This paper describes the characterization of various kinds of cement (grey, mortar, and white) samples produced in South Korea in terms of naturally occurring radionuclides (NORs) and chemical composition. The daughter nuclides of ^{238}U and ^{232}Th, and natural ^{40}K were determined by gamma-ray spectrometry, whereas chemical composition was determined by X-ray fluorescence spectrometry. The absolute efficiency of voluminous cement samples was determined by voluminous radioactive certified reference materials (CRM) and the EFFTRAN program. Methodologies used for concentrations determination were validated using matrix CRMs. Radiological indices and parameters, such as internal and external hazards indices, gamma and alpha indices, and annual effective dose, were calculated to estimate the risk in building materials. Results show that the natural radioactivity levels in different cement samples are lower than critical values, and the samples do not contain any significant source of radiation hazard. The chemical composition and quality of various locally produced cement samples is also examined. Some oxide (MgO, SO_3, and alkali) concentrations exceed the reference values, which may affect the properties of concrete. The statistical cluster analysis is used to discriminate cement samples. Approximate source apportionment of NORs is also determined by Pearson's correlations between radionuclides and oxide compounds. The concentrations of NORs in cement samples data are compared with values from other studies.

KEYWORDS: Cement, Natural radionuclides, Gamma-ray spectrometry, WD-XRF, Radiological-risk assessment, Source apportionment.

1 INTRODUCTION

Cement is one of the most important construction materials and plays a central role in developing countries' economic growth. South Korea is the top ten cement producer globally, and an average of 55 Mt of cement is produced per year [1-3]. Generally, cement is produced by mixtures of different geological raw materials, such as lime, silica, alumina, phosphogypsum, and iron ore. Limestone is the primary material for high-content Ca. It can be mixed with other materials, such as clay, mudstone, iron ore, and silica sand, to obtain the final cement's precise chemical composition [4]. Portland cement is popular for the most sustainable concrete and mortar design [4]. The cement contains major oxides, such as CaO (58.1%–68.0%), SiO_2 (18.4%–24.5%), Al_2O_3 (3.10%–7.56%), and Fe_2O_3 (0.16%–5.78%), as well as minor oxides SO_3 (0.0%–5.35%), MgO (0.02%–7.10%), Na_2O (0.10%–0.78%), and K_2O (0.04%–1.66%) [5]. The chemical composition assurance of cement is important in industries. Each oxide concentration must be within the range of standard specifications; otherwise, it leads to several physical and mechanical issues, such as unsoundness and failure in late-stage construction [4].

Along with the quality analysis of the chemical composition, radiological risk assessment due to naturally occurring radionuclides (NORs) is also important to human life. In developed countries, people spend more than 80% of their time indoors and are thus significantly exposed to NORs [6]. The concentrations of NOR in cement depend on the raw-material composition and local geological conditions. Accordingly the chemical composition and NOR activities of various kinds of cement samples need to be characterized.

Several studies have been carried out to determine the NOR (^{226}Ra, ^{232}Th, ^{40}K) concentrations in building materials and chemical composition analysis for quality estimation [1-3,5-9]. However, limited data are available from South Korea [10,11]. In addition, very limited data of white cement related to chemical composition and NORs available globally. In the present study, we analysed the chemical composition and NORs radioactivity in various cement samples produced in South Korea. The chemical composition of cement and potential radiological-risk indices and parameters were calculated. Results were compared with data from different countries.

