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NOTE

Assessment of the average effective dose from the analysis of ^{226}Ra, ^{232}Th and ^{40}K in soil samples from Punjab, India

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The activity concentrations of the natural radionuclides, namely ^{238}Ra, ^{232}Th and ^{40}K, are measured for soil samples collected from some locations of the Malwa region of Punjab. An HPGe detector, based on high-resolution gamma spectrometry system is used for the measurement of activity concentrations. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil from the studied areas varies from 23.3 Bq kg^{-1} (Jagraon) to 43.6 Bq kg^{-1} (Ludhiana), 104 Bq kg^{-1} (Raikot) to 148 Bq kg^{-1} (Kauli) and 290 Bq kg^{-1} (Rajpura) to 394 Bq kg^{-1} (Sarhind) having an overall mean value of 32 Bq kg^{-1}, 126 Bq kg^{-1} and 348 Bq kg^{-1} respectively. The radium equivalent activity (Ra_{eq}) in these soil samples ranges from 200 Bq kg^{-1} (Jagraon) to 264 Bq kg^{-1} (Kauli) having a mean value of 237 Bq kg^{-1}. Corresponding indoor and outdoor annual effective doses range from 0.45 to 0.59 mSv and 0.11 to 0.15 mSv and average 0.53 and 0.13 mSv. The external hazard index (H_{ex}) for soil samples of the study area are lower than unity; therefore, according to the Radiation Protection 112 (European Commission, 1999) report, soil from these regions is safe and can be used as a construction material without posing any significant radiological threat to population.

Keywords: gamma ray spectrometry, HPGe detector, Ra_{eq} activities, annual effective dose, external hazard index

INTRODUCTION

Human beings have always been exposed to natural radiations arising from within and outside the earth. Exposure to ionizing radiations from sources occurs because of naturally occurring radioactive elements in soil and rocks, cosmic rays entering the earth's atmosphere from outer space, and internal exposure from radioactive elements through food, water and air. Natural radioactivity in soil primarily comes from U and Th series and natural K. Artificial radionuclides can also be present such a ^{137}Cs, resulting from fallout from weapons testing and from accidents such as Chernobyl. The radiological implication of these radionuclides is caused by the gamma ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters. Therefore, the assessment of gamma-radiation doses from natural sources is of particular importance because natural radiation is the largest contributor to the external dose of the world's population (UNSCEAR, 1988).

External gamma-dose estimation produced by terrestrial sources is essential not only because it contributes considerably to the collective dose, but also because of variations of the individual dose related to this pathway. These doses vary depending upon concentrations of natural radionuclides, ^{238}U, ^{232}Th, their daughter products, and ^{40}K present in the soils and rocks, which in turn depend upon the local geology of each region in the world (Radhakrishna *et al*., 1993; Quindos *et al*., 1994; Kies *et al*., 2011; Wysocka, 2011; Solecki and Tchorz-Trzeciakiewicz, 2011; Vaupotič *et al*., 2011; Yang *et al*., 2011). The natural radioactivity of soil samples is usually determined from the ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K contents. Because 98.5% of the radiological effects of the uranium series are produced by radium and its daughter products, the contribution from ^{238}U and other ^{226}Ra, precursors are normally ignored.

High levels of uranium and its decay products in rock and soil and thorium in monazite sands are the main sources of high background in natural areas. There are a few regions in the world known to be High Background Radiation Areas (HBRAs) due to local geology and geochemical effects that cause enhanced levels of terrestrial radiation (Bennett, 1997; UNSCEAR, 1993, 2000). Very high background-radiation areas are found along the

