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Radiological Risk Assessment in Soil Samples of Western Haryana, India

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***Abstract*—**The measurements of ^{226}Ra, ^{232}Th and ^{40}K using gamma spectrometry and radon concentration and exhalation rates measurements using solid state nuclear track (LR-115, Type-II plastic) detectors are used to asses a first order exposure risk for the persons residing in Fatehbad and Hissar districts of Western Haryana, India. The concentration of Radium, Thorium and Potassium in the soil samples varies from 13.37 Bq m^{-3} to 24.67 Bq m^{-3}, 34.67 Bq m^{-3} to 67.34 Bq m^{-3} and 298.78 Bq m^{-3} to 405.67 Bq m^{-3} respectively with average values of 18.78, 47.35 and 361.57 Bq m^{-3} respectively. The radium equivalent activity (Ra_{eq}) calculated for the same soil samples varies from 92.72 Bq m^{-3} to 140.6 Bq m^{-3} with an average value of 111.80 Bq m^{-3}. The values of absorbed dose and annual effective dose (indoors and outdoors) are found to vary from 44.18 nGy h^{-1} to 65.23 nGy h^{-1}, 0.22 mSv y^{-1} to 0.32 mSv y^{-1} and 0.05 mSv y^{-1} to 0.08 mSv y^{-1} respectively. The radon concentration and exhalation rates have also been reported. The radium equivalent activities in all the soil samples were found to be lower than the limit (370 Bq kg^{-1}) set in the Organization for Economic Cooperation and Development (OECD) report and the value of H_{ex} in all the samples is less than unity.

***Keywords*—**Gamma ray spectrometry, dose, radon, exhalation rate.

I. Introduction

EXTERNAL gamma dose estimation due to the 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 the concentrations of the 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 [1],[2]. The natural radioactivity of soil samples is usually determined from the ^{238}U, ^{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. The measurement of natural radioactivity due to gamma rays from the dose rate is needed to implement precautionary measures whenever the dose is found to be above the recommended limits. The growing worldwide interest in natural radiation exposure has lead to extensive surveys in many countries. External gamma dose estimation due to the 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 the concentrations of the natural radio nuclides, ^{226}Ra, ^{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 [1]-[4].

Radon (^{222}Rn) a decay product of radium in the naturally occurring uranium series is a radioactive inert gas and is responsible for about half of the radiation dose received by general population [5]. In all natural soils radon atoms are continuously produced in the α-decay chains of uranium and thorium. A fraction of them emanates into the air-filled pore volume of the soil and eventually escapes to the atmosphere. With the advancement of detector techniques in recent years it has become possible to monitor both ^{220}Rn and ^{222}Rn activities on a continuous basis in environmental samples. The respective half-lives of the two species (3.85 days for ^{222}Rn; 56 seconds for ^{220}Rn) mainly determine the application in environmental studies. Radon isotopes can isolate themselves and migrate away from the parent mineral due to diffusion process through the soil and enter the atmosphere. The radon and its progeny attached to aerosols present in the ambient air constitute significant radioactive hazards to human lungs. During respiration radon progeny deposits in the lungs and irradiate the tissue thereby damaging the cells and may cause lung cancer. Henshaw et al. [6] claimed that indoor radon exposure is associated with the risk of leukemia and certain other cancers, such as melanoma and cancers of the kidney and prostate. The concentration of radon and its decay products show large temporal and local fluctuations in the indoor atmosphere due to the variations of temperature, pressure, nature of building materials, ventilation conditions and wind speed etc.

Therefore, measurements of natural radioactivity in soil are of a great interest for many researchers throughout the world, which led to worldwide national surveys in the last two decades. To evaluate the terrestrial gamma dose rate for outdoor occupation, it is very important to estimate the natural radioactivity level in soils. The 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 though the data on the content of radioactive elements is available for some states of India, but the data available for Haryana is quite meagre. The detailed investigations for the measurement of natural radioactivity have been carried out for the first time. The main objective of the present study is to study the level of

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radioactive element viz. radium, thorium and potassium in Western region of Haryana for health risk assessment.

II. Materials and Methods

A. Measuring Activity Concentration of Radium, Thorium, Potassium

Soil samples were collected from the ten different locations of Fatehbad and Hissar districts of Western Haryana, India. The soil was collected from an auger hole at a depth of about 0.75 meters from the ground so as to get the natural soil. After collection, samples were crushed into fine powder by using mortar and pestle. Fine quality of the sample was obtained using scientific sieve of 150 micron-mesh size. Before measurement samples were dried in an oven at about 383^{o}K for 24 hours. Each sample was packed and sealed in an airtight PVC container and kept for about four week period to allow radioactive equilibrium among the radon (^{222}Rn), thoron (^{220}Rn), and their short lived progenies. On an average 0.25 kg. of soil was taken for each sample. For calibration of the low background counting system, a secondary standard was obtained, calibrated with the primary standard obtained from the International Atomic Energy Agency. The concentration of ^{226}Ra was determined using a photon peak of 609 keV (46.1%) from ^{214}Bi. The 186 keV photon peak of ^{226}Ra was not used because of interfering peak of ^{235}U, with energy of 185.7 keV. ^{232}Th concentration was determined using the gamma transitions of 583 keV (86%) from ^{208}Tl. ^{40}K concentration was determined using the gamma transition of 1461 keV (10.7%).

