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# Estimation of Radioactivity and Associated Radiological Hazards of Cement Used in Sri Lanka

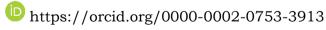
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### **Abstract**

Cement used in construction work can cause internal and external radiation exposure due to the presence of natural radionuclides  $^{226}\text{Ra},~^{232}\text{Th}$  and  $^{40}\text{K}$ . The radiation exposure risk can be estimated by finding the indoor absorbed dose rate and the annual effective dose. If the annual effective dose is within the internationally accepted value, use of cement can be considered safe and the risk will be within acceptable levels. The specific activities of  $^{226}\text{Ra},~^{232}\text{Th}$  and  $^{40}\text{K}$  in 85 samples from eleven types of cement were measured using gamma spectroscopy with a HPGe detector and the annual effective dose was calculated to determine the radiological hazard from the natural radioactivity in the samples. The average specific activities measured in Bq kg-1 ranged from 21.4  $\pm$  0.9 to 66.8  $\pm$  1.2 ;13.8  $\pm$  0.9 to 62.1  $\pm$  2.3 and 83.7  $\pm$  4.9 to 239.9  $\pm$  5.7 for  $^{226}\text{Ra},~^{232}\text{Th}$  and  $^{40}\text{K}$  respectively.

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The highest activity of both <sup>226</sup>Ra and <sup>232</sup>Th were obtained in CEM III- PPC 1 cement which contains 25 % fly ash from a coal power plant. The lowest activities of <sup>226</sup>Ra, <sup>40</sup>K and <sup>232</sup>Th were observed in CEM-X imported OPC 3 cement. It was observed that for all the studied cement samples the annual effective dose ranged from 0.15 to 0.51 mSv y<sup>-1</sup>.and less than the recommended maximum permissible public dose 1.0 mSv y<sup>-1</sup>. The results obtained in this study indicate that there is no radiological hazard arising from the use of the studied cement varieties in building construction.

**Keywords:** building construction, cement, equivalent dose, radiological hazard, radionuclides,

### **Introduction:**

All building materials, such as bricks, sand and cement derived from rocks and soil contain naturally occurring radionuclides of uranium (238U) and thorium (232Th) series and the radioactive isotope of potassium (40K). These radionuclides are also present in raw materials used in making cement, such as limestone, gypsum and fly ash. Radium (226Ra) which is a member of the 238U decay series is the most important isotope radiologically since radium and its daughter products produce 98.5% of the radiological effects of the uranium series. Therefore, the contribution from <sup>238</sup>U can be considered as that of <sup>226</sup>Ra and its daughter products. The radiation exposure from building materials due to these radionuclides is due to both internal and external exposures. The external exposure is caused by direct gamma radiation from an external source. The internal exposure is caused by the inhalation of the gaseous daughter products radon (222Rn) and thoron (220Rn) and their shortlived secondary decay products. Cement is used as a construction material for houses and other buildings in Sri Lanka. It is also used to make cement blocks and concrete as well as for plastering walls of houses and buildings. The demand for cement in Sri Lanka is increasing as a result of the increasing population and the expansion of infrastructure. Fly ash from coal power plants is used as a raw material in the manufacture of some brands of cement.

The aim of this study was to determine the specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in different brands of cement and in the raw materials used to manufacture cement and to assess the associated

radiological hazard from the use of cement. Cement samples were analyzed using high-resolution gamma spectrometry.

The potential radiological hazard was determined by computing the annual effective dose. The specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in Sri Lankan made cement were compared with the corresponding value of cement manufactured in some other countries.

The specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were also measured for raw materials of cement such as fly ash, gypsum, limestone, dolomite and clinker.

### **Materials & Methods**

### Sampling and sample preparation

A total of 85 cement samples manufactured in 2013 and 2014, from 11 main brands used in Sri Lanka were collected from the local market and from a cement factory. The types of cement and the proportions of raw materials in cement products are shown in Table 1.

**Table 1:** Brands of cement samples and the proportions of raw materials

Type of	No. of	Clink	Lime-	Fly	Minor
cement	Samp-	er	stone	ash	constituents
cement	les	(%)	(%)	(%)	(%)
CEM-I-OPC 1	10	95-	1	-	0-
		100			5(Gypsum/Dolomit
					e)
CEM-II-PLC	10	80-94	6-20	1	0-
					5(Gypsum/Dolomit
					e)
CEM-III-PPC 1	10	70	-	25	5 (Gypsum)
CEM-IV-OPC 2	5	91	1	5	4 (Gypsum)
CEM-V-OPC 3	5	94	1	2	4 (Gypsum)
CEM-VI-PPC 2	20	75	-	20	5 (Gypsum)
CEM-VII-OPC	5	95-	-	-	0-
4		100			5(Gypsum/Dolomit
CEM-VIII	5	95-	-	-	0-
Imported OPC		100			5(Gypsum/Dolomit
1					e)

CEM-IX	5	95-	-	-	0-
Imported OPC		100			5(Gypsum/Dolomit
2					e)
CEM-X	5	95-	-	-	0-
Imported OPC		100			5(Gypsum/Dolomit
3					e)
CEM-XI	5	65-89	-	11-35	0-
Imported PPC					5(Gypsum/Dolomit
1					e)

(OPC-Ordinary Portland Cement, PLC-Portland Limestone Cement, PPC-Portland Pozzolana Cement)

Thirty samples of raw materials were also collected from a cement factory. (Five samples each of limestone, dolomite, clinker, gypsum and 10 samples of fly ash).