2 MATERIALS AND METHODS

2.1 Sample Collection and Preparation

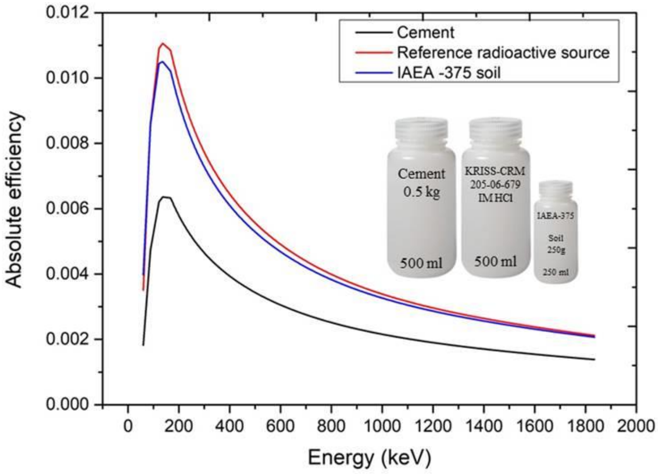
Ten different cement samples (about 2 kg per sample) were obtained from locally produced cement companies and are categorised as per the color and mixture; Three Portland cement (PC; C1–C3) are grey color; three readymade or mortar cement (MC; C4–C6) are mixture of cement and aggregates (sand or gravel); four white cement (WC; C7–C10) are white color cement. These samples are being used in various purposes such as construction and repair. The sample company names are not revealed as this is a scientific study and we do not assess specific company products.

Sample preparation for NOR measurement: About 500 g of cement samples were transferred into cylindrical polyethylene bottles and hermetically sealed. The sealed samples were left at room temperature for more than four weeks to enable them to reach the secular equilibrium between ^{222}Rn and ^{226}Ra.

Sample preparation for chemical composition measurement: A fusion bead was prepared with 1 g of cement sample mixed with 10 g of borate flux (50%-50% of lithium tetra-borate and lithium meta-borate flux). The final mixture was treated at 1050 ℃ and a 34 mm dia. fusion bead was prepared using KATANAX auto-bead machine. Each sample was prepared in duplicate and measured three times. The major advantage of the fusion bead method is to avoid grain size and mineralogical effects.

Gamma-ray spectrometry measurement: Samples were measured using high-resolution gamma-ray spectrometer with a 52% relative efficiency. The energy resolution of the detector is 1.8 keV at 1332.5 keV of ^{60}Co. The absolute efficiency of the HPGe detector was determined using the Korea Research Institute of Standards and Science (KRISS) gamma-emitting mixed radioactive nuclides certified reference material (CRM, no. 205-05-679, 450 ml, 1 M HCl solution) prepared in a bottle with identical geometry to that of samples to minimize the efficiency error from the geometrical difference. KRISS multi radioactive-nuclide CRM contained ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y, covering an energy range of interest of 59–1836 keV. The EFFTRAN (Monte Carlo efficiency transfer code) program [12] was used to transfer the efficiency from standard matrix geometry to cement sample geometry. This program included the coincidence correction and mass attenuation factor owing to the matrix. A typical, absolute full-energy peak efficiency of the HPGe detector for cement and reference samples is shown in Fig. 1. Each sample's absolute efficiency plot was obtained from the EFFTRAN program as samples had a different density.

Figure 1: Absolute full energy peak detection efficiency of the HPGe detector for various samples.



In the U series, ^{226}Ra has a relatively long half-life (1600 y), so we calculated the ^{226}Ra concentration instead of ^{238}U. The ^{226}Ra activity concentration was determined by the average of two daughter nuclides ^{214}Pb (351.9 keV) and ^{214}Bi (609.3 and 1120.3 keV). The ^{232}Th activity concentration was determined by the average of three daughter nuclides ^{228}Ac (911.2 keV), ^{212}Pb (238.6 keV) and ^{208}Tl (583.1 keV). The ^{40}K activity concentration was determined from its only gamma emission at 1460.8 keV. The International Atomic Energy Agency (IAEA) reference material soil-376 was measured under a similar condition to validate the efficiency transfer program and NOR activity calculation.