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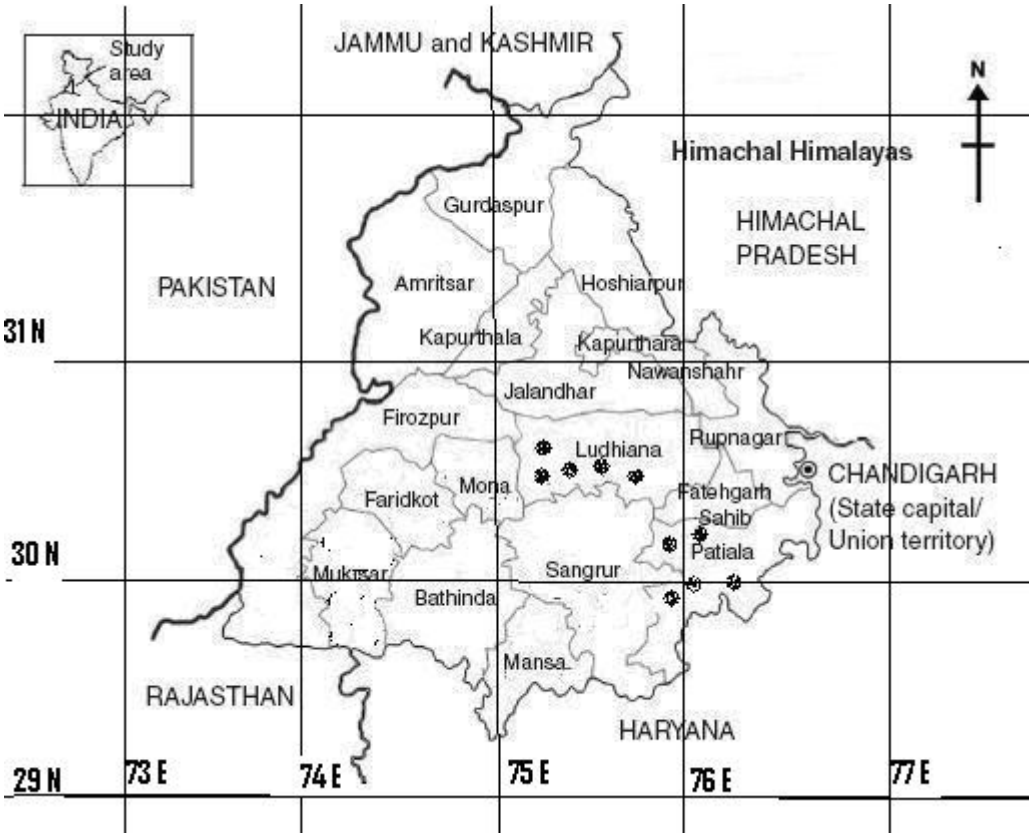

Fig. 1. Map of Punjab showing the area surveyed during this investigations.

southwest coast of India (Sunta *et al*., 1982; Sunta, 1993; Mishra, 1993; Paul *et al*., 1998); Ramsar and Mahallat in Iran (Sohrabi, 1993; Ghiassi-nejad *et al*., 2002); in the United States and Canada (NCRP, 1987), and in some other counties (UNSCEAR, 2000). Radium in soil/water and radon in the air are responsible for the radiation at Ramsar in Iran. In India, there are many monazite sand-bearing placer deposits causing high background radiation along its long coastline. Ullal in Karnataka (Radhakrishna *et al*., 1993), Kalpakkam in Tamil Nadu, (Kannan *et al*., 2002) coastal parts of the Tamil Nadu and Kerala states and the southwestern coast of India are known high background-radiation areas (Mishra, 1993; Sunta, 1993; Zhu *et al*., 1993; Sohrabi, 1993). Some of these areas have been under study for many years to determine the risks and effects of long-term, low-level, and natural radiation exposure (Sohrabi, 1998).

Therefore, measurements of natural radioactivity in soil are of a great interest for many researchers throughout the world, which promoted worldwide national surveys in the last two decades. To evaluate the terrestrial gamma-dose rate for outdoor occupations, it is very important to estimate the natural radioactivity level in soils. Specific levels are related to the types of rock from which the soils originate. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. From the review of literature it is observed that although the data on the content of radioactive elements is available for some states of India, data available for Punjab are quite meagre. We have measured natural radioactivity for the first time in this area. The main objective of this study is to determine the level of radioactive elements *viz*. radium, thorium and potassium in Malwa region of Punjab for health-risk assessment.

Experimental Procedure

Survey area

Figure 1 shows the geographic location of the states of the Punjab, as well as the location of the sampling sites. The geographic location of the study area in Punjab is between latitude 30.12°–30.55°N and longitude 74.43°–76.25°E. Punjab sediments are derived from Siwalik Himalayas and occur as alluvium. The surveyed area is bounded on the western side by Pakistan; on the south by Rajasthan and Haryana.

Sampling

For the measurement of the natural radioactivity in soil, surface-soil samples were collected randomly from ten different locations in the Patiala and Ludhiana districts of Malwa region of Punjab. The soil was collected from an auger hole at a depth of about 0.75 m so as to sample the natural soil. After collection, samples were crushed into fine powder by using mortar and pestle. The final grain-sizes of the samples were obtained by straining through a 150 micron-mesh size. An average 0.25 kg of soil is used per sample. Before measurement samples were dried in an oven at a temperature of 383°K for 24 hours. Each sample was packed and sealed in an airtight PVC container and kept for about a four-week period to allow radioactive equilibrium among the daughter products of radon (^{222}Ra), thoron (^{220}Ra) and their short lived decay products.

