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Estimating radiation dose from building materials

M. Gupta* and R.P. Chauhan

Department of Physics, National Institute of Technology, Kurukshetra-136119, India

Background: Natural radioactivity in materials under certain conditions can reach the hazardous radiological levels. So, it becomes necessary to study the natural radioactivity in different materials to assess the dose for the population in order to know the health risks and to have a baseline for future changes in the environmental radioactivity due to human activities. ***Materials and Methods:*** The present study deals with the measurement of radioactivity using "γ-ray spectrometry" from naturally occurring radionuclides in the soil, stone and sand samples used as building materials in North-Eastern Haryana state of India. The places are in the vicinity of Shivalik range of Himalayas. ***Results:*** The activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K varied from 18±1.5 to 156±6Bqkg^{-1}, 23±1 to 300±5Bqkg^{-1} and 32±0.5 to 1705±14 Bqkg^{-1} respectively in various samples. The absorbed dose rate in soil, sand and stone samples is investigated at 1 m above ground level. Ra equivalents, Internal and external hazard indices have also been calculated. ***Conclusion:*** The natural radioactivity levels measured in the samples under present study are below the recommended limits except for black stone (SB) and red stone (SR). However, these samples satisfy the universal standards. **Iran. J. Radiat. Res., 2011; 9(3): 187-194**

Keywords: *Absorbed dose, natural radioactivity, γ-ray spectrometry, Radium, thorium, potassium.*

INTRODUCTION

The knowledge of natural radionuclides concentration levels and their distribution in the environment have become a focus of much attention in assessing the human exposure. The high geochemical mobility of radionuclides in the environment allows them to move easily and to contaminate much of the environment with which humans come in contact. The measurement of natural radioactivity due to gamma rays from the dose rate (above recommended limits) is needed to implement the precautionary measures. The growing world wide interest in natural radiation exposure has lead to extensive surveys in many countries. Human beings are always exposed to ionizing radiation from natural sources. Natural radioactivity is wide spread in the earth's environment and it exists in various geological formations in soils, rocks, plants, water and air [1]. To evaluate the terrestrial gamma dose rate for outdoor occupation, it is very important to estimate the natural radioactivity levels in soils. The natural radioactivity in soil samples is usually determined from the ^{226}Ra, ^{232}Th and ^{40}K contents. Since 98.5% of the radiological effects of the uranium series are produced by radium and its daughter products, the contribution from the ^{238}U and the other ^{226}Ra precursors are normally ignored [2]. Most of the dose rate of natural radioactivity is due to the daughter products of the radioactive decay chains of ^{238}U (55.8%) and ^{232}Th (14%), along with the ^{40}K (13.8%) [3], which are ubiquitous in the earth crust, soil, stones, plants, water and air and have become important tracers of chemical, biological and physical processes occurring in the atmosphere. The contribution of ^{235}U to radiation exposure is negligible, because the ratio ^{235}U/^{238}U is less than 1%. Exhalation of radon is associated with the presence of radium (^{226}Ra) and its ultimate precursor of uranium (^{238}U). Radon, thoron and their progenies present in the environment contribute the maximum of the natural radiation dose to the occupational workers and general public [4]. The United States Environmental Protection Agency (US-EPA) has reported that inhalation of radon is the

Corresponding author:
Dr. Monika Gupta,
Department of Physics, National Institute of Technology, Kurukshetra-136119, India.
Fax: *+911 744 238050*
E-mail: *monika.kkr@gmail.com*

second highest killer causing cancer [5]. The radiological impact caused by nuclides is due to radiation exposure (gamma rays) of the body and irradiation of the lung tissues from inhalation of radon and its progeny [6]. Radon and its daughters are the most important radionuclide present in the ambient air as well as in the indoor atmosphere [7]. The assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population [8].

The objective of the study is focused on determining the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and their dose contribution. Knowledge of radioactivity present in the materials enables one to assess any possible radiological hazard to mankind by the use of such materials for building construction. The obtained results will serve as base line data for radioactivity level in this environment.

Geography of the study area

The geographic location of the study areas belong to Haryana state of India, which are situated in India's northwest between 27°37' and 30°35' Northern latitude and 74°28' and 77°36' East longitude as shown in figure 1 (Map of the study area). The sediments of the state are derived from Shivalik Himalayas and occur in the form of alluvium. The state is situated towards the depressions of the rivers Ganges and Indus. It is a broad level plain standing nearly on the watershed between the basins of the two rivers. It is a vast ground of moist land. In the whole of the region except the flood plains of the Yamuna and the Ghaggar, the alluvium is of the old type containing sand, clay, silt and hard calcareous concentrations.