The cement samples were dried in a temperature-controlled furnace for 14 hours at 110 °C to remove moisture. These samples were not sieved as the particles were very fine.

Some raw materials, such as clinker, dolomite and limestone were initially broken into small parts by using a hammer, milled using a grinder and sieved by a sieve of mesh size 250  $\mu$ m. Fly ash samples were not sieved as they were already in powdered form. Gypsum samples were also sieved using a 250  $\mu$ m mesh size without grinding. All raw material samples were oven dried at 110 °C for 14-21 hours to remove moisture.

Dried homogeneous samples of cement and raw materials of cement were thereafter sealed tightly in plastic containers (diameter 8.0 cm and height 2.5 cm) and kept for 30 days until radioactive equilibrium was reached.

## Measurement of specific radioactivity

The specific activities of the radionuclides were determined by high resolution gamma spectrometry using a HPGe (Hyper Pure Germanium) detector with a relative efficiency of 20.6% and an energy resolution of 1.85 keV FWHM (full width at half-maximum) for the 1332 keV gamma ray line of 60Co. GENIE 2000 software was used to analyze the spectra. The high resolution HPGe gamma spectrometry system consists of a p-type intrinsic Germanium co-

axial detector (type: EG and ORTEC model GEM 13200) mounted vertically and coupled to a multi-channel analyser. (Canberra S100 MCA, with 4023 channels). The detector was mounted in a cylindrical lead shield with a thickness of 10 cm to reduce the effect of background radiation.

The spectrometer was calibrated for energy and efficiency over the photon energy range of 186 keV - 2700 keV using IAEA reference materials, RGU-1 (U-ore), RGTh-1 (Th-ore) and RGk-1 (K<sub>2</sub>SO<sub>4</sub>), packed in containers whose geometry was identical to that of the geometry of cement and raw material samples. The accumulation time for gamma ray spectra ranged between 61,200 and 64,800 seconds, sufficient for a statistical error of less than 1%. Background measurements were also taken over the same period of time. For all samples, the <sup>226</sup>Ra specific activity was measured from the gamma ray line of 351.9 keV (37.1%) from <sup>214</sup>Pb and 609.3 (46.1%), 1120.2 (15.1%) and 1764.5 (15.9%) keV lines from <sup>214</sup>Bi. The specific activity of 232Th was determined from 583.1 (86%) and 2614 keV gamma lines of <sup>208</sup>Tl, 238.6 (43.6%) keV line from <sup>212</sup>Pb and 338.4 (12%) and 911.1 (29%) keV lines from <sup>228</sup>Ac. The <sup>40</sup>K concentration was calculated from its 1460.8 keV gamma line. The values inside the parentheses are the absolute probabilities of the gamma decay.

### Estimation of specific activity

The activities of radionuclides in the samples were computed using equation (1). The specific activities in Bq kg<sup>-1</sup> of the radionuclides were calculated by dividing the activity by the mass of the dried samples expressed in kilogram using equation (2).

The activity A and specific activity  $A_{sp}$  are given by,

$$A = \frac{Z}{r\epsilon t}$$

$$A_{\rm sp} = \frac{A}{M} = \frac{Z}{Mr\epsilon t}$$
(2)

Where, Z is the background subtracted counts under the photopeak, r - the gamma emission probability, t -the counting time of samples and  $\epsilon$  the photopeak efficiency for the corresponding gamma energy and M the dry mass of the sample.

The photopeak efficiency  $\epsilon$  was calculated using equation (3),

$$\epsilon = \frac{Z'}{rA_{st}t'} \tag{3}$$

Where,  $A_{st}$  - Activity of the IAEA reference material (RGU), t'-counting time of the reference material, r - gamma emission probability, Z'- the background subtracted count under the photopeak for the reference material.

### **Estimation of Uncertainty of Specific Activity**

The uncertainty of the specific activity depends on the uncertainty of the measured parameters, namely the count-rate, the efficiency of the detector and the mass of the sample.

$$A_{sp} = \frac{Z}{Mret} = \frac{C}{Mre} \tag{4}$$

Where,  $c = \frac{Z}{t}$  is the count-rate.

