

Measurement of Radioactivity of ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K in Soil of Different Geological Origins in **Northern India**

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ABSTRACT

Radioactivity of the nuclides ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K was measured in soil by y-ray spectrometry using HPGe detector. A criterion was set in order to analyse soil samples from plain, semi-hilly and hilly areas in northern India. More than three γ -ray energy peaks were used for the determination of ^{226}Ra and ^{232}Th activity concentrations to obtain more accurate results. Some of these peaks have interfering energies, which was caused by the limited resolution of the detector, but they were resolved theoretically and used in the analysis because of their significance in reducing the random error to its minimum level. Relationships between the measured radionuclides have been discussed elaborately. Radionuclides ^{238}U and ^{226}Ra were found in disequilibrium with ratio of specific activities ($^{238}U/^{226}Ra$) less than unity for most of the samples. In some cases this disequilibrium may be significant enough to modify the y-ray dose factors.

Keywords: Soil, Natural Radionuclides, Disequilibrium, Dose

1. Introduction

There is a great interest all over the world in the study of terrestrial radioactivity to carry out the radiological characterisation of soils and building materials. This kind of study is important not only for the assessment of public dose rates and health risks but also to keep reference-data record to document the possible changes in the environmental radioactivity induced by humans in future as far as the nuclear activities are concerned [1].

Natural environmental radioactivity depends on the geological and geographic conditions, and appears at different levels in the soils of each different geological region [2]. It comes mainly from uranium (²³⁸U) series, thorium $(^{232}$ Th) series and natural potassium $(^{40}$ K). Abnormal occurrences of uranium and its decay products in rocks and soils, and thorium in monazite sands have been identified in several areas of the world, e.g., Yangjiang in China, Rasmar in Iran, Kerala coast of India, etc. [3-5].

This study deals with the measurement of specific activities of the naturally occurring radionuclides (²³⁸U, 232 Th. and 40 K) in soil and consequently the assessment of γ -radiation dose. Soil samples were collected and their γ -radiation measured in our laboratory by γ -ray spectrometry using HPGe detector. The samples were collected from four sites (Bathinda, Amritsar, Pathankot and Dalhousie). These sites cover different geographical features, especially sandy soil at Bathinda and rocks at Dalhousie, making them interesting sites for radiological studies. Each site is different geologically and as well as climatically from the other. Amritsar and Bathinda fall in plain area of Punjab state of India. Pathankot is semi-hilly area at the border of Punjab and Himachal Pradesh states. Dalhousie is situated in the mountains of Chamba district of Himachal Pradesh. Although Amritsar and Bathinda both are plain areas, the nature of soil at these places is quite different from each other. Soil at Amritsar mainly consists of silty clay of low to medium plasticity. Texture of the soil at Bathinda is sandy to silty. Majority of the soil at Pathankot is a mixture of pebbles and sandy loam. Soil at Dalhousie is formed of hard and compact sedimentary rock (silty sandstone). Rainfall is frequent in Dalhousie all over the year, while the climate is drier in Amritsar and Bathinda.

2. Sample Collection and Measuring Methods

Soil samples were collected from 24 points at selected

four sites with 6 samples from each site. Each sample was taken from a depth of 5 cm - 10 cm at a randomly chosen point within the site area. The only constraint was that no sample was taken close to a field boundary, tree, building, or other obstruction. However, sampling points at each site were selected at a minimum distance of 2 km from each other in order to cover a large area and to observe a significant local spatial variation in terrestrial radioactivity. In order to obtain representative samples, they were thoroughly mixed, sieved to remove stones, pebbles grass roots and straws and then crushed to pass through a 200 micron mesh sieve to homogenise the contents. Then they were air-dried for several days and finally, were packed in plastic cylindrical containers. which were well sealed with celluloid tape. The volume of the soil sample in the container was kept constant on 245 cm³. The samples filled in the containers were left for at least 4 weeks before measuring by γ -spectrometry in order to ensure that radioactive equilibrium was reached between ²²⁶Ra, ²²²Rn, and hence ²²²Rn progeny. The volume of the soil samples was equal to that of the standard sample to maintain the geometrical identity. Therefore, due to the varying density of soils, the sample weight was also varying.