The Haryana state of India can be subdivided into two natural areas i.e. Sub-Himalayan terrain and the Indo-Gangatic planes. The plane is fertile and the height above the sea level is 700-900 ft. The slope is from north to south. The climate of the

Figure 1. Map of the study area.

area is continental, with extremes of heat in summer and markedly cold in winters. The maximum temperature is recorded in the month of May and June when it goes up to 46 °C. Mountain hills are Aravali hills in the southwest and shivalik hills in the north. Mines and minerals found in the state are lime stone; slate stone, dolomite, building stone, china clay and marble.

MATERIALS AND METHODS

Measurement of natural radioactivity levels by gamma spectrometry technique

In order to measure the natural radioactivity, the samples were collected from undisturbed sites at each location. After removing the stones and organic materials, samples were crushed into a fine powder by using mortar and pestle. Before measurement, the samples were sieved and dried at 110 °C for 24 hours to remove the moisture content. Each sample was packed and sealed in an airtight PVC container and kept for about 4-week period (>7 half-lives of ^{222}Rn and ^{224}Ra) to allow the radioactive equilibrium among the radon (^{222}Rn), thoron (^{220}Rn) and their short-lived decay products. Gamma transitions of 1461 keV for ^{40}K, 186 keV for ^{226}Ra, 295 and 352 keV for ^{214}Pb, 609, 1120 and 1764keV for ^{214}Bi, 338, 463, 911 and 968 keV for ^{228}Ac, 727 keV for ^{212}Bi, 238 keV for ^{212}Pb were used for the

laboratory measurement of activity concentration.

The detector involved in this study is one of the high-purity germanium (HPGe) detectors used at Inter-University Accelerator Centre, New Delhi to perform the measurement of radioactivity. The detector is a coaxial n-type HPGe detector (EG&G, ORTEC, Oak Ridge, USA).The detector having a resolution of 2.0 keV at 1332 keV and a relative efficiency of 20% was placed in 4 inch shield of lead bricks on all sides to reduce the background radiation from the materials and cosmic rays [9]. The output of the detector is analyzed using a 4K ADC system connected to PC, the spectrum is analyzed using the locally developed software "CANDLE (Collection and Analysis of Nuclear Data using Linux nEtwork)". For calibration of the low background counting system, a secondary standard was obtained, which was calibrated with the primary standard obtained from the International Atomic Energy Agency (IAEA). The efficiency for the system was determined using secondary standard source of uranium ore in the same geometry as available for the sample counting. Efficiency values are plotted against energy for particular geometry and are fitted by least squares method to an empirical relation that takes care of the nature of efficiency curve for the HPGe detector. For activity measurements the samples were counted for a period of 72,000s and the spectra are analyzed for the photopeak of uranium, thorium daughter products and ^{40}K. The activity concentration of ^{40}K (C_K) was measured directly by its own gamma ray of 1461 keV. As ^{226}Ra and ^{232}Th are not directly gamma emitters, their activity concentrations (C_{Ra} and C_{Th}) were measured through gamma rays of their decay products. Weighted averages of several decay products were used to estimate the activity concentrations of ^{226}Ra and ^{232}Th. The peaks corresponding to 1.46 MeV (^{40}K), 1.76MeV (^{214}Bi) and 2.614MeV (^{208}Tl) were considered for the evaluation of the activity levels of ^{40}K, ^{238}U series and ^{232}Th, respectively [10].

The net count rate under the most prominent photo peaks of radium and thorium daughter peaks are calculated from respective count rate after subtracting the background counts of the spectrum obtained for the same counting time. Then the activity of the radionuclide is calculated from the background subtracted area of prominent gamma ray energies. Gamma ray spectrum of stone sample is shown in figure 2. The activity of radium, thorium and potassium is calculated using the equation 1 where CPS is the net count rate per second, B.I. is the branching intensity and E_{ff} is the efficiency of the detector.

