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RESEARCH ARTICLE

PRELIMINARY INVESTIGATION OF NATURALLY OCCURRING RADIONUCLIDE IN SOME FIVE LOCALLY MANUFACTURED CEMENT TYPES COMMONLY USED IN KASHMIR VALLEY AS BUILDING MATERIAL

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ABSTRACT

The present study was aimed at the determination of specific activity of locally manufactured and commercially available five cement types used as building material in Kashmir valley, India by using a NaI (Tl) gamma ray spectrometer. The study envisages that the mean values of specific activity concentrations in the different analyzed cement samples were found to vary from 44.4±0.81 to 51.02±2.3 $Bqkg^{-1}$ for ^{226}Ra; 20.11±3.60 to 36.91±2.9 $Bqkg^{-1}$ for ^{232}Th and 25.29±1.42 to 55.78±2.81 $Bqkg^{-1}$for ^{40}K, the mean value specific activity for ^{226}Ra in all the investigated cement brand were above the world average of 32 $Bqkg^{-1}$. The radiation hazard indices determined are well below the limits and all the analysed cement brands used in Kashmir valley meet the safety requirement and do not pose any radiological hazard to human health.

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INTRODUCTION

One inescapable feature of life on the earth is exposure to the ionizing radiations. Ionizing radiation of the environment is the most ubiquitous form of exposure therefore determination of health risk of background radiation is of great importance in health physics (UNSCEAR, 2010). The main sources of the background radiation are cosmic and terrestrial radiations. Cosmic radiations include energetic particles produced by spallation reactions in the outer space of the atmosphere which penetrate into the earth's atmosphere and contribute as one of the main sources of background radiation. Interaction of these particles with atmosphere molecules may produce cosmogenic radionucleides. Long half lived radionuclides which were formed at the time of formation universe have formed terrestrial radionuclides which exist in air, soil, rocks, water and building materials. The terrestrial predominant radionuclides, with respect to absorbed dose in human, are ^{232}Th and ^{238}U are head of decay series in which radionucleides of the chain contribute to human exposure and increase total radiation on earth (UNSCEAR, 2000). Studies conducted on natural radioactivity have shown that the presence of potassium (^{40}K) and other daughter radionucleides from Thorium (^{232}Th) and Uranium (^{238}U) decay series in various components in the environment result in radiation exposure of the global population (El- Tahir, 2012). The primordial radionucleides are predominant in almost all raw and produced materials widely used in the building industries including; cement, brick, sand, tile, limestone, gypsum and those derived from rocks and soil (El- Tahir, 2012; White, 1981). Natural radiations in building materials related to external and internal

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exposure. The external exposure comes from direct terrestrial gamma-rays and internal exposure from inhalation of inert radioactive inert gas Radon (^{222}Rn, a daughter product of ^{226}Ra). The presence of these natural radionucleides in building raw materials depend on geological, geographical conditions and geochemical characteristics of the materials themselves.

Building materials are commonly originated from soils and rocks and also contain the naturally occurring radioactive materials. In order to assesses the radiological risk linking to standards on natural activity in building materials, it is important to study the levels of radiation emitted by them. Cement is commonly used in construction of building both in urban and rural areas in Kashmir valley. Due to its high production rate and widely used by the population deserves special attention. An attempt has been made through this study to report the activity concentrations of natural radionucleides such as ^{226}Ra, ^{232}Th, and ^{40}K in some locally manufactured and commercially available cement samples and the associated absorbed dose rate in the ambient air due to these cements.

MATERIALS AND METHODS

Sampling and sample preparation

A total five main types of cement commercially available (ten samples from each cement type) used in building construction in Kashmir valley were collected from different construction sites and shops. These samples were used without any process of homogenization since they are in powder form. The samples were dried in an oven at 125^{0}C for 24 hours to ensure the moisture is completely removed. The samples were weighted, packed in cylindrical geometry, labeled and hermetically sealed in air tight plastic containers. The sealed samples were then stored for four weeks in a safe place to enable them to attain a state of secular equilibrium, where the rate of progeny becomes equal to that of the parent (Ra^{226} and Th^{232}) (Akkurt *et al.,* 2010).

Radiometric analysis

The radioactivity of Ra^{226}, Th^{232}, and K^{40} in the samples was determined using the gamma-ray spectrometer consisting of a NaI (Tl) detector (crystal size 40.0 mm x 60.0 mm) connected to 1024 channel multichannel analyser (MCA). Before measurement, the system is calibrated using Cs^{137} and Co^{60} radioactive sources produce γ-ray energies of 662 KeV, 1173 KeV and 1332 KeV, respectively.