Soil samples were analysed for the activity of natural radionuclides using a High Purity Germanium (HPGe) coaxial n-type detector at Inter University Accelerator Center(IUAC), New Delhi. The detector had a resolution of 2.0 KeV at 1332 KeV and relative efficiency of 20% relative to a 3" × 3" NaI(Tl) detector. Spectrum was analysed using the software "CANDLE" (Collection and Analysis of Nuclear Data using Linux Network)"developed locally at Inter University Accelerator Center. The detector was shielded using 4" lead on all sides to reduce the background level of the system. Each sample was measured for a time period of 72,000 seconds. Activity concentration of each radionuclide was computed according to the following formula:

Activity =
$$\frac{PA \times 100 \times 100}{I \times \varepsilon \times 72000}$$
 (1)

where PA is peak area for a given energy, I is γ -ray intensity in percentage and ε is efficiency in percentage of the detector for that energy. It is well known that ²³⁸U and ²²⁶Ra may not be in

It is well known that ²³⁸U and ²²⁶Ra may not be in secular equilibrium, therefore, an independent (from ²²⁶Ra) method was used to measure ²³⁸U.

2.1. Estimation of ²³⁸U Concentration

Direct determination of 238 U using semiconductor γ -ray spectrometer is very hard because 238 U does not have intensive γ -rays (lines) of its own. But it has several daughter products which have more intensive lines and

activities equal to those of their parents in the state of secular equilibrium. The most intensive energies, used to determine²³⁸U contents, either come from²²⁶Ra and its progeny or from ²³⁴Th [6]. However, ²²⁶Ra is generally not in equilibrium with ²³⁸U because of geochemical reasons, therefore, the idea of using the energies of ²²⁶Ra progeny was discarded. ²³⁴Th is surely in equilibrium with 238 U and many authors use the 63.3 keV (3.6%) and 92.6 keV (4.9%) lines of 234 Th to determine 238 U. However, the interference of the 92.6 keV line from X-rays of Bi, Po, U and Th, and the interference of the 63.3 keV line from a 63.7 keV line of the thorium series may cause uncertainty in the results [7]. Moreover, the self absorption of these low-energy γ -rays in the sample material is significant and depends strongly on the varying composition and density of the sample. These facts may lead to the increase of the random error [8].

In the present study, ²³⁸U was determined through the determination of ²³⁵U assuming ²³⁵U/²³⁸U isotopic ratio to be constant for soils and rocks at 7.2×10^{-3} . ²³⁵U is determined using its own intensive line at 185.7 keV (57.5 %). Intensity of this peak relative to ²³⁸U is about 2.6% that gives a little bit lower peak intensity and higher statistical uncertainty than that of the 63.3 keV peak of ²³⁴Th. However, the 185.7 keV peak suffers from interference only from ²²⁶Ra, and the uncertainty caused by self-absorption is less because the higher γ -ray energy. The photo peak at the 185.7 keV line is interfered by the 186.1 keV line of ²²⁶Ra, which makes it necessary to resolve by using some mathematical relations. In the present analysis, the peak area under the 186 keV peak, was treated as sum:

$$PA(186) = PA(^{235}U, 185.7) + PA(^{226}Ra, 186.1)$$
 (2)

To deduce ²²⁶Ra contribution, it was assumed to be in equilibrium with its progeny. The intense γ -rays of ²¹⁴Pb and ²¹⁴Bi were used subsequently to determine ²²⁶Ra activity. For example, 295.2 keV γ -ray of ²¹⁴Pb were chosen for this purpose to yield:

$$PA(^{226}Ra, 186.1) = \frac{\varepsilon(186.1) \times I(^{226}Ra, 186.1)}{\varepsilon(295.2) \times I(^{214}Pb, 295.2)} \times PA(^{214}Pb, 295.2)$$
(3)

where I(^AX, E_{γ}) and $\epsilon(E_{\gamma})$ are the emission probability and detection efficiency, respectively, for the E_{γ} (keV) γ -ray of radionuclide ^AX. Similarly, PA(²²⁶Ra, 186.1) was computed also using the 351.9, 609.3 and 1120.3 keV lines and the average value for PA(²²⁶Ra, 186.1) was computed and subtracted from the measured PA(186) to yield the value of PA(²³⁵U, 185.7) according to Equation (2). Activity of ²³⁵U was, then, calculated using Equation (1). Finally, ²³⁸U was computed from the following equation:

$$= \frac{T_{1/2}(^{235}U) \times M(^{235}U)}{T_{1/2}(^{238}U) \times M(^{238}U)} \times \frac{^{235}U(Bq/kg)}{F}$$
(4)

where $T_{1/2}(^{A}X)$ is the half-life and M(^AX) is atomic mass of radionuclide ^AX. Factor F is $^{235}U/^{238}U$ isotopic, which is assumed to be constant for soil and rocks at 7.2×10^{-3} [7].

3. Results and Discussion

3.1. Specific Activities of 238 U, 226 Ra, 232 Th and 40 K

Table 1 illustrates the specific activities of the natural radionuclides (²³⁸U, ²²⁶Ra, ²³²Th, and ⁴⁰K) in the samples collected from four sites. Activity of ²³⁸U, collectively for all the 24 soil samples, found to be ranged from 22.63 to 116.23 Bg/kg with an average value of 50.5 Bg/kg. Activity of ²²⁶Ra varied from 19.47 to 96.08 Bq/kg with an average value of 54.13 Bg/kg. The ²³²Th activity ranged from 30.2 to 136.12 Bq/kg with an average value of 83.04 Bq/kg. While the activity of ⁴⁰K ranged from 189.47 to 508.05 Bq/kg with an average of 337.53 Bq/kg. Dalhousie location 3 has the highest values of ²³⁸U, ²²⁶Ra and ²³²Th, while Pathankot location 1 has the highest value of ⁴⁰K. Average activity concentrations of ²³⁸U, 226 Ra and 232 Th are significantly higher, and activity concentration of ⁴⁰K is lower as compared to the worldwide average concentrations in soils of various countries, which are 33 Bq/kg for ²³⁸U, 32 Bq/kg for ²²⁶Ra, 45 Bq/kg for 232 Th and 420 Bq/kg for 40 K [2]. Each out of 24 soil samples follows the activity order as 226 Ra $< ^{232}$ Th $< ^{40}$ K. For a detailed study, 238 U/ 226 Ra, 232 Th/ 226 Ra, $^{40}\text{K}/^{226}\text{Ra}$ and $^{40}\text{K}/^{232}\text{Th}$ ratios are given in Table 2. It is generally expected that ²³⁸U and ²²⁶Ra being in the same series, are in equilibrium, however, diversity of their ratio from unity was found in the present measurements as explained separately in section 4.2. ²³²Th/²²⁶Ra varied in the range of 1.25 and 1.85 with an average value of 1.55. 40 K/²²⁶Ra ranged from 3.4 to 10.15 with an average value of 6.61. 40 K/ 232 Th ranged from 2.72 to 6.54 with an average value of 4.27. This ratio can be used as an indicator of the relative occurrence of these radionuclides. Studying each site individually, it can be seen from Table 1 that the average activity values of Bathinda samples are highest for ²³⁸U, ²³²Th and ⁴⁰K, while ²²⁶Ra is highest for Dalhousie samples. The lowest average activity values of ²³⁸U, ²²⁶Ra and ²³²Th are for Pathankot samples. Average activity of ⁴⁰K is lowest for Amritsar samples. However, the contents of ²³⁸U and ²³²Th at Bathinda have insignificant difference from the corresponding contents at Dalhousie. Similarly, activity of ²²⁶Ra at Bathinda is approximately equal to that at Amritsar.

3.2. Study of 238U/226Ra Activity Concentration Ratio

Table 2 shows that 15 out of 24 samples have ${}^{238}U/{}^{226}Ra$ activity ratio less than unity. This ratio ranged from 0.56 to 1.56 having an average value of 0.95. The average value of ${}^{238}U/{}^{226}Ra$ in UNSCEAR, 2000 report is about 1.03. However, after a detailed study of this report, it is found that most of the values of ${}^{238}U$ presented in the report are based on assuming ${}^{238}U$ and ${}^{226}Ra$ in equilibrium. Compiling only those ${}^{238}U$ values, which were measured independently by some means, it has been observed that 9 out of 17 countries have experienced ${}^{238}U/{}^{226}Ra$ activity ratio less than unity as can be seen in **Table 3**.