WD-XRF spectrometry measurement: The major and minor oxide compositions of cement samples were determined using high-resolution sequential type wavelength dispersive X-ray fluorescence (WD-XRF) spectrometry (Bruker S8 Tiger). The experiment was performed using the fusion bead method. The spectrometry was calibrated with various reference materials and validated with the National Institute of Standards and Technology (NIST) Standard Reference Materials® (SRMs) 1881b (Portland cement blended with Fly Ash) and 1889b (Portland cement blended with lime stone). The composition of fifteen oxides, namely, CaO, Al_2O_3, SiO_2, Fe_2O_3, SO_3, K_2O, MgO, MnO, Na_2O, P_2O_5, SrO, TiO_2, ZnO, ZrO_2, and NiO, were determined in all cement and standard reference samples. Loss on ignition (LOI) of cement samples should be less than 3% and the studied samples LOI at 400 ℃ for 2 h varied from 0.7% to 2.5%.

3 RESULTS AND DISCUSSION

3.1 Chemical Composition of Cement by XRF Analysis

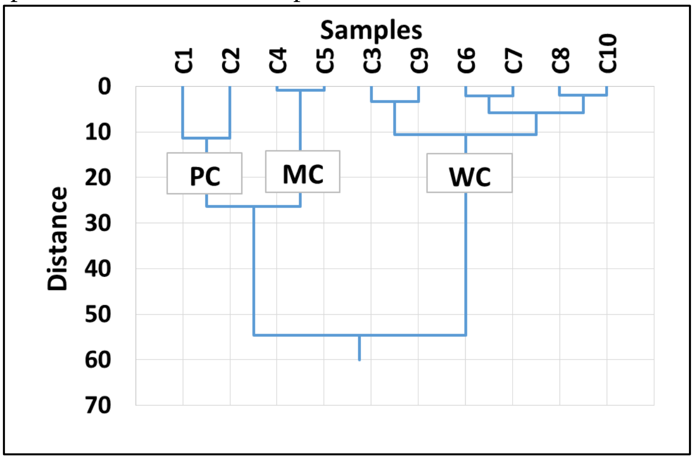
The major chemical composition of cement and reference (NIST SRMs) samples was determined using the Quanta express method [5]. The measured values of reference materials agree well with certified values within the range of ±8%. The major dominant oxide compositions of Korea cement samples and the standard range for Portland cement ASTM C150 (American Society for Testing and Materials) [13] are shown in Table 1. The composition of Korea Portland cement standard (KS L5204) is also similar to of ASTM C150.

Table 1: Major chemical composition (%) of cement samples and standard reference range of American Portland cement standard ASTM C150 [13].

	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	K_2O	SO_3	Na_2O
Portland cement (PC)								
C1	63.5	21.76	4.03	3.39	3.62	1.45	2.77	0.13
C2	62.0	22.40	5.14	3.97	1.41	1.19	2.68	0.17
C3	49.5	8.99	14.29	1.48	6.48	0.62	2.02	0.14
ASTM	61–67	19–23	2.5–6.0	<6	<5	N/A	<3	<1.0
Readymade/mortar cement (MC)								
C4	22.3	45.67	8.27	2.39	1.54	2.81	0.87	2.22
C5	22.4	45.18	7.92	2.44	0.94	2.68	0.62	2.08
C6	44.9	12.23	1.02	1.24	13.65	0.32	0.80	0.07
White cement (WC)								
C7	47.9	13.04	0.55	0.26	11.69	0.24	0.94	0.13
C8	50.1	17.34	0.95	0.26	6.39	0.29	11.69	0.22
C9	49.9	13.66	12.32	0.32	6.09	0.10	2.20	0.09
C10	48.8	9.83	1.16	0.61	12.42	0.13	1.39	0.06