Measurement of natural radioactivity

Using an HPGe detector based on high-resolution gamma spectrometry system, the activity of samples was counted. The detector is a co-axial n-type high purity germanium detector (make EG&G, ORTEC, Oak Ridge, TN, USA). The detector has a resolution of 2.0 keV and relative efficiency of 20% for 1.332 MeV gamma energy of Co-60. The output of the detector was analyzed using a 4K MCA system connected to a PC. The spectral data were analyzed using the software "CANDLE" (Collection and Analysis of Nuclear Data using Linux nEtwork) developed locally by Inter University Accelerator Centre, New Delhi. The detector is shielded using 10 cm lead on all sides to reduce the background level of the system (Kumar *et al*., 2001). The efficiency calibration for the system was carried out using a secondary, standard source of uranium ore in geometry available for the sample counting. Efficiency values were plotted against energy for the particular geometry and are fitted by least-squares method to an empirical relation that accounts for the nature of the efficiency curve for the HPGe detector. For calibration of the low background-counting system, a secondary standard was used. The secondary standard was calibrated with the primary standard (RGU-1) obtained from the International Atomic Energy Agency. Gamma transitions of 1461 keV for ^{40}K, 186 keV and 609 keV of ^{214}Bi for ^{226}Ra, 338, 463, 911, 968 keV for ^{228}Ac, 727 keV for ^{212}Bi, 238 keV for ^{212}Pb were used for the laboratory measurement of activity concentration potassium, radium and thorium. The samples were counted for a period of 72,000 seconds, and the spectra of the photo peak of radium, thorium daughter products and ^{40}K were analyzed. The net count rate under the most prominent photo peaks of radium and thorium daughter peaks were calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclide was calculated from the background-subtracted area of the prominent gamma-ray energies. The concentrations of radionuclides are calculated using the following equation:

$$\text{Activity}\,(\text{Bq}) = \frac{CPS \times 100 \times 100}{B.I. \times Eff} \pm \frac{CPS_{\text{error}} \times 100 \times 100}{B.I. \times Eff} \quad (1)$$

where, CPS = net count rate per second, $B.I.$ = branching intensity, and Eff = efficiency of the detector.

Radium equivalent activity

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. It is calculated through the following relation (Yu *et al*., 1992):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.07C_K \quad (2)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. When defining Ra_{eq} activity, it has been assumed that 370 Bq kg^{-1} ^{226}Ra or 259 Bq kg^{-1} ^{232}Th or 4810 Bq kg^{-1} ^{40}K produce the same gamma-dose rate.

Calculation of air-absorbed dose rate

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 conversion factor of 0.0414 nGy h^{-1}/Bq kg^{-1} for ^{40}K, 0.461 nGy h^{-1}/Bq kg^{-1} for ^{226}Ra, and 0.623 nGy h^{-1}/Bq kg^{-1} for ^{232}Th (UNSCEAR, 1993) assuming that ^{137}Cs, ^{90}Sr and the ^{235}U decay series can be neglected. They contribute very little to the total dose from the environmental background (Kocher and Sjoreen, 1985; Jacob *et al*., 1986; Leung *et al*., 1990).

$$D\ (\text{nGy h}^{-1}) = 0.461C_{Ra} + 0.623C_{Th} + 0.0414C_K \quad (3)$$

where, C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq kg^{-1}) of radium, thorium and potassium in the samples.

Calculation of annual effective dose

To estimate annual effective doses, the following must be considered: (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. The annual, estimated, average, effective-dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv Gy^{-1}, which is used to convert the absorbed rate to human effective-dose

Table 1. The values ^{226}Ra, ^{232}Th and ^{40}K activity content using gamma-ray spectrometry and Ra_{eq} activity in the soil samples from Malwa Region of Punjab

Sr. No.	Sample location (Village)	Radium concentration in soil C_{Radium} (Bq kg^{-1})	Thorium concentration in soil $C_{Thorium}$ (Bq kg^{-1})	Potassium concentration in soil $C_{Potassium}$ (Bq kg^{-1})	Radium equivalent activity Ra_{eq} (Bq kg^{-1})
Patiala					
1	Nabha	23.6	118	388	220
2	Patiala	41.6	126	298	243
3	Samana	31.1	137	313	250
4	Kauli	27.2	148	359	264
5	Raj Pura	23.5	119	290	215
Ludhiana					
1	RaiKot	40.4	104	344	214
2	Ludhiana	43.6	129	385	255
3	Khanna	36.1	134	376	255
4	Sarhind	32.5	139	394	258
5	Jagraon	23.3	107	333	200

equivalent with an outdoor occupancy of 20% and 80% for indoors (UNSCEAR, 1993).

