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Natural radioactivity in Indian vegetation samples

K. Kant[1*], R. Gupta[2], R. Kumari[3], N. Gupta[4], M. Garg[5]

[1]*Department of Physics, Aggarwal College Ballabgarh, Haryana, India– 121 004*
[2]*Department of Physics and Astro Physics, University of Delhi, Delhi, India*
[3]*Department of Physics, Aggarwal College Ballabgarh, Distt. Faridabad, Haryana, India*
[4]*Department of Humanities and Applied Sciences, AITM, Palwal Haryana, India*
[5]*Faculty of Humanities and Applied Sciences, YMCAUST, Faridabad (Haryana), India*

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**** Corresponding author:***
Dr. Krishan Kant,
Fax: *+91 129 2300769*
E-mail:
kkant_67@rediffmail.com

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ABSTRACT

***Background*:** Vegetation (food stuff) is grown in soil that contains many radioactive elements such as ^{238}U (uranium), ^{232}Th (thorium) and ^{40}K (potassium), which may get deposited either due to radioactive fallout or/and by absorption from the soil and can pose serious health hazards. ***Materials and Methods*:** Natural radioactivity, radiological hazards and annual effective dose assessment was carried out in vegetation samples (vegetables, cereals and fruits) collected from fields and market. Gamma spectrometry using HPGe detector was used. ***Results*:** The measured specific activity concentration of ^{238}U (uranium), ^{232}Th (thorium) and ^{40}K (potassium) varied from 10.25 ± 0.94 Bq/kg to 29.13 ± 0.69 Bq/kg, 22.20 ± 2.46 Bq/kg to 58.21 ± 1.15 Bq/kg, and 1158.4 ± 26.05 Bq/kg to 1962.2 ± 18.17 Bq/kg respectively in various vegetable and cereal samples and varied from 2.5 ± 0.16 Bq/kg to 9.8 ± 0.15 Bq/kg, 7.4 ± 1.24 Bq/kg to 18.4 ± 1.39 Bq/kg, and 287.13 ± 11.23 Bq/kg to 815.72 ± 12.50 Bq/kg respectively in various fruit samples studied in the present work. From these values, hazard indices, the minimum and maximum values of absorbed dose and indoor and outdoor annual effective doses were calculated for various samples used in the present investigation. ***Conclusion*:** The various values obtained were found to be within the recommended limits. The absorbed dose and annual effective dose for the vegetable and cereal samples in which fertilizers were used to enhance the crop yield were higher than that in fruit samples.

Keywords: *Radiological hazards, vegetation, radium equivalent activity.*

INTRODUCTION

We live on and in a radioactive world and are 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 [1-3]. Our soil, in which our food stuff is grown, contains many radioactive elements. There are several radioactive elements in this category, such as ^{238}U (uranium), ^{232}Th (thorium) and ^{40}K (potassium), also termed as primordial radioisotopes, for they were present when the earth was formed. Natural radioactivity in soils comes from ^{238}U and ^{232}Th series and natural ^{40}K. Uranium occurs in minerals such as pitchblende, uraninite etc. It is also found in phosphate rocks, lignite and monazite sands, whereas ^{40}K is present in mineral waters and brines, and in various minerals such as carnallite, feldspar, saltpeter, greens and sylvite [4-6]. ^{238}U and ^{232}Th give rise to the radium and thorium atoms found in all humans, acquired from the food we eat and that food, of course, obtained these materials from the soil in which it grew or on which it grazed [7]. Potassium is also in this category and ^{40}K- a naturally occurring radioactive isotope (isotope of interest in the present study) comprises a very

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small fraction (about 0.0118%) of naturally occurring potassium; its concentration in the earth's crust is about 1.8 mg/kg, or 13 picocurie per gram (pCi/g) [8]. It is the predominant radioactive component in human tissues and in most food [9-13] and is an important radionuclide in terms of the dose associated with naturally occurring radionuclides. The growing worldwide interest in natural radiation exposure has lead to extensive surveys in many countries. External gamma dose estimation due to terrestrial sources is essential not only because it contributes considerably (0.46mSv/y) to the collective dose but also because of the variations of the individual doses related to this pathways. The doses vary depending upon the concentration of the natural radionuclides, ^{238}U, ^{232}Th and their daughter products and ^{40}K, present in food stuffs, which in turn depend upon the local geology of each region in the world [14-18]. The health hazard of potassium-40 is associated with cell damage caused by the ionizing radiation that results from radioactive decay, with the general potential for subsequent cancer induction. Thus measurement of radioactivity in potassium enriched food stuffs is very important from radiation protection point of view and in the present work we have reported the radioactivity in potassium enriched vegetation.