Among the grey or Portland cement samples, C3's sample composition (CaO, SiO_2, Al_2O_3, and MgO) is out of the specified range of reference. The results of sample C3 specifications are lower than the standard specifications (ASTM C150). The lower quality cement may affect the strength and sound properties of concrete [4]. The readymade/mortar (remitar) cement is a mixture of cement and sand/gravel. Thus, its composition varies with the aggregate composition. Sample C4 and C5 samples contain high amount of Si, Al, K, and Na oxides owing to the dominated. In sample C6, cement composition is more dominated than aggregates. The chemical composition of white cement (C7–C10) significantly varies due to the different preparation procedures and selection of raw materials. The samples C3 (grey color) and C9 (white color) are from same company and fall in same cluster group. These two samples have similar chemical composition, however purposes of uses and characteristics (i.e. color) are different. Relatively same raw materials might be used to produce the different type of cements. Moreover, standard specifications are unavailable for white cement. However, selected oxide (e.g., MgO and SO_3) ranges should be of lower content; otherwise, the properties of concrete such strength and soundness would be risen, particularly in early age. MgO exceeds 5% in all white cement samples, and SO_3 exceeds in C8, resultant of these excess oxide influences the changes of soundness of properties, particularly with aging.

Figure 2: Cluster groups of Korea cement samples.



A statistical cluster analysis (CA) using Ward's method is applied to the cement samples by chemical composition to distinguish them (Fig. 2). Each sample is connected with other samples through distance. The distance parameter characterizes the similarity (small <10) and dissimilarity (largest >10) among them. The cement samples fall in three groups, and a good correlation is observed between similarity of group and type of sample (PC, MC, and WC) categorised physically, except two outliers (C3 and C6). These two outlier sample compositions are not similar to other relative type of samples (grey or mortar). The CA is simple and the best tool to identify outliers among samples. Generally, cement quality is verified by Bogue formula using oxide composition [5]. Bogue calculations also support our aforementioned conclusion about the quality of Portland cement that somehow sample C3 quality is lower than those of C1 and C2. This finding may lead to some physical and mechanical problems with aging. The CA is the best tool for the assessment of classification and quality of cement samples.

3.2 NORs in Cement Samples by Gamma-Ray Spectrometry

The activity concentration of NORs in cement samples and IAEA reference soil-375 are presented in Table 2 along with uncertainties. Good agreement is found between experimental and certified values of IAEA soil-375 sample, and the percentage deviation is within ±5%. The specific activity (A) and minimum detectable activity (MDAC) of NOR in cement and soil were determined using Equations (1) and (2) [7]:

$$A\ (Bqkg^{-1}) = \frac{N}{\varepsilon_\gamma \cdot \gamma_a \cdot t_s \cdot m_s} \tag{1}$$

$$MDAC\ (Bqkg^{-1}) = \frac{1.64 \cdot \sqrt{B}}{\varepsilon_\gamma \cdot \gamma_a \cdot t_s \cdot m_s} \tag{2}$$

where N and B are the counts under the peak and background, respectively; ε_γ is the efficiency of HPGe detector of specific energy; γ_a is the gamma-ray abundance; t_s is the measurement time in seconds; m_s is the mass of the sample in kilograms. The average MDAC of ^{226}Ra, ^{232}Th, and ^{40}K in cement is 0.41, 0.45, and 1.51 Bq/kg, respectively, whereas that of IAEA soil sample is 1.11, 089, and 2.06 Bq/kg, respectively.

Table 2: Activity concentrations of NORs (Bq/kg) in the studied samples and mean activity concentrations of top cement producing countries in the world [6-8].