The annual effective doses are determined as follows:

Indoor (nSv)
= (absorbed dose) nGy^{-1} × 8760 h × 0.8 × 0.7 Sv Gy^{-1} (4)

Outdoor (nSv)
= (absorbed dose) nGy^{-1} × 8760 h × 0.2 × 0.7 Sv Gy^{-1}. (5)

External hazard index (H_{ex})

The external hazard index H_{ex} can be calculated by Eq. (6):

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1 \quad (6)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively (Beretka and Mathew, 1985). The values of this index must be less than unity in order to keep the radiation hazard significant. The radiation exposure from the radioactivity of a construction material is limited to 1.5 mGy Y^{-1}. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370 Bq kg^{-1}).

Results and Discussions

Table 1 summarizes the results of measurements of natural radionuclide (^{226}Ra, ^{232}Th and ^{40}K) concentrations in the collected soil samples. World average concentrations are 35 Bq kg^{-1}, 30 Bq kg^{-1} and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. Table 1 shows that, in general, the average and ranges of activity concentration of ^{232}Th in soil of these areas are higher than the world figures reported in UNSCEAR (2000). However, the concentration for ^{226}Ra is very much comparable and a concentration for ^{40}K is lower as compared with world figures. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil from the studied areas varies from 23.3 Bq kg^{-1} (Jagraon) to 43.6 Bq kg^{-1} (Ludhiana), 104 Bq kg^{-1} (Raikot) to 148 Bq kg^{-1} (Kauli) and 290 Bq kg^{-1} (Rajpura) to 394 Bq kg^{-1} (Sarhind) with overall mean values of 32 Bq kg^{-1}, 126 Bq kg^{-1} and 348 Bq kg^{-1}, respectively. Comparatively high values of ^{226}Ra and ^{232}Th in soil samples from the study area as compared to world averages may be correlated with the presence of uranium in the soil of this area which is also reported in earlier studies (Mehra *et al*., 2006). Our values for radium content in soil lie in the range (2.5–207 Bq kg^{-1}) reported for Indian soils (Nageswara Rao *et al*., 1996) using gamma-ray spectrometry and are less than the permissible value (370 Bq kg^{-1}), which is acceptable as safe limit (OECD, 1979). The values of ^{226}Ra are comparable with those reported for the adjoining districts of the Malwa region (Mehra *et al*., 2007, 2009), but the values of ^{232}Th and ^{40}K are higher than those as reported by Mehra *et al*. (2007, 2009). These higher values of thorium and potassium might result from

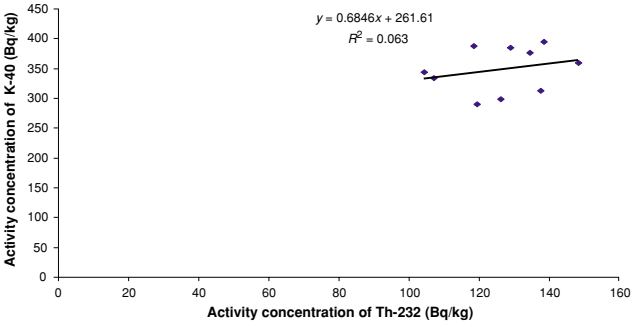

Fig. 2. Linear regression of the activity concentration of ^{40}K on the activity concentration of ^{232}Th.