The spectrum was analysed by Leybold Cassy Lab Multi-Channel Analyser model Pocket- CASSY 559901 (Germany made). The activity of K^{40} was measured directly from 1460.7 (10.7%) KeV photo peak of the gamma ray spectrum. To determine the activity concentration of Ra^{226}, the average value of gamma ray lines 295.1 (19.2%) and 351.9 (37.1%) KeV from Pb^{214} to 609.3 (46.1%) and 1764.5 (15.9%) KeV gamma rays from Bi^{214} are used. The activity concentration of Th^{232} was determined using the average value of gamma rays peaks 238.6 (43.6%) KeV from Pb^{212}, 338. 4 (12%), 911.1 (29%) and 968.9 (17.4) KeV from Ac^{228}, 583.1 (86%) and 2614 KeV from Tl^{208}. Each sample was examined for 7200 seconds. The analysis of results is performed using Microsoft Excel software. The activities for the natural radionucleides were calculated using the following relation [Beretka and Mathew, 2003]:

$$A(Bqkg^{-1}) = \frac{N}{\gamma \times \varepsilon \times t \times m} \quad (1)$$

Where A is the activity of the radionuclide in $Bqkg^{-1}$, N represents the counts under the most prominent photo peak, calculated from subtracting the respective count rate from the back ground spectrum obtained for the same counting time, ε is the detector efficiency of the specific gamma-ray, γ is the absolute transition probability of gamma decay, t is the counting time(s) which is 7200 seconds and m is the mass of the sample.

Assessment of the Radiological Hazards of the materials

The common used radiological hazard index Ra_{eq} is called the radium equivalent activity. It is defined as the weighted sum of activities of the Ra^{226}, Th^{232}, and K^{40} radionuclides based on the assumption that 370 $Bq.Kg^{-1}$ of Ra^{226}, 259 $Bq.Kg^{-1}$ of Th^{232} and 4810 $Bq.Kg^{-1}$ of K^{40} produce the same gamma ray dose constant. The index is calculated from the following relation suggested by Beretka and Mathew: (Beretka, 2003)

$$\mathrm{Ra_{eq}} = (\mathrm{A_{Th}} \times 1.43) + \mathrm{A_{Ra}} + (\mathrm{A_K} \times 0.077) \quad (2)$$

Where A_{Ra},A_{Th}, and A_K are the specific activities ($BqKg^{-1}$) of Ra^{226}, Th^{232}, and K^{40} respectively.

Due to more than one radionuclide contribution to the dose; it is practical to present to present investigation levels in the form of an activity index. The European Commission has proposed in their guidance document the induction of an activity concentration index used to asses safety requirement for building materials. The radiation hazard index used to estimate the level of γ- radiation hazard associated with natural radionucleides is called the representative index I_γ, is defined by the following relation: (Miah *et al.,* 1998)

$$I_\gamma = \frac{1}{200Bq.Kg^{-1}} A_{Ra} + \frac{1}{300Bq.Kg^{-1}} A_{Th} + \frac{1}{3000Bq.Kg^{-1}} A_K \quad (3)$$

Where A_{Ra},A_{Th}, and A_K have the same meaning as in the equation (2).

Values of index $I_\gamma \leq 0.5$ corresponds to a dose rate criteria of 0.3 $mSv.y^{-1}$, whereas $I_\gamma$corresponds to a criteria of 1.0 $mSv.y^{-1}$(European Commission, 1999). Thus the material with $I_\gamma > 6$ should be avoided to use as building material since these values correspond to the dose rates higher than 1.0 $mSv.y^{-1}$which is highest than recommended values (Miah *et al.,* 1998; European Commission, 1999). Due to radon inhalation originated from building materials (Dragovic *et al.,* 2006). The $I_\alpha$was determined using the following formula:

$$I_\alpha = \frac{A_{Ra}}{200(Bq.Kg^{-1})} \quad (4)$$

Where A_{Ra} is the specific activities concentration of Ra^{226} assumed in equilibrium with ^{238} U. The recommended exemption and upper limit of ^{226}Ra activity concentration in building materials are 100 and 200 $Bq.Kg^{-1}$, respectively as suggested by many countries in the world [Dragovic *et al*., 2006]. These considerations reflected in the I_α. The recommended upper limit activity concentration of ^{226}Ra is 200 $Bq.Kg^{-1}$, for which $I_\alpha = 1$. A direct connection between radioactivity concentrations of natural radionucleides and their exposure rate is known as the absorbed dose in the air at 1 meter above the ground surface. The mean activity concentrations of Ra^{226} (of the U^{238} series), Th^{232}, and K^{40} ($Bqkg^{-1}$) in the lignite and soil samples are used to calculate the absorbed dose rate given using the following formula provided by UNSCEAR (UNSCEAR, 2000) and European Commission (European Commission, 1999). UNSCEAR and the European Commissions have provided the dose conversion coefficients for the standard room centers.