Present study				Literature data			
Sample	^{226}Ra	^{232}Th	^{40}K	Country	^{226}Ra	^{232}Th	^{40}K
C1	24.5 ± 0.9	15.4 ± 1.2	264± 9	China	57	37	173
C2	34.5 ± 2.1	30.2 ± 1.6	264 ± 9	India	54	65	440
C3	17.3 ± 0.9	10.3 ± 0.6	100 ± 4	Vietnam	39.86	25.46	243
C4	18.7 ± 0.8	25.1 ± 1.3	868 ± 29	EU	45	31	216
C5	21.2 ± 0.9	33.3 ± 1.7	745 ± 25	Brazil	61.7	58.5	564
C6	28.6 ± 1.5	9.8 ± 0.5	21.4 ± 0.9	S. Korea	34.5	19.4	241
C7*	33.1 ± 1.2	3.8 ± 0.2	<1.5	Turkey	41	26	265
C8*	33.4 ± 1.3	7.1 ± 0.3	5.9 ± 0.4	Turkey*	31.7	15.1	100
C9*	20.8 ± 1.2	6.6 ± 0.5	<1.5	India*	24.4	19.8	176.7
C10*	11.2 ± 0.7	5.7 ± 0.4	<1.5	Kuwait*	17	8.7	199
**IAEA soil Exp./ certi.	19.9 ± 0.8 (20.0 ± 1)	20.2 ± 1.3 (20.5 ± 1.4)	402 ± 18 (424 ± 8)	Greece*	14–26	7–13	5–67

*white-cement sample data

**Certified values of reference materials of IAEA-375 mentioned in the brackets.

Table 2 shows the activity levels of ^{226}Ra, ^{232}Th, and ^{40}K in studied cement samples are about the mean values of the global range (39.4 for ^{226}Ra, 27.6 for ^{232}Th, and 289 Bq/kg for ^{40}K) and below the global typical values of 50, 50, and 500 Bq/kg, respectively, as per the guidelines of the UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation), 1993 [14]. The mean concentration of ^{226}Ra, ^{232}Th, and ^{40}K with standard deviation in three types of cement are 25.4 ± 8.6, 18.6 ± 10.3 and 209 ± 94 Bq/kg in Portland cement; 22.8 ± 5.1, 22.7 ± 11.9, and 545 ± 454 Bq/kg in mortar cement; 24.6 ± 10.7, 5.8 ± 1.5, and 2.6 ± 2.2 Bq/kg in white cement, respectively. By contrast, the mean activity of NORs in MC is slightly higher than other types of sample's mean (PC and WC) because of the mortar cement is mixed with aggregate (sand or gravel). The ^{226}Ra, ^{232}Th, and ^{40}K activities are lower than the mean value of the global range. The mean activity of ^{226}Ra is similar in PC and WC means that the composition of clinker is same. The ^{232}Th and ^{40}K activity levels depend on the additives (fly-ash, iron oxide, and clay) used in the cement production, in which negligible quantities are mixed with the clinker to maintain the precise composition of white cement, particularly the color. Literature data [3,7,9] and the present study show that the main contribution of ^{226}Ra in cement is from limestone, and those of ^{232}Th and ^{40}K come from the additives to clinker such as clay and fly-ash.

3.3 Radiological Hazard Parameters and Indices

To assess the radiological safety of building materials produced in Korea, simple radiological parameters and health indices, such as radium equivalent activity (Ra_{eq}), internal (H_{in}) and external (H_{ex}) hazard indices, gamma (I_γ) and alpha (I_α) indices, annual effective dose (AED) were adopted in this study. Those parameters and indices have been actively applied to building materials by a number of researchers to estimate the radiological risk caused by NORs [6-10].

3.3.1 Radium Equivalent Activity

The activities of NORs in building materials are distributed non-uniformly. A general index equation was derived to estimate the specific radioactivity level of NORs denoted as radium equivalent index (Ra_{eq}), and is given as Equation (3) [7].

$$Ra_{eq} = A_{226_{Ra}} + 1.43 \cdot A_{232_{Th}} + 0.077 \cdot A_{40_K} \tag{3}$$

where A is the specific activity of the corresponding radionuclide in Bq/kg. The radium equivalent dose values are presented in Table 3. As per the UNSCEAR report [14], the maximum allowed Ra_{eq} is 370 Bq/kg for safe in building materials which means that annual external dose will be below the 1.5 mSv/y. In the present study, Ra_{eq} values range from 19–126 Bq/kg in all kinds of cement samples, and they are below the recommended value.