It can be shown that the most probable error of the specific activities can be expressed by the equation 5 (Topping, 1972).

$$\left(\delta A_{sp}\right)^{2} = A_{sp}^{2} \left\{ \left(\frac{\delta C}{C}\right)^{2} + \left(\frac{\delta \epsilon}{\epsilon}\right)^{2} + \left(\frac{\delta r}{r}\right)^{2} + \left(\frac{\delta M}{M}\right)^{2} \right\}$$
 (5)

Where,  $\delta A_{sp}$  - The uncertainty of the specific activity,  $\delta c$  - The uncertainty of the count-rate,  $\delta r$  - The uncertainty of gamma emission probability,  $\delta \epsilon$  - The uncertainty of the efficiency,  $\delta M$  - The uncertainty of the measurement of mass.

## **Estimation of Minimum Detectable Activity (MDA)**

The MDA is a measure of the lowest level at which sample activity can be distinguished from the background. The minimum detectable activity (MDA) of the gamma spectroscopic system was calculated using the equation (6) (Curie, 1968).

$$MDA = \frac{k\alpha\sigma}{\epsilon rtM} (Bq kg^{-1})$$
 (6)

Where,  $\sigma$  – The standard deviation of the background in the region of interest which is the square root of the number of counts of the

spectrum in a given time, since it has a Poisson distribution, and  $k\alpha$  –The statistical coverage factors equal to 1.645 at 95% confidence level.

#### Assessment of the radiation hazard

In this study, the indoor annual effective dose was determined to assess the potential radiation hazard.

### The Annual Effective Dose Rate (A.E.D.)

The indoor annual effective dose due to the gamma ray emission from the radionuclides <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in cement used as a building material to construct a reference room with dimensions 6 m x 4 m x 3 m can be calculated from the Equation 7 given below (Interim report, The Netherlands, 1985). It is assumed that the ceiling, walls and the floor of the room are made of concrete with density 2400 kg m<sup>-3</sup> and thickness 12 cm.

$$A.E.D. = DtTF \tag{7}$$

Where, D - Absorbed dose rate in air mGy h<sup>-1</sup>, t - the number of hours in a year = 8760 h y<sup>-1</sup>, F - Conversion factor of absorbed dose in air to effective dose = 0.7 (Sv Gy<sup>-1</sup>), T = The indoor occupancy factor.

External exposure to gamma radiation from natural radioactive elements occurs both indoors and outdoors. For calculation of annual dose it is important to take into account the occupancy factor. Here it is assumed that people spend 20% of their time outdoors and 80% indoors on an average worldwide (UNSCEAR, 2008). Hence the indoor occupancy factor *T* is taken as 0.8.

The absorbed dose rate (*D*) can be expressed as a sum of contributions from the different radionuclides in the building material (Equation 8).

$$D = q_{Ra}C_{Ra} + q_{Th}C_{Th} + q_kC_k$$
 (8)

Where,  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the specific activities in Bq kg<sup>-1</sup> of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively,  $q_{Ra}$ ,  $q_{Th}$ ,  $q_k$  are the factors for converting the radioactivity concentrations in a building material to the

absorbed dose rate in air and at 1 m distance away from the surface of the material (Interim report, The Netherlands, 1985).

$$q_{Ra}$$
 = 620 x 10<sup>-9</sup> mGy h<sup>-1</sup>/Bq kg<sup>-1</sup>  
 $q_{Th}$  = 890 x 10<sup>-9</sup>mGy h<sup>-1</sup>/Bq kg<sup>-1</sup>  
 $q_k$  = 54 x 10<sup>-9</sup> mGy h<sup>-1</sup>/Bq kg<sup>-1</sup>

### **Results and Discussion**

### Specific activity

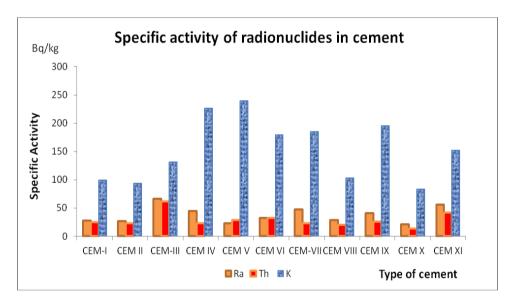
The specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were determined for all the studied cement samples and raw materials of cement. The mean specific activity values of these radionuclides with the statistical uncertainty are presented in Table 2 and Table 3 respectively. These results are also shown graphically in Figure 1 and Figures 2, 3 and 4.