Using HPGe detector of high-resolution gamma spectrometry system, the activity of samples was counted. The detector was a co-axial n-type high purity germanium detector (Make EG&G, ORTEC, Oak Ridge, USA).The detector had a resolution of 2.0 keV at 1332 keV and relative efficiency of 20%. The output of the detector was analyzed using a 4K ADC system connected to PC, the spectrum was analyzed using the locally developed software "CANDLE (Collection and Analysis of Nuclear Data using Linux nEtwork)". The detector was shielded using 4" lead on all sides to reduce the background level of the system [4]. The efficiency calibration for the system was carried out using secondary standard source of uranium ore in geometry available for the sample counting. Efficiency values were 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. The samples were counted for a period of 72000 seconds and the spectra are analyzed of the photo peak of uranium, thorium daughter products and ^{40}K. 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 radionuclides was calculated from the background subtracted area prominent gamma ray energies.

B. Measurement of Radon Exhalation Rate

For radon exhalation measurements the samples were collected in clean, dry polyethylene bags. These samples were chosen to understand particularly the migration and exhalation of radon in the naturally occurring soils. A Glass bottle of about 1 litre capacity was used as an emanation chamber. The samples (0.25 kg) were placed at the bottom of the bottles, which were than closed for about 1 month in order to establish equilibrium between radium-radon members of the decay series. After one month the bottles were opened and LR-115 type-II plastic track detectors were suspended inside the bottles in a bare mode. The height of the detector is selected such as direct alpha particle does not reach the detectors. The bottles were than closed for about three month to record alpha activity under equilibrium conditions. After exposure the detectors were removed and etched in 2.5N NaOH solution at 60^{0}C for 90 minutes, using a constant temperature bath. The tracks were counted using Olympus microscope at a magnification of 400x. The resulting track density was then converted in Bqm^{-3} by appropriate calibration factor [7]. The 'radon exhalation rate' in terms of area is obtained from (1).

$$E_A = \frac{CV\lambda}{A[T+1/\lambda\,(e^{-\lambda T}-1)]} \quad (1)$$

where E_A is radon exhalation rate in terms of area ($mBq\ m^{-2}\ hr^{-1}$); C the integrated radon exposure as measured by LR-115 plastic track detector ($Bq\ m^{-3}\ hr$); V the effective volume of the bottle (m^3); λ the decay constant for radon (hr^{-1}); T the exposure time (hr); A the area of the bottle (m^2) [8]. This formula is also modified to calculate the radon exhalation rate in terms of mass ($mBq\ kg^{-1}\ hr^{-1}$).

$$E_M = \frac{CV\lambda}{M[T+1/\lambda\,(e^{-\lambda T}-1)]} \quad (2)$$

where E_M is radon exhalation rate in terms of mass ($mBq\ kg^{-1}hr^{-1}$) and M is the mass of the sample (0.25kg).

III. Results and Discussion

A. Estimation of Radium, Thorium and Potassium in Soil Samples

The concentrations of radium, thorium, and potassium in collected soil samples were calculated using (3).

$$Activity\ (Bq) = \frac{CPS \times 100 \times 100}{B.I. \times Eff} \pm \frac{CPS_{error} \times 100 \times 100}{B.I. \times Eff} \quad (3)$$

where, *CPS* = Net count rate per second
B.I. = Branching Intensity, and
Eff = Efficiency of the detector.

Table I summarizes the results of measurements of natural radionuclide (^{226}Ra, ^{232}Th and ^{40}K) concentrations in the collected soil samples. The errors shown in the results are the statistical uncertainties in track counting. 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. The concentration of radium, thorium and potassium in the soil samples varies from 13.37 Bq m^{-3} to 24.67 Bq m^{-3}, 34.67 Bq m^{-3} to 67.34 Bq m^{-3} and 298.78 Bq m^{-3} to 405.67 Bq m^{-3} respectively with average values of 18.78, 47.35 and 361.57 Bq m^{-3} respectively. Table I 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 [9]. However, the concentration for ^{40}K is very much comparable and a concentration for ^{226}Ra is lower as compared with world figures. Our values for radium content in soil lie in the range (2.5-207.0 Bq kg^{-1}) reported for Indian soils [10] using gamma ray spectrometry and are less than the permissible value (370 Bq kg^{-1}), which is acceptable as safe limit [11]. These values reported for radium content in soils of study area are generally low as compared to the values reported by Mehra et al. [4] for radium concentration in soils of Malwa region of Punjab. These values of radium concentration are comparable with those reported by Singh et al. [12] for Bathinda district of Punjab. The values of activity concentrations of ^{226}Ra and ^{232}Th are less and value of activity concentrations of ^{40}K is on the higher side than those reported earlier in Kullu area [13]. However, a detailed investigation is required to reach at some conclusion.