3.3.2 External and Internal Hazard Indices

The external (H_{ex}) and internal (H_{in}) indices can help characterize the building materials. As per the UNSCEAR, 2000 reports, the radiation dose from building materials should be less than 1.5 mSv/y, and H_{ex} and H_{in} indices values should be less than 1 for safe building materials [14]. Inhalation of the uranium progeny of radon affects the respiratory organs. Internal exposure of NORs in the body can be calculated using the H_{in} index. The H_{ex} and H_{in} were calculated using Equations (4) and (5) [7] and are presented in Table 3. These indices range within 0.05–0.34 and 0.08–0.40, respectively, and they are below the recommended values found in all kinds of cement samples.

$$H_{ex} = \frac{A_{226Ra}}{370} + \frac{A_{232Th}}{259} + \frac{A_{40K}}{4810} \tag{4}$$

$$H_{in} = \frac{A_{226Ra}}{185} + \frac{A_{232Th}}{259} + \frac{A_{40K}}{4810} \tag{5}$$

3.3.3 Gamma and Alpha Indices

The limitation of excess gamma (I_γ) and alpha (I_α) radiation dose owing building materials were estimated using Equations (6) and (7) [7] and are presented in Table 3. For building materials, I_γ and I_α values (equivalent dose value) of ≤0.5 (0.3 mSv/y), ≤1 (1 mSv/y) and >1 (>1 mSv/y) are deemed as good, satisfactory, and unsuitable for construction purposes, respectively [8]. ^{226}Ra activity should be <200 Bq/kg as the limit of internal exposure is 1 mSv/y.

$$I_\gamma = \frac{A_{226Ra}}{300\,Bqkg^{-1}} + \frac{A_{232Th}}{200\,Bqkg^{-1}} + \frac{A_{40K}}{3000Bqkg^{-1}} \tag{6}$$

$$I_\alpha = \frac{A_{226Ra}}{200Bqkg^{-1}} \tag{7}$$

The calculated gamma and alpha indices range within 0.07–0.49 and 0.06–0.17, respectively. The ^{226}Ra activities in the studied samples are well below the limit value (<1 mSv/y). According to these indices, all samples fall within the satisfactory range and can be used to construct buildings.

3.3.4 Annual Effective Dose (AED)

Some buildings emit excessive radiation doses and thus affect humans staying in these buildings. The potential radiation contribution to the human body owing to indoor air is calculated by the external absorption dose (D_r) rate using Equation (8) [8]. Most people are deemed to spend 80% of their time within a building, and 0.7 (nGy/h) is set as conversion coefficient from the D_r to Annual Effective Dose (AED) for adults [14]. The annual absorption dose to the human body is calculated using Equation (9) [8], and results are presented in Table 3.

$$D_r(nGyh^{-1}) = 0.462 \cdot A_{226_{Ra}} + 0.604 \cdot A_{232_{Th}} + 0.0417 \cdot A_{40_K} \tag{8}$$

$$\text{AED (mSv}y^{-1}) = D_{in}(nGyh^{-1}) \cdot 8760h \cdot 0.7\ (Svy^{-1}) \cdot 0.8 \cdot 10^{-6} \tag{9}$$

The D_r values are found to range within 8–61 nGy/h owing cement as a building materials, and this value is less than the global average dose rate of 84 nGy/h [14]. The AED absorbed by the human body owing to various kinds of cement range between 0.04–0.30 mSv/y and which is also less than the critical value of 1 mSv/y set by the UNSCER for building materials [14].