**Table 2:** The range and the mean specific activities with uncertainty of \$^{226}Ra, \$^{232}Th and \$^{40}\$K of all types of cement samples in Bq kg<sup>-1</sup>

Type of		<sup>226</sup> Ra (Bq	<sup>232</sup> Th (Bq	<sup>40</sup> K (Bq kg <sup>-1</sup> )
Cement		kg-1)	kg-1)	
CEM-1 OPC 1	Mean	27.9 ±	25.1 ± 0.9	99.5 ± 4.2
		0.7		
	Max.	33.7 ±	29.4 ± 3.5	126.6 ±
		2.3		13.6
	Min.	20.8 ±	$18.7 \pm 2.6$	50.6 ±
		2.0		13.3
CEM-II PLC	Mean	26.5 ±	$23.4 \pm 0.8$	93.3 ±
		0.6		3.5
	Max.	29.5 ±	26.2 ± 2.9	103.9 ±
		1.9		11.9
	Min.	22.8 ±	$20.9 \pm 2.6$	62.5 ±
		1.9		10.5
CEM-III PPC	Mean	66.8 ±	62.1 ± 2.3	131.5 ±
1		1.2		5.9

	1	T	T	1
	Max.	71.3 ±	$68.9 \pm 6.3$	168.3 ±
		2.8		14.6
	Min.	62.7 ±	57.7 ±5.7	103.2 ±
		2.9		13.1
CEM-IV OPC	Mean	44.9 ±	$23.0 \pm 1.3$	226.4 ±
2		0.9		6.7
	Max.	46.2 ±	$26.7 \pm 3.1$	240.5 ±
		2.2		14.9
	Min.	42.4 ±	$19.4 \pm 2.4$	209.3 ±
		2.2		15.2
CEM-V OPC 3	Mean	23.1 ±	28.8 ± 1.2	239.9 ±
		0.8		5.7
	Max.	25.5 ±	31.7 ± 2.9	258.5 ±
		1.9		12.5
	Min.	19.9 ±	26.3 ± 2.4	224. 4 ±
		1.9		12.3
CEM-VI PPC	Mean	32.4 ±	32.8 ± 1.2	179.5 ±
2		0.7		3.8
	Max.	52.9 ±	43.0 ± 3.6	220.1 ±
		2.6		11.6
	Min.	18.7± 0.9	20.4±2.2	158.5±
				12.5
CEM-VII OPC	Mean	47.5 ±	22.8 ± 1.2	185.5 ±
4		0.9		5.3
	Max.	49.0 ±	24.5 ± 2.6	197.8 ±
		2.1		11.4
	Min.	46.5 ±	21.0 ± 2.6	174.1 ±
		2.1		11.6
CEM-VIII	Mean	29.1 ±	20.6 ± 1.1	102.8 ±
Imported		0.9		4.9
OPC 1	Max.	29.8 ±	25.0 ± 2.5	110.8 ±
		2.1		10.6
	Min.	28.6 ±	18.8 ± 2.3	82.4 ±
CEM-IX	Mean	40.9 ±	25.7 ± 2.1	195.2 ±
Imported OPC	liiodii	0.9		5.6
2	Max.	44.2 ±	28.4 ± 3.2	218.4 ±
		2.2		12.5
	Min.	38.6 ±	24.1 ± 2.6	174.6 ±
		2.1		12.5
I	1	I.	T.	I.

CEM-X	Mean	21.4 ±	13.8 ± 0.9	83.7 ± 4.9
Imported OPC		0.9		
3	Max.	23.5 ±	15.6 ± 1.8	102.2 ±
		1.8		10.4
	Min.	19.5 ±	12.5 ± 1.8	72.9 ±
		1.9		10.8
CEM-XI	Mean	55.9 ±	41.4 ± 1.8	152.3 ±
Imported PPC		1.1		5.6
1	Max.	56.6 ±	43.2 ± 4.3	159.7 ±
		2.1		13.6
	Min.	54.1 ±	39.6 ± 3.9	138.3 ±
		2.1		1.0



**Figure 1:** Mean Specific activities of radionuclides in cement samples

As can be seen from Table 2 and Figure 1 the specific activities of 226Ra and  $^{232}$ Th were maximum in CEM-III PPC 1 cement. The specific activity of  $^{226}$ Ra was found in the range 21.4 ± 0.9 to 66.8 ± 1.2 Bq kg<sup>-1</sup> for all cement samples. For  $^{232}$ Th it was found in the range 13.8 ± 0.9 to 62.1 ± 2.3 Bq kg<sup>-1</sup>. The lowest value for  $^{226}$ Ra and  $^{232}$ Th were observed in CEM-X imported OPC 3 cement.

In the case of <sup>40</sup>K, the CEM-V OPC 3 cement samples showed the highest concentration, whereas, CEM-X imported OPC 3 cement samples showed the lowest concentration. The highest and the

lowest concentrations were 239.9  $\pm$  5.7 Bq kg<sup>-1</sup>and 83.7  $\pm$  4.9 Bq kg<sup>-1</sup> respectively.