MATERIALS AND METHODS

Sample collection and preparation

In the present investigation, samples (commercially available in the market and from the fields) of different types of vegetation were collected and analyzed for the activity concentration of the natural radionuclides namely ^{238}U, ^{232}Th and ^{40}K using gamma spectrometry as carried out by other workers [19]. Subsequently radium equivalent was calculated.

The samples were powdered, charred under a low flame, and then ashed at 450 °C to get a uniform white ash in muffle furnace. The samples were shaken in a sieve of 250 micron-mesh size and particles of size ≤ 250 microns were obtained. Vegetation samples were dried for 10–15 h at 110 °C in an electric oven (hot air oven) to obtain a constant dry weight. Sieved samples were packed and sealed in 300 ml air tight PVC container and kept for about four weeks period to allow radioactive equilibrium among the radon (^{222}Rn), thoron (^{220}Rn), and their short lived progenies. On an average 0.3 kg of ash 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 an energy of 185.7 keV. ^{232}Th concentration was determined using the gamma transitions of 583 keV (86%) from ^{208}Tl [20]. ^{40}K concentration was determined using the gamma transition of 1461 keV (10.7%).

Measurement technique

Using HPGe detector of high-resolution gamma spectrometry system available at inter university accelerator center (IUAC), New Delhi, the activity of samples is counted. The detector is a co-axial n-type high purity germanium detector (Make EG&G, ORTEC, Oak Ridge, USA). The detector has a resolution of 2.0 keV at 1332 keV and relative efficiency of 20%. 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 (**C**ollection and **A**nalysis of **N**uclear **D**ata using **L**inux n**E**twork)". The detector is shielded using 4'' lead on all sides to reduce the background level of the system [21]. The efficiency calibration for the system is carried out using secondary standard source of uranium ore in geometry 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. The samples were counted for a period of 72000 seconds and the

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spectra were 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 are calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclides is calculated from the background subtracted area prominent gamma ray energies.

Formulas used in measurements

Measuring activity concentration of uranium, thorium and potassium

The concentrations of Uranium, Thorium and Potassium were calculated using the following equation:

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

Where, CPS - Net count rate per second
B.I. - Branching Intensity, and
Eff - Efficiency of the detector.

(ii) Radium equivalent activity

The widely used radiation hazard index Ra_{eq} is called the radium equivalent activity; the radiation equivalent activity is a weight sum of activities of the three natural radionuclides ^{238}U, ^{232}Th and ^{40}K based on the estimation that 10 Bq/kg of ^{226}Ra, 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce the same gamma ray dose rate [22].

The radium equivalent activity was calculated using the following equation

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_k \quad (2)$$

Where, C_u, C_{Th} and C_k are the specific activities of ^{238}U, ^{232}Th and ^{40}K in Bq/ kg respectively.

Assessment of radiological hazards

As more than one radionuclide contributes towards the gamma doses (i.e. ^{226}Ra, ^{232}Th and ^{40}K), therefore radiological hazards have been presented in terms of a single quantity called 'hazard index' and the measured specific activity concentrations have been used to assess the radiological hazards in terms of external hazard index (Hex), Internal hazard index (H_{in}), Gamma index (I_γ) and Alpha index (I_α) using equations 3, 4, 5 and 6 respectively [22-24].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (4)$$

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad (5)$$

$$I_\alpha = \frac{A_{Ra}}{200} \quad (6)$$

To limit the external gamma radiation dose from materials below 1.5 mGy/y, the external hazard index, H_{ex} should obey the following relation $H_{ex} \leq 1$ [25].