Estimation of Radium Equivalent Activity (Ra_{eq})

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in soil and stone samples is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium Equivalent Activity (Ra_{eq}) in Bqkg^{-1} to compare the specific activity of samples containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. It is calculated through the following expression [11]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \qquad (2)$$

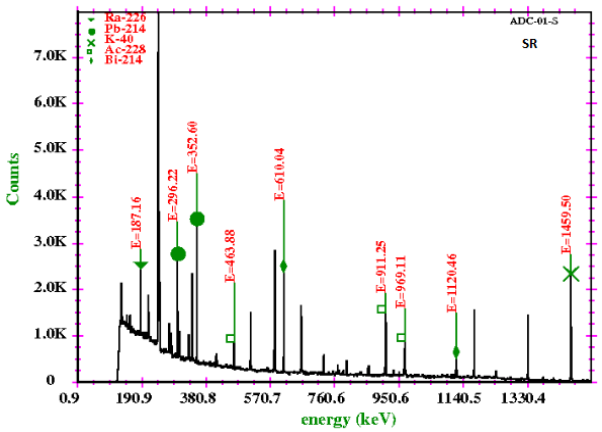

Figure 2 . Spectra of the Stone (SR) sample.
Ra-226(▼), Pb-214(●), K-40(X), Ac-228(□), Bi-214(♦)

$$\text{Activity(Bq)} = (CPS \times 100 \times 100)/(\text{B.I.} \times E_{ff}) \pm (CPS_{error} \times 100 \times 100)/(\text{B.I.} \times E_{ff}) \qquad (1)$$

where; C_{Ra}, C_{Th} and C_K are the concentrations in (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, in the samples respectively. While defining Ra_{eq} activity according to equation 2, it has been assumed that 370 $Bqkg^{-1}$ of ^{226}Ra or 259 $Bqkg^{-1}$ of ^{232}Th or 4810 $Bqkg^{-1}$ of ^{40}K produces the same gamma dose rate.

Estimation of absorbed and effective dose

The measured activity of ^{226}Ra, ^{232}Th and ^{40}K were converted into doses ($nGyh^{-1}Bq^{-1}kg^{-1}$) by applying the factors 0.462, 0.604 and 0.0417 for radium, thorium and potassium, respectively [12]. These factors were used to calculate the total absorbed gamma dose rate in air at 1m above the ground level using the equation 3.

Where; C_{Ra}, C_{Th} and C_K are the activity (Bq kg^{-1}) of radium, thorium and potassium in the samples respectively. To estimate annual effective doses, account must be taken of (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 $SvGy^{-1}$, which is used to convert the absorbed rate to annual effective dose with an outdoor occupancy of 20% and 80% for indoors [3]. The annual effective doses are determined as follows (equations 4 and 5).

The Internal Index

Several α indices have been proposed to assess the exposure level due to radon inhalation originating from building materials [13, 14]. The internal index is defined as follows:

$$I_\alpha = C_{Ra} / 200 Bqkg^{-1} \quad (6)$$

Where; C_{Ra} ($Bqkg^{-1}$) is the activity concentration of ^{226}Ra. The recommended exemption level and the recommended upper level for ^{226}Ra suggested by the International Commission on Radiological Protection [15] are 100 $Bq.kg^{-1}$ and 200 $Bq.kg^{-1}$, respectively.

The External Index

The gamma index (I_γ) is defined in order to examine the applicability of using materials in construction. For a typical material it is given by the following expression [13]:

$$I_\gamma = \sum_x C_x / A_x \leq 1 \quad (7)$$

Where C_x (Bq kg^{-1}) is the measured activity of each nuclide in the building material , A_x (Bq kg^{-1}) is the activity concentration of each nuclide in the material, and it is assumed to produce the same gamma dose rate, i.e. 300, 200 and 3000 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively [13].

RESULTS AND DISCUSSION

The average concentration of the radionuclides, Ra^{226}, Th^{232} and K^{40}, as well as the corresponding statistical error in the samples under investigation was calculated using the equation 1 and is shown in table 1. The activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K varied from 44±3 to 61±4 $Bqkg^{-1}$, 51±2 to 67±2 $Bqkg^{-1}$and 430±5 to 751±8 $Bqkg^{-1}$ respectively in various soil samples, the activity concentrations in stone sample found to vary from 29±2 to 156±6 $Bqkg^{-1}$, 31±2 to 300±5 $Bqkg^{-1}$and 396±5 to 1705±14 $Bqkg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K respectively and the activity concentrations in sand samples found to vary from 18±1.5 to 36±3 $Bqkg^{-1}$, 23±1 to 50±2 $Bqkg^{-1}$and 32±0.5 to 455±5 $Bqkg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K respectively. Table 2 indicates that the range of radium equivalent from minimum