Navas et al. [9] have reported ²³⁸U/²²⁶Ra ratio as low as 0.55 in the upper layer of soils (0 cm - 20 cm). They explained it by the significant differences in mobility of these radionuclides. According to them, ²³⁸U may be intensively leached from the soil surface and transferred to deeper soil sections where it accumulates. In comparison ²²⁶Ra is very immobile and keeps constant. This disturbance from equilibrium might be attributed to the weathering of rocks and the activity of water on the soil [6]. It is found that there is a positive correlation between 238 U/ 226 Ra ratio and water content in the soil [10]. Present study of ²³⁸U/²²⁶Ra provides further evidences for this fact as Dalhousie is known for frequent rain falling all over the year and average ²³⁸U/²²⁶Ra at this site is lowest to the value of 0.83. On the other side, Amritsar and Bathinda have drier climate and exhibit higher ²³⁸U/²²⁶Ra ratio. More surprisingly Bathinda shows ²³⁸U/²²⁶Ra ratio more than unity. This may be attributed to the spread of fly ash enriched in uranium contents, which is originated from the combustion of coal from a thermal plant running in this area for more than 50 years.

3.3. Dose Distribution of *γ*-Radiations

Distribution of ²²⁶Ra, ²³²Th and ⁴⁰K in soil is not uniform. Therefore, in order to compare their combined radiological effect, a common index called the radium equivalent activity (Ra_{eq}) was used. The radium equivalent activity is a weighted sum of activities of the above three radionuclides based on the assumption that 370 Bq/Kg of ²²⁶Ra, 259 Bq/Kg of ²³²Th and 4810 Bq/Kg of ⁴⁰K produces the same gamma dose rate. It is calculated through the following relation [11]:

$$Ra_{eq} = 370 \left(\frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \right)$$
(5)

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077_{K}$$
(6)

where C_{Ra} , C_{Th} and C_K are the activity concentrations (in Bq/kg) for ²²⁶Ra, ²³²Th and ⁴⁰K, respectively. It can be seen from **Table 1** that the radium equivalent for all the 24 soil samples in the present study ranged between

77.87 Bq/kg for Pathankot location 6 and 325.71 Bq/kg for Dalhousie location 3 at an average value of 198.86 Bq/kg, which is lower than the allowed maximum value of 370 Bq/kg [11,12]. The contributions of 226 Ra, 232 Th and 40 K in the average value of Ra_{eq} are in the ratio

Table 1. Activity concentration of ²³⁸U, ²²⁶Ra, ²³²Th, and ⁴⁰K in some Indian soils (Bathinda, Amritsar, Pathankot and Dalhousie).