According to the results shown in Table 3 and Figures 2, 3 and 4 the concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in fly ash are much higher than that of the other raw materials. About 25% and 20% fly ash is added to CEM-III PPC 1 cement and CEM-VI PPC 2 cement respectively during manufacturing. About 5% of fly ash is added to CEM-IV OPC 2 cement and about 2% of fly ash is added to CEM-V OPC 3. The reason for the highest specific activities of <sup>226</sup>Ra and <sup>232</sup>Th in the CEM-III PPC 1 cement is that it contains 25% of fly ash produced by combustion of coal. The power plant uses different types of coal and had used Indonesian coal during the period of samples of this brand of cement were collected.

CEM-IV OPC 2 cement samples were collected in June 2013. During this period the power plant used a mixture of South African and Indonesian coal. The specific activities of radionuclides in fly ash emitted during combustion of the mixture of South African and Indonesian coal was higher than what was produced from Indonesian coal. Therefore, a higher concentration of <sup>226</sup>Ra was found in CEM- IV OPC 2 cement than some other local brands of cement.

The worldwide average specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in the Earth's crust were estimated as 32, 45 and 412 Bq kg<sup>-1</sup> respectively (UNSCEAR, 2008). In all the studied cement samples except in CEM-III PPC 1 the specific activities of <sup>232</sup>Th were below the world average value. For all cement samples, the specific activity of <sup>40</sup>K was below the world average value. As can be seen in Table 2 the specific activity of <sup>226</sup>Ra in some brands of cement were higher than the world average value of 32 Bq kg<sup>-1</sup>.

Table 3 shows the mean specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in Bq kg<sup>-1</sup>, for the raw material samples, clinker, gypsum, dolomite and limestone collected from a cement factory.

**Table 3:** Mean specific activities with uncertainty of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K for the raw materials used in cement samples.

Type of raw material	<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> )
*Fly ash-1	73.9 ± 1.7	71.7 ± 4.9	357.4 ± 10.7
*Fly ash-2	63.4 ± 1.9	66.9 ± 3.4	189.9 ± 10.2
*Fly ash-3	144.7 ± 1.6	115.9 ± 1.5	422.2 ± 7.6
Clinker	27.5 ± 0.8	30.5 ± 1.3	64.9 ± 4.2
Gypsum	1.6 ± 0.2	$2.2 \pm 0.3$	21.4 ± 2.7
Limestone	11.4 ± 0.9	12.2 ± 0.9	38.7 ± 5.1
Dolomite	2.8 ± 0.5	$0.6 \pm 0.0$	1.62 ± 0.1

<sup>\*</sup>Fly ash-1 (fly ash emitted from burnt Indonesian coal)

The specific activities of radionuclides were significantly higher in fly ash than the other raw materials. The specific activities of <sup>226</sup>Ra and <sup>232</sup>Th in raw materials other than fly ash were lower than the world average of 50 Bq kg<sup>-1</sup> for building materials (UNSCEAR, 1993). It was observed that for all raw materials the specific activity of <sup>40</sup>K was less than the world average value of 500 Bq kg<sup>-1</sup> (UNSCEAR, 1993).

<sup>\*</sup>Fly ash-2 (fly ash emitted from burnt mixture of Indonesian and South-African coal)

<sup>\*</sup>Fly ash-3 (fly ash emitted from burnt mixture of South-African and Russian coal)

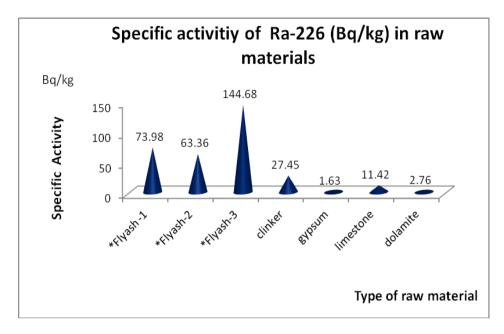


Figure 2: Mean Specific Activities of <sup>226</sup>Ra in raw material samples

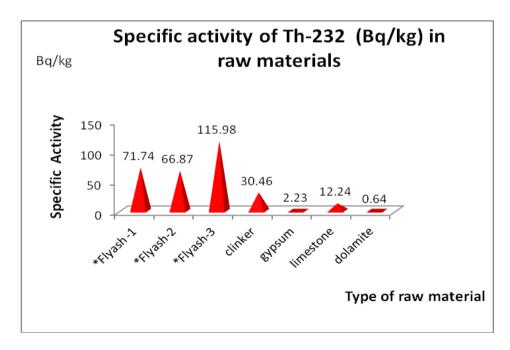


Figure 3: Mean Specific Activities of <sup>232</sup>Th in raw material samples

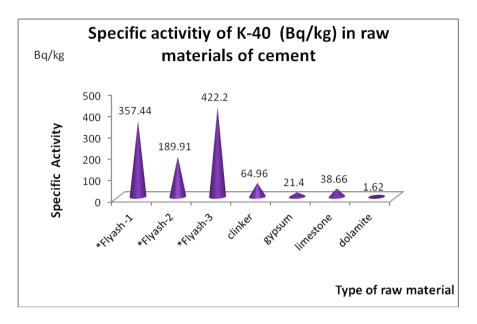


Figure 4: Mean Specific Activities of <sup>40</sup>K in raw material samples

According to the Table 3 and Figures 2, 3 and 4 the highest concentrations of radionuclides was observed in fly ash samples. The concentrations of radionuclides were very low in gypsum and dolomite samples.