$$D(nGyh^{-1}) = 0.462A_{Ra} + 0.6A_{Th} + 0.042A_K \quad (4)$$

Where D is the absorbed dose rate in $nGyh^{-1}$, A_{Ra}, A_{Th}, and A_K are the activity concentration of Ra^{226} (U^{238}), Th^{232} and K^{40}, respectively. The dose coefficients in the units of $nGyh^{-1}$ per $Bqkg^{-1}$ are taken from the UNSCEAR (2000) report.

The absorbed dose rate in air at 1 meter above the ground surface does not directly provide the radiological risk to which an individual is exposed (Dragovic *et al.,* 2009). The absorbed dose can considered in terms of the Annual Effective Dose Equivalent (E_T) from the indoor terrestrial gamma radiation which is converted from absorbed dose by taking into account two factors, namely the conversion coefficient from the absorbed dose in air to effective dose and the occupancy factor. The Annual Effective Dose Equivalent can be estimated using the following formula (UNSCEAR, 2000):

$$E_T(\mu Sv.y^{-1}) = D(nhh^{-1}) \times 24h \times 365.25days \times 0.8 \times 0.7(SvGy^{-1}) \times 10^{-6} \quad (5)$$

To limit the radiation exposure attributable to natural radionucleides in the samples to permissible dose equivalent limit of $1\, mSv.y^{-1}$. External hazard index due emitted gamma rays of the samples is calculated and examined according to the following relation: (Krieger, 1987)

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad (6)$$

In addition to H_{ex}, radon and its short lived products are also hazardous to the respiratory organs. The H_{in} due to internal exposure to radon and its daughter products is quantified by H_{in} , which is given by the following equation as (UNSCEAR, 2000):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{2\ 59} + \frac{A_K}{4810} \leq 1 \quad (7)$$

Where A_{Ra},A_{Th} and A_K are the concentration in $Bqkg^{-1}$ of Ra^{226}, Th^{232} and K^{40} respectively.

RESULTS AND DISCUSSION

A total of 50 samples of locally manufactured and commercially available five cement types commonly used as building material in Kashmir Valley, India, were collected, their radioactivity content was analyzed with special emphasis in the radium content of the samples and associated hazards. The activity concentration of ^{226}Ra, ^{232}Th, ^{40}K and Ra_{eq} in different of cement types used as building material have been calculated by using NaI (Tl) gamma ray spectrometer and presented in the Table 1.

Table 1. The mean value of activity concentration of the ^{226Ra} and ^{232}Th series and ^{40}K of the studied cement samples

Cement Trade Name	^{226}Ra	^{232}Th	^{40}K	^{Ra}eq
TCI Max Grade 53	47.29±4.51	20.11±3.60	29.88±4.56	76.28±4.22
TCI Max Grade 43	44.40±0.81	21.59±1.41	25.29±1.42	75.48±1.21
Khyber Grade 53	49.53±1.4	32.43±0.98	56.34±2.43	96.33±1.6
Saifco Grade -53	43.21±.2.11	31.78±1.31	55.78±2.81	88.08±2.07
Saifco Grade-43	51.02±2.3	36.91±2.9	42.38±1.32	104.28±2.17

The comparison of the mean values of ^{226}Ra, ^{232}Th, ^{40}K and Ra_{eq} of locally manufactured commercially available cement types of Kashmir Valley with the data published by other countries are presented in the table 2. The radiation hazard indices in the investigated cement samples have been estimated and reported in the table 3. The mean values of specific activity concentrations in the analyzed samples were found to vary from 44.4±0.81 to 51.02±2.3 $Bqkg^{-1}$ for ^{226}Ra; 20.11±3.60 to 36.91±2.9 $Bqkg^{-1}$ for ^{232}Th and 25.29±1.42 to 55.78±2.81 $Bqkg^{-1}$for ^{40}K.