Sample	²³⁸ U (Bq/kg)	²²⁶ Ra (Bq/kg)	²³² Th (Bq/kg)	⁴⁰ K (Bq/kg)	Ra _{eq} (Bq/kg)
Bathinda location 1	49.64 ± 4.13	57.16±1.08	104.08 ± 4.08	489.55 ± 5.15	243.69 ± 7.31
Bathinda location 2	41.7 ± 3.92	44.12 ± 0.88	72.64 ± 3.08	252.75 ± 3.10	167.46 ± 5.52
Bathinda location 3	97.32 ± 5.20	82.28 ± 1.48	124.36 ± 5.24	507.5 ± 5.53	299.19 ± 9.40
Bathinda location 4	58.57 ± 4.36	74.68 ± 1.24	121.44 ± 1.96	486.45 ± 5.28	285.8 ± 4.45
Bathinda location 5	45.6 ± 4.02	41.88 ± 0.80	71.28 ± 3.64	272.28 ± 3.28	164.78 ± 6.26
Bathinda location 6	52.33 ± 4.20	33.92 ± 0.56	62.68 ± 3.52	253.73 ± 3.10	143.09 ± 5.83
Average	57.53 ± 4.30	55.67 ± 1.01	92.75 ± 3.59	377.04 ± 4.24	217.33 ± 6.46
Standard deviation	20.33	19.33	27.28	129.06	67.59
Amritsar location 1	35.26 ± 3.73	56.36 ± 1.08	90.64 ± 3.28	301.66 ± 3.50	209.20 ± 6.04
Amritsar location 2	31.40 ± 3.61	35.40 ± 0.66	49.72 ± 1.71	231.56 ± 2.88	124.33 ± 3.33
Amritsar location 3	55.47 ± 4.28	42.48 ± 0.78	65.48 ± 2.91	214.25 ± 2.73	152.61 ± 5.15
Amritsar location 4	37.81 ± 3.80	46.31 ± 0.90	57.73 ± 2.46	257.89 ± 3.11	148.72 ± 4.66
Amritsar location 5	40.27 ± 3.87	51.08 ± 0.98	74.10 ± 3.67	312.24 ± 3.60	181.09 ± 6.51
Amritsar location 6	103.46 ± 5.31	95.08 ± 1.56	132.16 ± 5.12	493.18 ± 5.18	322.04 ± 9.28
Average	50.61 ± 4.10	54.45 ± 0.99	78.31 ± 3.19	301.80 ± 3.50	189.67 ± 5.83
Standard deviation	27.17	21.16	29.92	101.25	71.13
Pathankot location 1	42.36 ± 3.93	56.96 ± 1.04	100.08 ± 3.80	508.05±5.32	239.19±6.88
Pathankot location 2	26.78±3.46	20.38±0.47	33.28±1.89	189.47±2.52	82.56±3.37
Pathankot location 3	23.83±3.35	34.72±0.72	61.96 ± 4.16	242.08 ± 3.00	141.96 ± 6.90
Pathankot location 4	33.42 ± 3.67	59.32 ± 1.11	101.92 ± 3.88	442.5 ± 4.78	239.14 ± 7.03
Pathankot location 5	70.29 ± 4.63	64.30 ± 1.14	89.72 ± 3.29	403.39 ± 4.41	223.66 ± 6.18
Pathankot location 6	30.41 ± 3.58	19.47 ± 0.51	30.20 ± 1.77	197.6 ± 2.56	77.87 ± 3.24
Average	37.85 ± 3.77	42.53 ± 0.83	69.53 ± 3.13	330.52 ± 3.77	167.40 ± 5.60
Standard deviation	17.13	20.24	32.58	137.66	76.58
Dalhousie location 1	23.47 ± 3.34	29.26 ± 0.55	37.14 ± 1.30	223.78 ± 2.82	99.60 ± 2.63
Dalhousie location 2	74.86 ± 4.73	75.40 ± 1.80	126.72 ± 5.40	485.25 ± 5.13	293.97 ± 9.92
Dalhousie location 3	116.23 ± 5.55	96.08 ± 1.68	136.12 ± 5.24	454.28 ± 4.88	325.71 ± 9.55
Dalhousie location 4	22.63 ± 3.31	38.07 ± 0.71	51.56 ± 2.23	264.80 ± 3.16	132.19 ± 4.14
Dalhousie location 5	61.50 ± 4.43	87.44 ± 1.44	109.32 ± 4.12	297.10 ± 3.47	266.64 ± 7.60
Dalhousie location 6	37.43 ± 3.79	57.00 ± 0.96	88.52 ± 3.59	319.48 ± 3.60	208.18 ± 6.37
Average	56.02 ± 4.19	63.88 ± 1.19	91.56 ± 3.65	340.78 ± 3.84	221.05 ± 6.70
Standard deviation	36.14	26.97	40.27	105.43	90.73