The specific activities of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in Bq kg<sup>-1</sup> obtained for some kinds of raw materials in some countries are shown in Table 4.

**Table 4:** Comparison of specific activities of the radionuclides in raw materials considered for the present study with those from other countries.

Country	No. of sampl es	Raw material	<sup>226</sup> Ra Bq kg <sup>-</sup>	<sup>232</sup> Th Bq kg <sup>-</sup>	40K Bq kg- 1	Reference
Denmark	7	gypsum	10	07	n.m	(UNSCEA
					•	R, 1982)
	10	fly ash	150	90	n.m	(UNSCEA
						R, 1982)
	13	clinker	66	55	n.m	(UNSCEA
						R, 1982)
Italy	n.m.	limeston	11	2	22	(Rizzo et
		e				al., 2001)
	n.m.	gypsum	6.0	2.0	32	(Rizzo et
						al., 2001)

		1			1	/I INIC OF :
Germany	23	gypsum	<	<11	n.m	(UNSCEA
			19		•	R, 1982)
	28	fly ash	211	130	n.m.	(UNSCEA
						R, 1982)
Poland	106	fly ash	63-	33-	n.m.	(UNSCEA
			610	320		R, 1982)
United	73	gypsum	22	07	n.m.	(UNSCEA
kingdom						R, 1982)
Banglade	n.m.	limeston	68	107	1660	(Alam et
sh		e				al., 1999)
		gypsum	255	21.4	121	Chowdhu
						ry et al.,
Egypt	n.m.	limeston	20.4	4.4	19.3	(Ahamed
		e				and
						Hussein,
						1998)
			31.7	55	90	,
		gypsum	31.7	33	89	(E1-
D =:1		1:	04.2	7.0	005	Taher,
Brazil	n.m.	limeston	24.3	7.0	205	(Malanka
		е	6.0	1	154	et al.,
		gypsum	6.3	n.d.	154	(Malanka
0 1:		1:	00.6	40.0		et al.,
Saudi	n.m.	limeston	28.6	49.2	66	(E1-
Arabia		e	22.2	47.0	0.0	Taher,
		gypsum	33.3	47.2	88	(E1-
7D 1	100	T · .	16.5	7.7	00.1	Taher,
Turkey	128	Limesto	16.5	7.7	88.1	(Turhan,
		ne	000	1.177	1.00	2008)
		fly ash	232.	117.	466.	(Turhan,
			3	1	2	2008)
		Gypsum	10.8	3.6	44.5	(Turhan,
			0.6.5	0:5	200	2008)
Present	30	fly ash	94.0	84.8	323.	
study		Gypsum	1.63	2.23	21.4	
		Limesto	11.4	12.2	38.6	
		Clinker	27.4	30.4	64.9	
		Dolomit	2.76	0.64	1.62	

 $(n.m-not\ calculated;\ n.d.-not\ detected)$ 

It can be seen from Table 4, that the radioactivity in raw materials used for manufacturing cement varies from one country to another.

The values given in this table were not the representative values for the countries mentioned, but they were typical for the regions where samples were collected. Table 4 also shows that specific activity of <sup>226</sup>Ra and <sup>232</sup>Th in fly ash considered in the present study is lower than those of other countries.

The mean values of specific activities of <sup>226</sup>Ra,<sup>232</sup>Th and <sup>40</sup>K calculated for cement samples for the present study and the corresponding values determined by other countries are shown in Table 5.

**Table 5:** Comparison of specific activities (Bq kg<sup>-1</sup>) of radionuclides in Sri Lankan made cement samples with the published data of other countries.

	Other Cot		1		T
Country	No.of sample s	<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
Australia	7	51.5	48.1	114.7	(Bereka and
					Mathew,
Austria	18	26.7	14.2	210.0	(Sorantin
					and Steger,
Algeria	12	41.0	27.0	422.0	(Amarani
					and Tahtat,
Bangladesh	18	62.3	59.4	328.9	(Chowdhury
					et al., 1998)
Brazil	1	61.7	58.5	564.0	(Malanka et
					al., 1993)
China	46	56.5	36.5	173.2	(Xinwei.,
					2005)
Egypt	85	78.0	33.3	37.0	(El Afiafi,
					2006)
Finland	11	40.0	20.0	251.0	(Mustonen,
					1984).
Ghana	50	35.94	25.44	233.0	(Kpeglo,
					2011)
Greece	20	62.8	23.8	284.1	(Papastefano
					u, 2005)
India	1	37.0	24.1	432.2	(Kumar,
					1999)
Italy	7	38.0	22.0	218.0	(Rizzo et al.,
					2001)