Table 2. Air-absorbed dose rates and annual effective doses at various locations of Malwa Region of Punjab

Sr. No.	Sample location	Absorbed dose (nGy h^{-1})				External hazard index (H_{ex})	Annual effective dose (m Sv)	
	(Village)	^{226}Ra	^{232}Th	^{40}K	Total		Indoor	Outdoor
Patiala								
1	Nabha	10.9	73.7	16.1	101	0.6	0.49	0.12
2	Patiala	19.1	78.5	12.4	110	0.66	0.54	0.13
3	Samana	14.3	85.6	13.0	113	0.68	0.55	0.14
4	Kauli	12.6	92.3	14.9	120	0.72	0.59	0.15
5	Raj Pura	10.8	74.4	12.0	97	0.59	0.48	0.12
Ludhiana								
1	RaiKot	18.6	64.9	14.2	98	0.58	0.48	0.12
2	Ludhiana	20.1	80.3	15.9	116	0.7	0.57	0.14
3	Khanna	16.6	83.7	15.6	116	0.69	0.57	0.14
4	Sarhind	14.9	86.3	16.3	118	0.7	0.58	0.14
5	Jagraon	10.7	66.8	13.8	91	0.55	0.45	0.11

industrial waste from the districts that is allowed to mix in the soil, but a detailed investigation is needed for reaching this conclusion. The values reported for radium content in soils of study area are generally low as compared to the values reported by Sharma *et al.* (2003) for radium concentration in soils of Kangra, Himachal Pradesh. Radium concentrations are comparable with those reported by Singh *et al.* (2005) for Bathinda district of Punjab. Activity concentrations of ^{226}Ra and ^{232}Th are less than and the activity concentrations of ^{40}K are larger than those reported earlier in the Kullu area by Narayan Dass *et al.* (1979) and Kaul *et al.* (1993). However, a detailed investigation is required to reach at some conclusion.

The radium equivalent activity (Ra_{eq}) in these soil samples ranges from 200 Bq kg^{-1} (Jagraon) to 264 Bq kg^{-1} (Kauli) with mean value of 237 Bq kg^{-1} which is less than the safe limit (370 Bq kg^{-1}) recommended by OECD (1979). No regular trend in the variation of the terrestrial radioactivity has been observed from the study area. A detailed analysis of the results indicates that there is some degree of positive correlation between the activity concentrations of ^{232}Th and ^{40}K ($R = 0.25$) in the soil samples (Fig. 2). The R^2 value is the percentage of the total variance in ^{40}K explained by the independent variable of ^{232}Th, in the regression equation. Since, the value of R^2 signifies 6.3% of the total variance in ^{40}K is explained by ^{232}Th. Hence the value of R^2 is significant at 95% confidence level. Both the values of R and R^2 being low means presence of ^{232}Th and ^{40}K are independent of each other and one variable has nothing to do with the prediction or measurement of the other variable. The respective correlation coefficient is comparable to those reported for soils from Rajsthan (Nageswara Rao *et al.*, 1996).

The calculated total absorbed-dose and annual effective-dose rates of samples are shown in the Table 2. The International Commission on Radiological Protection (ICRP) has recommended the annual effective dose equivalent limit of 1 mSv Y^{-1} for the individual members of the public and 20 mSv Y^{-1} for the radiation workers (ICRP, 1993). Also, from Table 2 it is observed that the absorbed-dose rate calculated from activity concentration of ^{226}Ra, ^{232}Th and ^{40}K ranges between 10.8 and 20.1, 64.9 and 92.3, and 12.0 and 16.3 nGy h^{-1}, respectively. The total absorbed dose in the study area ranges from 91 nGy h^{-1} to 120 nGy h^{-1} with an average value of 108 nGy h^{-1}.

The corresponding indoor and outdoor annual effective-doses range from 0.45 to 0.59 mSv and 0.11 to 0.15 mSv having average values of 0.53 and 0.13 mSv, respectively; while the world-wide, average, annual, effective-dose is approximately 0.5 mSv, and the indoor results for individual countries are generally within the 0.3–0.6 mSv range. Generally similar results are observed in all the samples, and no regular trend in the variation in the annual effective dose and absorbed-dose rate is observed from the soil samples. Our results for average, annual, effective dose are with in the range of world wide average value.

The calculated values of H_{ex} for the soil samples studied range from 0.55 to 0.72 (Table 2). Because these values are lower than unity, and therefore, according to the Radiation Protection 112 (European Commission, 1999) report, soil from these regions is safe and can be used as a construction material without posing any significant radiological threat to population.

CONCLUSIONS

(i) The concentration of ^{232}Th in soil samples of Malwa

region of Punjab are higher than the world figures reported in UNSCEAR (2000). However, the concentrations for ^{226}Ra is very much comparable to and concentrations of ^{40}K are lower than world figures.

(ii) The results show that the indoor and outdoor effective doses from natural radioactivity of soil samples is lower than the average national and world-recommended value of 1.0 mSv Y^{-1}.

(iii) The values reported for radium content in soils of study area are generally lower than the values reported for radium concentration in soils of Himachal Pradesh.

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