a 1	²³⁸ U/ ²²⁶ Ra	²³² Th/ ²²⁶ Ra	40xx 226m	⁴⁰ K/ ²³² Th
Sample			⁴⁰ K/ ²²⁶ Ra	
Bathinda location 1	0.87 ± 0.09	1.82 ± 0.11	8.56 ± 0.25	4.70 ± 0.23
Bathinda location 2	0.95 ± 0.11	1.65 ± 0.10	5.73 ± 0.18	3.48 ± 0.19
Bathinda location 3	1.18 ± 0.08	1.51 ± 0.09	6.17 ± 0.18	4.08 ± 0.22
Bathinda location 4	0.78 ± 0.07	1.63 ± 0.05	6.51 ± 0.18	4.01 ± 0.11
Bathinda location 5	1.09 ± 0.12	1.70 ± 0.12	6.50 ± 0.20	3.82 ± 0.24
Bathinda location 6	1.54 ± 0.15	1.85 ± 0.13	7.48 ± 0.21	4.05 ± 0.28
Average	1.07 ± 0.10	1.69 ± 0.10	6.83 ± 0.20	4.02 ± 0.21
Standard deviation	0.27	0.13	1.03	0.40
Amritsar location 1	0.63 ± 0.08	1.61 ± 0.09	5.35 ± 0.16	3.33 ± 0.16
Amritsar location 2	0.89 ± 0.12	1.40 ± 0.07	6.54 ± 0.20	4.66 ± 0.22
Amritsar location 3	1.31 ± 0.12	1.54 ± 0.10	5.04 ± 0.16	3.27 ± 0.19
Amritsar location 4	0.82 ± 0.10	1.25 ± 0.08	5.57 ± 0.18	4.47 ± 0.24
Amritsar location 5	0.79 ± 0.09	1.45 ± 0.10	6.11 ± 0.19	4.21 ± 0.26
Amritsar location 6	1.09 ± 0.07	1.39 ± 0.08	5.19 ± 0.14	3.73 ± 0.18
Average	0.92 ± 0.10	1.44 ± 0.09	5.63 ± 0.17	3.95 ± 0.21
Standard deviation	0.24	0.13	0.58	0.59
Pathankot location 1	0.74 ± 0.08	1.76 ± 0.10	8.92 ± 0.26	5.08 ± 0.25
Pathankot location 2	1.31 ± 0.20	1.63 ± 0.13	9.30 ± 0.34	5.69 ± 0.40
Pathankot location 3	0.69 ± 0.11	1.78 ± 0.16	6.97 ± 0.23	3.91 ± 0.31
Pathankot location 4	0.56 ± 0.07	1.72 ± 0.10	7.46 ± 0.22	4.34 ± 0.21
Pathankot location 5	1.09 ± 0.09	1.40 ± 0.08	6.27 ± 0.18	4.50 ± 0.21
Pathankot location 6	1.56 ± 0.22	1.55 ± 0.13	10.15 ± 0.40	6.54 ± 0.47
Average	0.99 ± 0.13	1.64 ± 0.12	8.18 ± 0.27	5.01 ± 0.31
Standard deviation	0.39	0.15	1.50	0.97
Dalhousie location 1	0.80 ± 0.13	1.27 ± 0.07	7.65 ± 0.24	6.03 ± 0.29
Dalhousie location 2	0.99 ± 0.09	1.68 ± 0.11	6.44 ± 0.22	3.83 ± 0.20
Dalhousie location 3	1.21 ± 0.08	1.42 ± 0.08	4.73 ± 0.13	3.34 ± 0.16
Dalhousie location 4	0.59 ± 0.10	1.35 ± 0.08	6.96 ± 0.21	5.14 ± 0.28
Dalhousie location 5	0.70 ± 0.06	1.25 ± 0.07	3.40 ± 0.10	2.72 ± 0.13
Dalhousie location 6	0.66 ± 0.08	1.55 ± 0.09	5.60 ± 0.16	3.61 ± 0.19
Average	0.83 ± 0.09	1.42 ± 0.08	5.80 ± 0.18	4.11 ± 0.21
Standard deviation	0.23	0.17	1.56	1.23

Table 2. Activity concentration ratios ²³⁸U/²²⁶Ra, ²³²Th/²²⁶Ra, ⁴⁰K/²²⁶Ra and ⁴⁰K/²³²Th in some Indian soils (Bathinda, Amritsar, Pathankot and Dalhousie).

Table 3. Activity concentration ratio ²³⁸U/²²⁶Ra in various countries (UNSCEAR, 2000).