Table 2. The mean value of concentrations of the natural radionucleides in cement ($BqKg^{-1}$) reported for different parts of world

Country	Specific Activity ($Bqkg^{-1}$)			Ra_{eq}($Bqkg^{-1}$)	Reference
	^{226}Ra	^{232}Th	^{40}K		
Algeria	41	27	422	112	(Amrani and Tahtat)
Cameroon	24	16.6-47.6	12.5-32.5	53.6-105.9	(Ndontchuenge *et al*., 2013)
Egypt	78.00	33.30	37.00	151.00	(El Afifi *et al*., 2006)
Brazil	61.70	58.50	564.00	188.80	(Manaca *et al*., 1999)
Ghana	35.94	25.44	251.00	90.12	(Kpeglo *et al*., 2011)
Italy	38.00	22.00	218.00	92.00	(Rizzo *et al*., 2001)
Nigeria	43.80	21.50	71.7	80.10	(Ademola *et al*., 2008)
Netherland	27.00	19.00	230.00	71.90	(Ackers *et al*., 1985)
Turkey	50	40	324	62-312	(Damla *et al*., 2010)
World Average	32	45	420	370	(Ndontchuenge *et al*., 2013)
TCI Max Grade 53	47.29±4.51	20.11±3.60	29.88±4.56	76.28±4.22	Current Study
TCI Max Grade 43	44.40±0.81	21.59±1.41	25.29±1.42	75.48±1.21	Current Study
Khyber Grade 53	49.53±1.4	32.43±0.98	56.34±2.43	96.33±1.6	Current Study
Saifco Grade 53	43.21±2.11	31.78±1.31	55.78±2.81	88.08±2.07	Current Study
Saifco Grade 43	51.02±2.3	36.91±2.9	42.38±1.32	104.28±2.17	Current Study

The lowest specific activity values were observed in TCI Max Grade 43, whereas the highest values are seen in Saifco Grade 43 cement.

Table 3. The average values of radiation hazard indices of the investigated samples

Cement Trade Name	D (nGy/h)	E_T (μSv/y)	H_{ex}	H_{in}	I_γ	I_α
TCI Max Grade 53	35.25	0.17	0.21	0.34	0.31	0.24
TCI Max Grade 43	34.81	0.17	0.21	0.33	0.30	0.22
Khyber Grade 53	44.83	0.22	0.27	0.40	0.37	0.25
Saifco Grade 53	41.51	0.20	0.25	0.37	0.33	0.22
Saifco Grade 43	47.64	0.23	0.29	0.43	0.39	0.26
World Average (Ndontchuenge *et al*., 2013)	69	0.410	< 1	< 1	1.0	0.5

The average specific activity values of ^{226}Ra, ^{232}Th, and ^{40}K determined in the cement samples varied from one brand to the another brand. This variation in the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the investigated cement types used as building material in Kashmir Valley may depend upon the Uranium, Thorium and Potassium content under the earth's crust from where the raw materials for a particular brand of cement were obtained. As per United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR), the world's mean value of ^{226}Ra, ^{232}Th, ^{40}K and Ra_{eq} specific activity were 32, 45, 420 and 370 $Bqkg^{-1}$ respectively (UNSCEAR, 2000).The mean activity concentration of ^{226}Ra in this study were found to be higher in the investigated cement brand than the world average. The radium equivalent activity calculated from the studied cement types is lower than the recommended maximum value of 370 $Bqkg^{-1}$, which corresponds to an annual effective dose of $1mSv$. This envisages that the investigated cement types are within the recommended safety limit when used as building construction materials. The radiation Hazard indices H_{ex} & H_{in}, Dose rate in air (nGy/h), Annual effective dose equivalent (μSv/y), and activity concentration indices (I_γ and I_α) obtained for different cement brands are presented in the table 3.The calculated H_{ex} & H_{in} indices in all the cement brands are less than unity and the dose rate in air and all other hazard indices are below the world average.

Conclusion

A preliminary investigation on naturally occurring radionuclides in locally manufactured and commercially available five cement brands was carried out by using NaI(Tl) gamma ray spectrometer. The study envisages that the mean value of specific activity for ^{232}Th and ^{40}K in the investigated cement brands are much below the permissible global value by UNSCEAR whereas the mean value specific activity for ^{226}Ra in all the investigated cement brand were above the world average of 32 $Bqkg^{-1}$. The radium equivalent activity, dose rate, annual effective dose, external hazard index, internal hazard index and activity concentration indices were calculated to determine the radiological implication implying the use of these cement brands as building materials. The entire analysed cement brands used in Kashmir valley met the safety requirement.

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