(iv) Estimation of absorbed and annual effective dose

The measured activity concentrations of ^{238}U, ^{232}Th and ^{40}K were converted into doses ($nGyh^{-1}$ per $Bqkg^{-1}$) by applying the factors 0.462, 0.604 and 0.0417 for uranium, thorium and potassium, respectively [26]. These factors were used to calculate the total absorbed gamma dose rate in air at one meter above the ground level using the following equation:

$$D\ (nGyh^{-1}) = (0.462\ C_U + 0.604\ C_{Th} + 0.0417\ C_K) \quad (7)$$

Where, C_U, C_{Th} and C_K are the activity concentrations (Bq/kg) of uranium, 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, which is used to convert the absorbed dose rate to annual effective dose with an outdoor occupancy of 20% and 80% for indoors [27].

The annual effective doses are determined as follows:

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$$\text{Indoor (mSv)} = (\text{Absorbed Dose})\text{nGyh}^{-1} \times 8760\text{h} \times 0.8 \times 0.7\text{SvG y}^{-1} \quad (8)$$

$$\text{Outdoor (mSv)} = (\text{Absorbed Dose})\text{nGyh}^{-1} \times 8760\text{h} \times 0.2 \times 0.7\text{SvG y}^{-1} \quad (9)$$

RESULTS

The concentration of the radionuclides, ^{238}U, ^{232}Th and ^{40}K, and radium equivalent activity calculated using equations 1 & 2 in the vegetable and cereal samples studied in the present investigation are given in table 1 and in fruit samples in table 2 respectively. From table 1, it is clear that the activity concentration for ^{238}U, ^{232}Th and ^{40}K varies from 10.25 ± 0.94 Bq/kg to 29.13 ± 0.69 Bq/kg, 22.20 ± 2.46 Bq/kg to 58.21 ± 1.15 Bq/kg, and 1158.4 ± 26.05 Bq/kg to 1962.2 ± 18.17 Bq/kg respectively in various vegetable and cereal samples studied in the present work. The radium equivalent activity varied from 177.78 Bq/kg to 252.07 Bq/kg in the vegetable and cereal samples used in the present investigation. The table 2 shows that the activity concentration for ^{238}U, ^{232}Th and ^{40}K varies from 2.5 ± 0.16 Bq/kg to 9.8 ± 0.15 Bq/kg, 7.4 ± 1.24 Bq/kg to 18.4 ± 1.39 Bq/kg, and 287.13 ± 11.23 Bq/kg to 815.72 ± 12.50 Bq/kg respectively in various fruit samples studied in the present work. The radium equivalent activity varied from 47.91 Bq/kg to 80.02 Bq/kg in fruit samples used in the present investigation.

The radiological hazards in terms of external hazard index (Hex), Internal hazard index (H_{in}), Gamma index (I_γ) and Alpha index (I_α) for the samples calculated using equations 3, 4, 5 & 6 are given in table 3 and 4. From table 3 and 4, it is clear that external hazard indices varies from 0.48 to 0.68 in various vegetable and cereal samples and varies from 0.13 to 0.22 in various fruit samples; the internal hazard index varies from 0.54 to 0.73 in various vegetable and cereal samples and varies from 0.14 to 0.24 in various fruit samples, the gamma index varies from 0.69 to 1.00 in various vegetable and cereal samples and varies from 0.19 to 0.32 in various fruit samples, the alpha index varies from 0.05 to 0.15 in various vegetable and cereal samples and varies from 0.01 to 0.05 in various fruit samples studied in the present work.

The radiation absorbed dose and annual effective dose from vegetable and cereal samples and from fruit samples calculated using equations 7, 8 & 9 are given in table 5 and table 6 respectively. The table 5 shows that the minimum and maximum values of absorbed dose, indoor annual effective dose and outdoor annual effective dose varied from 86.83 nGyh^{-1} to 125.18 nGyh^{-1}, from 0.425 mSv to 0.614 mSv and from 0.106 mSv to 0.153 mSv respectively in various vegetable and cereal samples under investigation. Table 6 shows that the calculated values of absorbed dose and annual effective dose (indoors and outdoors) varied from 23.39 nGyh^{-1} to 40.10 nGyh^{-1}, 0.114 mSv/y to 0.196 mSv/y and 0.028 mSv/y to 0.049 mSv/y, respectively in fruit samples.

Table 1. Activity Concentration of uranium, thorium and potassium in vegetable and cereal samples.