$$\text{Absorbed dose D } (nGyh^{-1}) = 0.462\ C_{Ra} + 0.604\ C_{Th} + 0.0417\ C_K \quad (3)$$

$$\text{Indoor annual effective dose (mSv)} = (\text{Absorbed dose})\ nGyh^{-1} \times 8760h \times 0.8 \times 0.7 SvGy^{-1} \times 10^{-6} \quad (4)$$

$$\text{Outdoor annual effective dose (mSv)} = (\text{Absorbed dose})\ nGyh^{-1} \times 8760h \times 0.2 \times 0.7 SvGy^{-1} \times 10^{-6} \quad (5)$$

to maximum was 149 to 212 Bq/kg in the soil samples with an average of 186±11 Bq/kg and 106 to 677 Bq/kg in stone samples with an average of 319±92 Bq/kg calculated using the equation 2. Similarly, 54 to 142 Bq/kg in sand samples with an average of 101±15 Bq/kg. In the present study, the samples were found to have Ra_{eq} values smaller that the upper recommended value [1] of 370 $Bqkg^{-1}$ except in some stone samples SB and SR.

Using equations 3 to 5, the absorbed and annual effective dose rates from the samples were calculated as shown in table 2. The minimum and maximum values of absorbed dose, indoor annual effective dose and outdoor annual effective dose were found to vary from 69$nGyh^{-1}$ to 98 $nGyh^{-1}$ with an average of 86±5 $nGyh^{-1}$, 0.34mSv to 0.48mSv with an average of 0.42±0.02mSv and 0.08mSv to 0.12mSv with an average of 0.10±0.01mSv, in the soil samples studied in the present work. All the samples analyzed in the present work satisfy the safety criterion, i.e. the annual effective dose is less than 1mSv, the recommended safety limit for general public [16]. Hence, these samples do not pose any health hazard for the occupants. The minimum and maximum values of absorbed dose, indoor annual effective dose and outdoor annual effective dose were found to vary from 50 to 306 $nGyh^{-1}$ with an average of 146±42 $nGyh^{-1}$, 0.2 to 1.5mSv with an average of 0.7±0.2 mSv and 0.06 to 0.37mSv with an average of 0.17±0.05 mSv, in the stone samples studied in the present work. Ra_{eq} is related to the external γ-dose and internal dose due to radon and its daughters. Similarly, the minimum and maximum values of absorbed dose, indoor annual effective dose and outdoor annual effective dose were found to vary from 24 to 66 $nGyh^{-1}$ with an average of 46±7 $nGyh^{-1}$, 0.1 to 0.3 mSv with an average of 0.2±0.03 mSv and 0.03 to 0.08 mSv with an average of 0.05±0.008 mSv in the sand samples.

Using equations 6 and 7, the internal and external indices from the samples were calculated as shown in a table 2. Based on

Table 1. Activity concentration of radium, thorium and potassium for soil, stone and sand samples.

Samples	Sample codes	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
Soil	WMP	49±3	57±2	586±6
	MMP	61±4	62±2	554±6
	NSP	51±3	66±2	751±8
	26P	44±3	51±2	430±5
	NA	60±4	67±2	726±8
Stone	STY	47±4	37±2	405±5
	STR	131±6	52±2	460±5
	SB	119±6	300±5	1682±14
	SR	156±6	258±5	1705±14
	STRS	29±2	31±2	414±5
	STBR	85±5	60±2	1049±10
	STNS	61±5	56±2	396±5
Sand	CSY	34±2	46±2	450±5
	FSY	36±3	50±2	455±5
	RSB1	23±2	25±1	34±0.5
	DS	24±2	32±1	260±3
	RSB2	18±1.5	23±1	32±0.5
	NS	34±2	45±2	303±3.5

the dose criterion of 1 $mSvY^{-1}$, table 2 indicates that I_α and I_γ is less than unity in most of the samples investigated, regardless of the ways and the amounts in which the material is used except in black (SB) and red stones (SR). As indicated in table 2, the ^{226}Ra activities in the samples investigated were lower than 200 Bq kg^{-1}, so the alpha indices in materials did not exceed the recommended upper level.