Japan	16	35.8	20.7	139.4	(Suzuki et
					al., 2000)
Netherlands	6	27.0	19.0	230.0	(Ackers et
					al., 1985)
Pakistan	25	26.7	28.6	272.9	(Khan and
					Khan, 2001)
Tunisia	2	21.5	10.1	175.5	(Hizem et al.,
					2005)
Turkey	145	40.0	28.0	248.3	(Turhan,
					2008).
Sri Lanka	65	38.5	31.8	165.0	

According to Table 5 the mean specific activity values for cement samples taken from different countries shows considerable variation. The reason for this is the type and different proportions of raw materials used in cement manufacturing.

#### Assessment of radiation hazard

The radiation hazard caused by cement used in Sri Lanka was determined by the radiological parameter, the annual effective dose rate (A.E.D). Table 6 shows the calculated values of this parameter and the absorbed dose rate for the different brands of cement studied.

**Table 6:** Annual effective dose rates and absorbed dose rates for cement samples.

Type of cement	Absorbed dose rate in	A.E.D.
	air (nGy h-1)	(mSv y-1)
CEM-1 OPC 1	45.01	0.22
CEM-II PLC	42.23	0.21
CEM-III PPC 1	103.85	0.51
CEM-IV OPC 2	60.57	0.30
CEM-V OPC 3	53.03	0.26
CEM-VI PPC 2	58.93	0.29
CEM-VII OPC 4	60.27	0.31

CEM-VIII Imported	42.23	0.21
OPC1		
CEM-IX Imported	59.09	0.29
OPC 2		
CEM-X Imported OPC	30.02	0.15
3		
CEM-XI Imported PPC	79.42	0.39
1		

The Annual effective dose inside the reference room considered was calculated by using Equation 7. The calculated values for different cement samples are shown in Table 6. The estimated average annual effective dose ranged from 0.15 to 0.51 mSv y<sup>-1</sup> and less than the recommended maximum permissible public dose 1.0 mSv y<sup>-1</sup> (International Basic Safety, IAEA, 1996).

### Conclusion

From the results it is evident that there are considerable variations in the specific activities of radionuclides <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in raw materials. The highest concentrations of these radionuclides were observed in fly ash.

The concentrations of radionuclides in the samples of different brands of cement were not same. The highest values of <sup>226</sup>Ra and <sup>232</sup>Th were observed in CEM-III PPC-1 cement where 25% fly ash was added, whereas the minimum values of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were in CEM -X imported OPC 3 cement. The highest concentration of <sup>40</sup>K was observed in CEM-V OPC 3 cement. These variations are due to the varying amounts of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the raw materials and different proportions of the raw materials used in manufacturing different brands.

It was observed that the annual effective dose values for all the studied samples were lower than the recommended maximum permissible public dose of  $1.0~\text{mSv}~\text{y}^{-1}$ . All the cement samples studied did not show any significant radiation hazard and could be considered safe for construction work.

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### References

- Ackers, J. G., DenBoer, J. F., Delong, P. & Wolschrijn, R. A. (1985). Radioactivity and radon exhalation rates of building materials in the Netherlands. *Science Total Environment* 45:151-156. https://doi.org/10.1016/0048-9697(85)90215-3
- Ahamed, N. & Hussein, A. (1998). Natural radioactivity in Gordanian soil and building materials and the associated radiation hazards. *Journal of Environmental Radioactivity* 39: 9 –22. https://doi.org/10.1016/S0265-931X(97)00046-5
- Alam, M. N., Chowdhury, M. I., Kamal, M., Ghose, S., Islam, M. N., Mustata, M. N., Miah, M. H. & Ansary, M. M. (1999). The <sup>226</sup>Ra,<sup>232</sup>Th and <sup>40</sup>K activities in beach sand minerals and beach soil of Cox-s Bazar, Bangladesh. *Journal of Environmental Radioactivity* 46:243-250. https://doi.org/10.1016/S0265-931X(98)00143-X
- Amarani, D. & Tahtat, M. (2001). Natural radioactivity in Algerian building materials. *Applied Radiation Isotopes* 54:687-689. https://doi.org/10.1016/S0969-8043(00)00304-3
- Bereka, J. & Mathew, P. J. (1985). Natural radioactivity of Australian building materials, Industrial wastes and by-products. *Health Physics* 48:87-95. DOI: 10.1097/00004032-198501000-00007.
- Chowdhury, L. M., Alam, M. N. & Ahmed, A. K. S. (1998). Concentration of radionuclides in building and ceramic materials of Bangladesh and evaluation of radiation hazard. *Journal of Radioanlytical Nuclear Chemistry* 231(1-2):117-122. https://doi.org/10.1007/BF02388016