Sr. No.	Sample Code	Activity Concentration (Bq/kg)			Radium Equivalent Activity (Ra_{eq}) (Bq/kg)
		^{238}U	^{232}Th	^{40}K	
1	VC-1	28.08 ± 1.28	42.31 ± 2.17	1158.4 ± 26.05	177.78
2	VC -2	15.55 ± 1.77	57.52 ± 2.34	1579.2 ± 17.58	219.40
3	VC -3	22.72 ± 1.63	47.32 ± 1.35	1268.5 ± 15.05	188.06
4	VC -4	29.13 ± 0.69	22.20 ± 2.46	1885.1 ± 28.03	206.03
5	VC -5	28.22 ± 1.62	26.48 ± 1.41	1590.9 ± 19.21	188.59
6	VC -6	10.25 ± 0.94	36.37 ± 1.20	1665.4 ± 17.63	190.49
7	VC -7	19.94 ± 0.40	49.17 ± 1.31	1820.6 ±27.24	230.44
8	VC -8	17.74 ± 1.59	58.21 ± 1.15	1962.2 ± 18.17	252.07
9	VC -9	18.38 ± 1.15	50.82 ± 1.17	1698.5 ± 28.04	221.84
10	VC -10	11.40 ± 0.90	39.85 ± 2.32	1775.0 ± 26.48	205.06
11	VC -11	19.21 ± 1.61	26.53 ± 1.89	1625.2 ± 18.28	182.29

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Table 2. Activity concentration of uranium, thorium, potassium in fruit samples.

Sr. No.	Sample Code	Activity Concentration (Bq/kg)			Radium Equivalent Activity (Ra_{eq}) (Bq/kg)
		^{238}U	^{232}Th	^{40}K	
1	FR-1	8.1 ± 0.15	12.3 ± 1.49	687.13 ± 12.23	78.60
2	FR-2	2.5 ± 0.16	15.1 ± 1.67	554.74 ± 14.09	66.81
3	FR-3	3.9 ± 0.25	13.0 ± 1.32	487.13 ± 11 .23	60.00
4	FR-4	4.6 ± 0.23	09.6 ± 1.22	384.13 ± 9.23	47.91
5	FR-5	9.8 ± 0.15	11.4 ± 1.29	287.13 ± 11.23	48.21
6	FR-6	5.6 ± 0.15	13.4 ± 1.76	437.56 ± 12.39	58.45
7	FR-7	4.6 ± 0.29	08.3 ± 1.53	737.39 ± 9.37	73.25
8	FR-8	3.5 ± 0.27	07.4 ± 1.24	815.72 ± 12.50	76.89
9	FR-9	8.5 ± 0.12	18.4 ± 1.39	587.13 ± 16.23	80.02
10	FR-10 (Mixed)	7.4 ± 0.18	08.1 ± 1.34	511.25 ± 8.17	58.35

MADL (Minimum Activity Detection Limit) = 2Bq/kg, 2Bq/kg and 4Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K respectively.

Table 3. External hazard index, internal hazard index, gamma index and alpha index in vegetable and cereal samples.

Sr. No.	Sample Code	H_{ex}	H_{in}	I_γ	I_α
1	VC-1	0.48	0.57	0.69	0.14
2	VC -2	0.59	0.64	0.87	0.08
3	VC -3	0.51	0.59	0.74	0.11
4	VC -4	0.56	0.54	0.84	0.15
5	VC -5	0.51	0.68	0.76	0.14
6	VC -6	0.51	0.73	0.77	0.05
7	VC -7	0.62	0.65	0.92	0.10
8	VC -8	0.68	0.58	1.00	0.09
9	VC -9	0.60	0.54	0.88	0.09
10	VC -10	0.55	0.57	0.83	0.06
11	VC -11	0.49	0.64	0.74	0.10

Table 4. External hazard index, internal hazard index, gamma index and alpha index in fruit samples.

Sr. No.	Sample Code	H_{ex}	H_{in}	I_γ	I_α
1	FR-1	0.21	0.23	0.32	0.04
2	FR-2	0.18	0.19	0.27	0.01
3	FR-3	0.16	0.17	0.24	0.02
4	FR-4	0.13	0.14	0.19	0.02
5	FR-5	0.13	0.16	0.19	0.05
6	FR-6	0.16	0.17	0.23	0.03
7	FR-7	0.20	0.21	0.30	0.02
8	FR-8	0.21	0.22	0.32	0.02
9	FR-9	0.22	0.24	0.32	0.04
10	FR-10 (Mixed)	0.16	0.18	0.24	0.04

Table 5. Radiation absorbed dose and annual effective dose from vegetable and cereal samples.