B. Evaluation of Radium Equivalent Activity

The distribution of natural radioactivity in the soil 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 materials containing different amount of ^{226}Ra, ^{232}Th, ^{40}K. About 98.5% of the radiological implications are due to ^{226}Ra and its daughter products in ^{238}U series. The natural abundance of ^{235}U is only 0.72% of the total uranium content and hence was not considered in the present study, except its ^{226}Ra contribution at 186 keV. The gamma transitions of energy 609 keV or 1760keV (due to ^{214}Bi) was used to determine the concentration of ^{226}Ra. The criterion for this model considers the external hazard due to gamma rays corresponds to a maximum radium equivalent activity of 370 $Bqkg^{-1}$ for the building material. This Ra_{eq} is calculated through (4)

$$Ra_{eq} = C_{Ra} + 1.43\, C_{Th} + 0.07\, C_K \quad (4)$$

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. While 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 [14].

Using above equation, the radium equivalent activity found in the soil samples is given in table1. The radium equivalent activity (Ra_{eq}) calculated for the same soil samples varies from 92.72 Bq m^{-3} to 140.6 Bq m^{-3} with an average value of 111.80 Bq m^{-3}. It is inferred that for all the soil samples analyzed, the radium equivalent activity value is well with in the permissible limits of 370 Bq kg^{-1}.

C. Estimation of Absorbed and Effective Dose

The external terrestrial γ-radiation absorbed dose rate in air at a height of about 1meter above the ground are calculated by using the conversion factor of 0.0414 nGy h^{-1} Bq^{-1} kg^{-1} for ^{40}K, 0.461 nGy h^{-1} Bq^{-1} kg^{-1} for ^{226}Ra, and 0.623 nGy h^{-1} Bq^{-1} kg^{-1} for ^{232}Th [15] assuming that ^{137}Cs, ^{90}Sr and the ^{235}U decay series can be neglected as they contribute very little to the total dose from environmental background [16]-[18].

$$D\ (nGyh^{-1}) = 0.461\, C_{Ra} + 0.623\, C_{Th} + 0.0414\, C_K \quad (5)$$

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

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 Sv Gy^{-1}, which is used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors [15]. The annual effective doses are determined as (6):

Indoor (nSv) = (Absorbed Dose) nGy^{-1} x 8760h x 0.8 x 0.7 $SvGy^{-1}$
Outdoor (nSv) = (Absorbed Dose) nGy^{-1} x 8760h x 0.2 x 0.7 $SvGy^{-1}$

(6)

The calculated total absorbed dose and annual effective dose rates of samples are shown in the Table II. 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 [19].

From Table II it is observed that the absorbed dose rate calculated from activity concentration of ^{226}Ra, ^{232}Th and ^{40}K ranges between 6.16 and 11.37, 21.60 and 41.95, and 12.37 and 16.79 nGy h^{-1}, respectively. The total absorbed dose in the study area ranges from 44.18 nGy h^{-1} to 65.23 nGy h^{-1} with an average value of 53.12 nGy h^{-1}.

The corresponding indoor and outdoor annual effective doses range from 0.22 to 0.32 mSv and 0.05 to 0.08 mSv with an average value of 0.26 and 0.07 mSv respectively while the world wide average annual effective dose is approximately 0.5 mSv and the results for individual countries being generally within the 0.3- 0.6 mSv range for indoors. Generally similar type of trend is observed in all the samples and no regular trend in the variation in the annual effective dose and absorbed dose rare is observed from the soil samples. Our results for average annual effective dose are with in the range of world wide average value.

D. External Hazard Index (H_{ex})

The external hazard index H_{ex} [20] can be calculated by (7).

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1 \quad (7)$$

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. The values of this index must be less than unity in order to keep the radiation hazard to be significant. The radiation exposure due to the radioactivity from a construction material is limited to 1.5mGy Yr^{-1}. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370Bq kg^{-1}). The calculated values of H_{ex} for the soil samples studied range from 0.26 to 0.39 (Table 2). Since these values are lower than unity, therefore, according to the Radiation Protection 112 report [21], soil from these regions is safe and can be used as a construction material without posing any significant radiological threat to population.