Country	²³⁸ U(Bq m ⁻³)/ ²²⁶ Ra(Bq m ⁻³)	
Algeria	30/50 (0.60)	
Egypt	37/17 (2.18)	
United States	35/40 (0.88)	
China	33/32 (1.03)	
Hong Kong	84/59 (1.42)	
Japan	29/33 (0.88)	
Kazakhstan	37/35 (1.06)	
Malaysia	66/67 (0.99)	
Thailand	114/48 (2.38)	
Armenia	46/51 (0.90)	
Syrian Arab Republic	23/20 (1.15)	
Ireland	37/60 (0.62)	
Bulgaria	40/45 (0.89)	
Hungary	29/33 (0.88)	
Russian Federation	19/27 (0.70)	
Croatia	110/54 (2.04)	
Portugal	49/44 (1.11)	

27.2:59.7:13.1. Average results of Ra_{eq} for each site are also listed in Table 1, which is highest at Dalhousie at the value of 221.05 Bq/kg and lowest at Pathankot at the value of 167.4 Bq/kg.

The external terrestrial γ -radiation absorbed dose rate in air at a height of about 1 m above the ground is calculated by using the following relation [2]:

$$D(nGy/h) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_{K}$$
(7)

Results of γ -radiation absorbed dose rate are shown in **Table 4**. It ranged between 35.48 nGy/h for Pathankot location 6 and 145.55 nGy/h for Dalhousie location 3 at an average value of 89.24 nGy/h, which is higher by a factor of 1.49 than the world's average value of 60 nGy/h [2].The contributions of ²²⁶Ra, ²³²Th and ⁴⁰K in the average value are in the ratio 28:56.2:15.78. Results for each

sample as well as average results for each site are also listed in **Table 4**.

To estimate annual effective dose, account was taken of the conversion coefficient from absorbed dose in air to effective dose and the indoor occupancy factor. The committee used 0.7 Sv/Gy for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.2 for the outdoor occupancy factor [2, 13]. Effective dose rate outdoors in units of mSv per year was calculated by the following formula:

Results for outdoor annual effective dose are shown in **Table 4**. It ranged from 0.4 to 0.18 mSv with an average

Table 4. Absorbed dose rates from ²²⁶Ra, ²³²Th, and ⁴⁰K and annual effective dose (Bathinda, Amritsar, Pathankot and Dalhousie).