Sr. No.	Sample Code	Absorbed Dose Rate ($nGyh^{-1}$)	Annual Effective Dose (mSv)	
			Indoor	Outdoor
1	VC-1	86.83	0.425	0.106
2	VC -2	107.78	0.528	0.132
3	VC -3	91.97	0.451	0.112
4	VC -4	105.48	0.517	0.129
5	VC -5	95.37	0.467	0.117
6	VC -6	96.15	0.471	0.118
7	VC -7	114.83	0.563	0.141
8	VC -8	125.18	0.614	0.153
9	VC -9	110.01	0.539	0.135
10	VC -10	103.35	0.507	0.126
11	VC -11	92.67	0.454	0.113

Table 6. Radiation absorbed dose and annual effective dose from fruit samples.

Sr. No.	Sample Code	Absorbed Dose Rate ($nGyh^{-1}$)	Annual Effective Dose (mSv)	
			Indoor	Outdoor
1	FR-1	39.82	0.195	0.048
2	FR-2	33.41	0.163	0.041
3	FR-3	29.97	0.147	0.036
4	FR-4	23.94	0.117	0.029
5	FR-5	23.39	0.114	0.028
6	FR-6	28.93	0.141	0.035
7	FR-7	37.89	0.185	0.046
8	FR-8	40.10	0.196	0.049
9	FR-9	39.52	0.193	0.048
10	FR-10 (Mixed)	29.63	0.145	0.036

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DISCUSSION

From the results obtained it is clear that the concentration of Radium, Thorium and Potassium and the radium equivalent activity varied considerably in different samples depending on the quantity and type of fertilizer used in the fields. From the data, it reveals that the activity concentration and the radium equivalent activity in the fruit samples in which no fertilizer/less fertilizer was used, is significantly lower than that in the vegetable and cereal samples collected from market and crop fields where fertilizers were used to enhance the crop yield. Our findings are in good agreement with the findings of other researchers reported in the literature [26, 28-33]. The concentration of the radionuclide ^{40}K is higher in all the vegetable and cereal samples than fruit samples. It is inferred that for all the samples analyzed, the radium equivalent activity value is well within the permissible limits of 370Bq/kg [26,34].

The external hazard index and internal hazard index for the studied samples is less than unity and therefore these vegetations are safe to be used for consumption. Value of gamma index $I_\gamma \leq 2$ corresponds to a dose rate criterion of 0.3 mSv/y, whereas $2 \leq I_\gamma \leq 6$ corresponds to a criterion of 1 mSv/y [23, 35]. Thus the vegetation with $I_\gamma > 6$ should be avoided to be used, since these values correspond to the dose rates higher than 1 mSv/y which is higher than the recommended safe limit values [26]. All the current 'I_γ' values of the studied samples follow the criterion ($I_\gamma \leq 2$) therefore it may be concluded that the samples are safe from health and hygiene point of view and don't pose any significant health hazards to the consumers. The recommended limit for concentration of ^{226}Ra is 200 Bq/kg, for which $I_\alpha = 1$ [36]. The observed values are less than unity showing that the samples are safe from the health and hygiene point of view and don't pose any environmental radiation hazards.

In all the samples, the indoor annual effective dose was less than the recommended safety limit of 1 mSy/y as recommended for general public [37]. The positive correlation was observed between the uranium activity and the absorbed dose. The absorbed dose and annual effective dose for the vegetable and cereal samples in which fertilizers are used to enhance the crop yield are higher than that in fruit samples.

CONCLUSION

The present investigations showed the inborn radioactivity in vegetation samples was modified during technological enhancement (use of fertilizers to enhance the crop yield) but all the samples were found to satisfy the safety criteria. Efforts should be made at national and international level to reduce ^{226}Ra activity in the fertilizers, like extracting uranium from phosphoric acid by solvent extraction method, so that the fertilizers are more eco-friendly and the radioactivity in the soil itself is not technologically enhanced.

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Conflicts of interest: *none to declare.*

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