In the present investigations the observed values of radium, thorium and potassium in soil, stone and sand samples are comparable with other Indian studies reported for soil, stone and sand as shown in table 3.

The natural radioactivity levels measured in the samples under present study are below the recommended limits except for black stone (SB) and red stone (SR). However, these samples satisfy the universal standards [12] limiting the radioactivity within the safe limits. Computed radiological data indicates that the black and red stone under study may enhance the indoor radiation dose when used as building construction materials.

Table 2. Activity concentration of radium equivalent, absorbed dose, dose equivalents for soil, stone and sand samples.

Samples	Sample codes	Ra_{eq} (Bq/kg)	D ($nGyh^{-1}$)	Indoor (mSv)	Outdoor (mSv)	Internal Index	External Index
Soil	WMP	176	82	0.40	0.10	0.2	0.6
	MMP	192	89	0.43	0.10	0.3	0.7
	NSP	202	94	0.46	0.11	0.2	0.7
	26P	149	69	0.34	0.08	0.2	0.5
	NA	212	98	0.48	0.12	0.3	0.8
Range		149-212	69-98	0.34-0.48	0.08-0.12	0.2-0.3	0.5-0.8
AM±SE*		186±11	86±5.0	0.42±0.02	0.10±0.01	0.2±0.02	0.7±0.05
Stone	STY	132	61	0.3	0.07	0.2	0.4
	STR	240	111	0.5	0.13	0.6	0.8
	SB	677	306	1.5	0.37	0.6	2.5
	SR	656	299	1.4	0.36	0.8	2.4
	STRS	106	50	0.2	0.06	0.1	0.4
	STBR	251	119	0.6	0.14	0.4	0.9
	STNS	171	78	0.4	0.09	0.3	0.6
Range		106-677	50-306	0.2-1.5	0.06-0.37	0.1-0.8	0.4-2.4
AM±SE*		319±92	146±42	0.7±0.2	0.17±0.05	0.4±0.1	1.1±0.3
Sand	CSY	134	62	0.3	0.07	0.2	0.5
	FSY	142	66	0.3	0.08	0.2	0.5
	RSB1	61	27	0.1	0.03	0.1	0.2
	DS	90	41	0.2	0.05	0.1	0.3
	RSB2	54	24	0.1	0.03	0.1	0.2
	NS	122	56	0.3	0.07	0.2	0.4
Range		54-142	24-66	0.1-0.3	0.03-0.08	0.1-0.2	0.2-0.5
AM±SE*		101± 15	46± 7	0.2 ± 0.03	0.05±0.008	0.15±0.02	0.35±0.05

*** SE** (standard error) = $\sigma/\sqrt{N}$, Where σ is SD (standard deviation) and N is the no of observations.

Table 3. Comparison of levels of present study with other Indian samples.

Sample Name	Location	^{226}Ra (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})	Reference
Soil	Fatehbad & Hissar (western Haryana)	19.24 - 32.21	30.21 - 64.25	346.25 - 607.32	Mehra, 2010 [17]
	Upper Siwaliks and Punjab	28.3- 81.0	61.2-140.3	363.4-1002.2	Singh *et al.* 2009 [18]
	Western Ghats-India	-	39.17-76.13	127.54-248.12	Manigandan *et al.* 2007 [19]
	Malwa region-Punjab	18.37-53.11	57.28-148.28	211.13-413.27	Mehra *et al.* 2007 [20]
	Punjab and Himachal Pradesh	18.22-90.30	34.80-124.68	80.42-181.41	Singh *et al.* 2005 [21]
	North-Eastern Haryana	44-61	51-67	430-751	Present Study
Stone	North Karnataka Gadag region Gulbarga region Kottur region	 - - -	 61.20-96.88 6.27-8.21 16.72-23.30	 966.75-1214.86 97.03-149.22 751.90-1135.16	Kerur *et al.* 2010 [22]
	North-Eastern Haryana	29-156	31-300	396-1705	Present Study
Sand	Gobichetti-palayam town	9.61-5.40	10.96-52.92	13.25-83.39	Brahmanandhan *et al.* 2007 [23]
	Kalpakkam,T.N	-	352-3872	324-405	Kannan *et al.* 2002 [24]
	North-Eastern Haryana	18-36	23-50	32-455	Present Study

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