		Annual effective dose			
Sample	²²⁶ Ra (nGy/h)	²³² Th (nGy/h)	⁴⁰ K (nGy/h)	Total (nGy/h)	(mSv)
Bathinda location 1	26.41 ± 0.50	62.86 ± 2.46	20.41 ± 0.21	109.69 ± 3.18	0.135 ± 0.004
Bathinda location 2	20.38 ± 0.41	43.87 ± 1.86	10.54 ± 0.13	74.80 ± 2.39	0.092 ± 0.003
Bathinda location 3	38.01 ± 0.68	75.11 ± 3.16	21.16 ± 0.23	134.29 ± 4.08	0.165 ± 0.005
Bathinda location 4	34.50 ± 0.57	73.35 ± 1.18	20.28 ± 0.22	128.14 ± 1.97	0.157 ± 0.002
Bathinda location 5	19.35 ± 0.37	43.05 ± 2.20	11.35 ± 0.14	73.76 ± 2.70	0.090 ± 0.003
Bathinda location 6	15.67 ± 0.26	37.86 ± 2.13	10.58 ± 0.13	64.11 ± 2.51	0.079 ± 0.003
Average	25.72 ± 0.47	56.02 ± 2.17	15.72 ± 0.18	97.47 ± 2.81	0.120 ± 0.003
Standard deviation	8.93	16.48	5.38	30.45	0.037
Amritsar location 1	26.04 ± 0.50	54.75 ± 1.98	12.58 ± 0.15	93.36 ± 2.63	0.114 ± 0.003
Amritsar location 2	16.35 ± 0.30	30.03 ± 1.03	9.66 ± 0.12	56.04 ± 1.46	0.069 ± 0.002
Amritsar location 3	19.63 ± 0.36	39.55 ± 1.76	8.93 ± 0.11	68.11 ± 2.23	0.084 ± 0.003
Amritsar location 4	21.40 ± 0.42	34.87 ± 1.49	10.75 ± 0.13	67.02 ± 2.03	0.082 ± 0.002
Amritsar location 5	23.60 ± 0.45	44.76 ± 2.22	13.02 ± 0.15	81.38 ± 2.84	0.100 ± 0.003
Amritsar location 6	43.93 ± 0.72	79.82 ± 3.09	20.57 ± 0.22	144.32 ± 4.03	0.177 ± 0.005
Average	25.16 ± 0.46	47.30 ± 1.93	12.59 ± 0.15	85.04 ± 2.53	0.104 ± 0.003
Standard deviation	9.78	18.07	4.22	31.78	0.039
Pathankot location 1	26.32 ± 0.48	60.45 ± 2.30	21.19 ± 0.22	107.95 ± 3.00	0.132 ± 0.004
Pathankot location 2	9.42 ± 0.22	20.10 ± 1.14	7.90 ± 0.11	37.42 ± 1.46	0.046 ± 0.002
Pathankot location 3	16.04 ± 0.33	37.42 ± 2.51	10.09 ± 0.13	63.56 ± 2.97	0.078 ± 0.004
Pathankot location 4	27.41 ± 0.51	61.56 ± 2.34	18.45 ± 0.20	107.42 ± 3.06	0.132 ± 0.004
Pathankot location 5	29.71 ± 0.53	54.19 ± 1.99	16.82 ± 0.18	100.72 ± 2.70	0.124 ± 0.003
Pathankot location 6	9.00 ± 0.24	18.24 ± 1.07	8.24 ± 0.11	35.48 ± 1.41	0.044 ± 0.002
Average	19.65 ± 0.38	41.99 ± 1.89	13.78 ± 0.16	75.43 ± 2.43	0.093 ± 0.003
Standard deviation	9.35	19.68	5.74	34.36	0.042
Dalhousie location 1	13.52 ± 0.25	22.43 ± 0.79	9.33 ± 0.12	45.28 ± 1.16	0.056 ± 0.001
Dalhousie location 2	34.83 ± 0.83	76.54 ± 3.26	20.23 ± 0.21	131.61 ± 4.31	0.161 ± 0.005
Dalhousie location 3	44.39 ± 0.78	82.22 ± 3.16	18.94 ± 0.20	145.55 ± 4.14	0.179 ± 0.005
Dalhousie location 4	17.59 ± 0.33	31.14 ± 1.35	11.04 ± 0.13	59.77 ± 1.81	0.073 ± 0.002
Dalhousie location 5	40.40 ± 0.67	66.03 ± 2.49	12.39 ± 0.14	118.82 ± 3.30	0.146 ± 0.004
Dalhousie location 6	26.33 ± 0.44	53.47 ± 2.17	13.32 ± 0.15	93.12 ± 2.76	0.114 ± 0.003
Average	29.51 ± 0.55	55.31 ± 2.20	14.21 ± 0.16	99.03 ± 2.91	0.121 ± 0.004
Standard deviation	12.46	24.32	4.40	40.21	0.049

value of 0.11 mSv, while the world wide outdoor average annual effective dose is approximately 0.07 mSv [2]. Only three samples, Pathankot location 2, Pathankot location 6 and Dalhousie location 1, have lower values than the worldwide average value, which are 0.05, 0.04 and 0.06 mSv, respectively. Average annual effective doses at Bathinda and Dalhousie are equal at 0.12 mSv and are higher than other two sites Amritsar by factor of 1.20 and Pathankot by a factor of 1.33.

4. Conclusions

1) For the determination of 226 Ra and 232 Th activity concentrations, more than three γ -ray energy peaks were used.

2) Average activity concentrations of 238 U, 226 Ra and 232 Th are higher and activity concentration of 40 K is lower as compared to the worldwide average concentration.

3) Dalhousie exhibit lowest ${}^{238}\text{U}/{}^{226}\text{Ra}$ ratio while Amritsar and Bathinda exhibit higher ${}^{238}\text{U}/{}^{226}\text{Ra}$ ratio due to their different climatic changes.

Average results of Ra_{eq} is highest at Dalhousie with value 221.05 Bq/kg and lowest at Pathankot with value 167.4 Bq/kg.

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