- Curie, L. A. (1968). Limits of qualitative determination and quantitative determination. *Analytical Chemistry 40* (3):586-593.
- El Afiafi, E. M, Hilal, M. A, Khalifa, S. M. & Aly, H. F. (2006). Evaluation of U, Th and K and emanated radon in some NORM and TENORM samples. *Radiation Measurements* 41:627-633. https://doi.org/10.1016/j.radmeas.2005.09.014
- El-Taher, A. (2010). Gamma spectroscopic analysis and associated adiation hazards of building materials used in Egypt. *Radiation Protection Dosimetry 138* (2):166-173. https://doi.org/10.1093/rpd/ncp205
- El-Taher, A. (2012). Assessment of natural radioactivity levels and radiation hazards for building materials used in Qassim area, Saudi Arabia. Rom. *Journal of Physics*; 57:726-735.
- Hizem, N., Fredj, A. B. & Ghedira, I. (2005). Determination of natural radioactivity in building materials used in Tunisian dwellings by gamma ray spectrometry. *Radiation Protection Dosimitry* 114: 533-537. https://doi.org/10.1093/rpd/nch489
- Interim report on potential consequences for the Dutch population of using waste substances with an enhanced concentration of radionuclides as building materials for houses by Health Council of the Netherlands (1985). The Hague, The Netherlands.
- International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources-Schedule II, International Atomic Energy Agency (IAEA) (1996).
- Khan, K. & Khan, H. M. (2001). Natural gamma emiting radionuclides in Pakistani Portland cement. *Applied Radiation Isotopes* 54:861-865. https://doi.org/10.1016/S0969-8043(00)00327-4
- . Kpeglo, D. O., Lawluvi, H., Faanu, A., Awudu, A. R., Deatanyah, P., Wotorchi, S. G., Arwui, C. C., Emi- Reynolds, G. & Darko, E. O. (2011). Natural radioactivity and its associated hazard in

- Ghanaian cement. *Journal of Environment and Earth Sciences* 3(2):160-166.
- Kumar, V., Ramachandran, T. V. & Prasad, R. (1999). Natural radioactivity of Indian building materials and by-products. *Applied Radiation Isotopes* 51:93-96. https://doi.org/10.1016/S0969-8043(98)00154-7
- Malanka, A., Pessina, V., Dallara, G., Luce, C. N. & Gaidol, L. (1993). Natural radioactivity in building materials from the Brazilian state of Espirito Santo. *Applied Radiation Isotopes* 46:1387-1392. https://doi.org/10.1016/0969-8043(95)00223-Z
- Mustonen, R. (1984). Natural radioactivity and radon exhalation rate from finish building materials. *Health Physics* 46:1195-1203. DOI: 10.1097/00004032-198406000-00003
- Papastefanou, C., Stoulos, S. & Manolopoulou, M. (2005). The radioactivity of building materials. *Journal of Radioanalytical Nuclear Chemistry* 266(3):367-372.
- Report to the General Assembly of the United Nations. Annex C. Ionizing Radiation, Sources and Biological Effects. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). (1982). United Nations Sales Publications (NY. USA).
- Report to the General Assembly of the United Nations, Annex C, Ionizing Radiation, Sources and Biological Effects. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). (1993). United Nations Sales Publications. (NY, USA).
- Report to the General Assembly of the United Nations, Annex B, Exposures of the public and workers from various sources of radiation. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). (2008). United Nations Sales Publications. (NY, USA).
- Rizzo, S., Brai, M., Basile, S., Bellia, S. & Hauser, S. (2001). Gamma activity and geochemical features of building materials:

- estimation of gamma dose rate and indoor radon levels in Sicily. *Applied Radiation Isotopes* 55:259-265. https://doi.org/10.1016/S0969-8043(00)00384-5
- Sorantin, P. & Steger, F. (1984). Natural radioactivity of building materials in Austria. *Radiation Protection Dosimetry* 7:59-61. https://doi.org/10.1093/oxfordjournals.rpd.a082963
- Suzuki, A., Lida, T., Moriizumi, J. & Sakuma, Y. (2000). The effects of different types of concrete on population doses. *Radiation Protection Dosimetry* 90:437-443. https://doi.org/10.1093/oxfordjournals.rpd.a033171
- Topping, J. (1972). Errors of Observations and Their Treatment. New York, Halsted Press.
- Turhan, S. (2008). Assessment of the natural radioactivity and radiological hazard in Turkish cement and its raw materials. *Journal of Environmental Radioactivity* 99:404--414 https://doi.org/10.1016/j.jenvrad.2007.11.001
- Xinwei, L. (2005). Radioactive analyses of cement and its products collected from Shannxi China. *Health Physics* 88:84-86. https://doi:10.1097/01.HP.0000142498.08451.c8