E. Radon Concentration and Radon Exhalation Rate Measurements

The calculated values of radon concentration and exhalation rate for soil samples are presented in Table III. The radon concentration varies from 223.78 Bq m^{-3} to 413.42 Bq m^{-3} and radon exhalation rate varies from 6.32 mBq kg^{-1} h^{-1} to 11.68 mBq kg^{-1} h^{-1} for mass exhalation rate and from 139.55 mBq m^{-2} h^{-1} to 257.81 mBq m^{-2} h^{-1} for surface exhalation rate in soil samples. These values are lower than those reported by Mehra et al. [3] for the soils of adjoining Malwa region of Punjab. The variation in values of radon exhalation rate may be due to the difference in radium content [22] and porosity of the soil [23].

TABLE I
ACTIVITY CONCENTRATION OF RADIUM, THORIUM, POTASSIUM, RADIUM EQUIVALENT

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 Ra_{eq} (Bq kg^{-1})
1	Bhattu Kallan	14.23±0.23	41.67±2.19	405.67±9.34	102.22
2	Sampla	16.43±0.19	45.65±2.34	398.23±8.69	109.59
3	Bhodiya Khera	22.43±0.25	67.34±1.98	312.45±9.81	140.6
4	Dhanger	24.67±0.26	56.78±2.39	384.34±8.04	132.77
5	Jandli Khurd	20.34±0.18	35.95±2.45	299.57±8.91	92.72
6	Pawra	20.21±0.19	45.93±2.96	376.64±9.89	112.25
7	Nehla	17.78±0.27	34.67±1.99	398.78±10.43	95.27
8	Barwala	19.97±0.16	51.42±2.24	384.34±8.30	120.4
9	Dhasnsu	18.34±0.21	39.67±2,71	298.78±9.10	95.98
10	Ganguwa	13.37±0.18	54.43±1.93	356.87±8.49	116.19

TABLE II
RADIATION ABSORBED DOSE AND ANNUAL EFFECTIVE DOSE FROM SOIL SAMPLES

Sr. No.	Sample location (Village)	Absorbed dose (nGy h^{-1})				External Hazard Index H_{ex}	Annual effective dose(m Sv)	
		Ra	Th	K	Total		Indoor	Outdoor
1	Bhattu Kallan	6.56	25.96	16.79	49.31	0.28	0.24	0.06
2	Sampla	7.57	28.44	16.49	52.5	0.3	0.26	0.06
3	Bhodiya Khera	10.34	41.95	12.94	65.23	0.39	0.32	0.08
4	Dhanger	11.37	35.37	15.91	62.65	0.37	0.31	0.08
5	Jandli Khurd	9.38	22.4	12.4	44.18	0.26	0.22	0.05
6	Pawra	9.32	28.61	15.59	53.52	0.31	0.26	0.07
7	Nehla	8.2	21.6	16.51	46.31	0.26	0.23	0.06
8	Barwala	9.21	32.03	15.91	57.15	0.33	0.28	0.07
9	Dhasnsu	8.45	24.71	12.37	45.53	0.26	0.22	0.06
10	Ganguwa	6.16	33.91	14.77	54.84	0.32	0.27	0.07

TABLE III
RADON CONCENTRATION AND RADON EXHALATION RATES FROM SOIL SAMPLES

Sr. No.	Sample location (Village)	Radon Concentration (Bqm^{-3})	Radon Exhalation Rates	
			Surface Exhalation Rate ($mBqm^{-2}h^{-1}$)	Mass Exhalation Rate ($mBqkg^{-1}h^{-1}$)
1	Bhattu Kallan	289.45	180.50	8.18
2	Sampla	342.89	213.83	9.69
3	Bhodiya Khera	413.42	257.81	11.68
4	Dhanger	389.34	242.80	11.01
5	Jandli Khurd	238.91	148.99	6.75
6	Pawra	413.32	257.75	11.68
7	Nehla	298.43	186.10	8.44
8	Barwala	330.9	206.35	9.35
9	Dhasnsu	223.78	139.55	6.32
10	Ganguwa	293.45	183.00	8.30

IV. CONCLUSION

In general, the values 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 ^{40}K is very much comparable and a concentration for ^{226}Ra is lower as compared with world figures. The results obtained have shown that the indoor and outdoor effective dose due to natural radioactivity of soil samples is lower than the average national and world recommended value of 1.0 $mSv.Y^{-1}$. The calculated values of H_{ex} for the soil samples are lower than unity, therefore, according to the Radiation Protection 112 report [21], soil from these regions is safe and can be used as a construction material without posing any significant radiological threat to population.

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