Table 3: Radiological hazard indicators for ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in Korea cement samples

Sample	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	I_γ	I_α	AED (mSv/y)
C1	66.93	0.18	0.25	0.25	0.12	0.16
C2	98.01	0.26	0.36	0.35	0.17	0.22
C3	39.71	0.11	0.15	0.14	0.09	0.09
C4	121.40	0.33	0.38	0.48	0.09	0.29
C5	126.16	0.34	0.40	0.49	0.11	0.30
C6	44.20	0.12	0.20	0.15	0.14	0.10
C7	38.55	0.10	0.19	0.13	0.17	0.09
C8	44.00	0.12	0.21	0.15	0.17	0.10
C9	30.36	0.08	0.14	0.10	0.10	0.07
C10	19.51	0.05	0.08	0.07	0.06	0.04

3.4 Source Apportionment of NORs

The correlation coefficients between major chemical composition and NORs are presented in Table 4. The major chemical composition and NOR data of all samples were considered for Pearson's correlation calculations [9]. Most correlation coefficients are very low because of the diverse composition of different kinds of cement samples and the limited number of samples. As shown in Table 4, the correlation coefficients between ^{232}Th and ^{40}K are close to individual oxides. Thus, these radionuclides source may be the same minerals. ^{232}Th and ^{40}K show a strong positive correlation with Fe_2O_3, SiO_2, TiO_2, K_2O, Na_2O, and MnO. These oxides are rich in clay and fly-ash minerals. Previous studies [6] also showed that aluminosilicate material is the major contributor to ^{232}Th and ^{40}K, which is confirmed in the present study. ^{226}Ra is not correlated with any oxide composition except CaO. This finding means that the major source of ^{226}Ra is limestone. As observed in NOR data of white cement from literature [8] and the present study, the ^{226}Ra activity is two to five times higher than those of the other two radionuclides. In white cement, the major mineral composition is limestone. Therefore, the source of ^{226}Ra is limestone. This correlation study can help predict the possible minerals used in the cement preparation and source of NORs. However, this conclusion is drawn from limited samples, and extensive data sets are required to strengthen the concept.

Table 4: NOR correlation with chemical composition using Pearson's coefficients

	Al_2O_3	CaO	Fe_2O_3	K_2O	MgO	MnO	Na_2O	SiO_2	SO_3	TiO_2	^{226}Ra	^{232}Th	^{40}K
^{226}Ra	-0.45	*0.35*	0.11	-0.17	-0.02	0.08	-0.26	-0.09	0.22	-0.02	1.00		
^{232}Th	*0.25*	-0.40	*0.82*	*0.86*	-0.81	*0.72*	*0.70*	*0.82*	-0.25	*0.97*	0.04	1.00	
^{40}K	*0.30*	-0.72	*0.58*	*0.99*	-0.74	*0.39*	*0.95*	*0.97*	-0.31	*0.88*	-0.22	*0.84*	1.00

4 CONCLUSION

NORs in cement samples were examined using gamma-ray spectrometry. The concentrations of radionuclides vary in different cement samples because the raw materials are obtained from various places. These NORs affect the human body, so various indices were calculated using the NOR concentration. All radiological indices were found to be lower than the critical values set by the UNSCEAR and below the mean values found in cement from other countries. Among the studied samples, NOR data of mortar cement shows slightly higher than Portland and white cements because aggregate (sand) is dominant. We have not identified any significant radiological risk to humans owing to the studied cement samples, so they are suitable for building construction. Along with the estimation of radiological risk from cement samples, cement quality was checked using chemical composition determined by X-ray fluorescence. The chemical composition of Portland cement is important for its physical and mechanical properties. Among the oxide compositions, MgO, SO_3, and alkali concentrations in cement show the most adverse effects on concrete. Thus it should be below the <5%, <3.5%, and <1% levels, respectively. We also observed that some of the cement samples contained high concentrations of the aforementioned oxides. Those oxides affect the integrity of the concrete as it ages. An appropriate source of NORs is identified using the chemical correlation with NOR concentration. The main contribution of ^{226}Ra from limestone and ^{232}Th and ^{40}K from aluminosilicate silicate minerals in clay or fly ash are identified and found to agree well with previous findings [6,9].

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