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DISTRIBUTION OF NATURAL RADIONUCLIDES IN SOIL SAMPLES OF QUAID-I-AZAM UNIVERSITY, ISLAMABAD, PAKISTAN

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The concentration of natural gamma-emitting radionuclides of the uranium and thorium series as well as ^{40}K and a fission product ^{137}Cs have been determined by gamma-ray spectroscopy in soil samples collected from various locations of Quaid-i-Azam University (QAU) campus, Islamabad and its peripheries. For detection, analysis and data acquisition a high purity germanium detector (HPGe) coupled with a multichannel analyzer was used. The mean specific activity, in Bq kg^{-1} dry mass of soil samples, due to ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs is 45.5±15.9, 55.2±20.9, 355.7±123.8 and 7.1±4.9 respectively. The average value of Radium equivalent activity was found to be 151.9 Bq kg^{-1}. The external and internal hazard indices were below unity. The average value of absorbed dose rate was 65 nGyh^{-1}. The results show that the soil of QAU do not pose any significant environmental or radiological health problem.

Keywords: Soil samples, Natural radioactivity, Gamma spectroscopy, Radium equivalent activity, Hazard indices, Absorbed dose rate.

1. Introduction

Radionuclides have been essential constituents of the earth since its creation and man is continuously exposing to the ionizing radiations from these radionuclides [1]. The fallout from the frequent nuclear weapon tests in the past and releases of radioactive materials from different nuclear installations have resulted in the increase of radioactivity level in the environment [2]. Therefore, knowledge of the distribution pattern of both anthropogenic and natural radionuclides is essential in maintaining some sense of control of prevailing radiation levels [3].

Natural radioactivity is present everywhere and all rocks; soils and minerals contain naturally occurring radionuclides such as ^{40}K, ^{238}U, ^{232}Th and their progeny [4]. In addition, a large scale radiation exposure can also occur as a result of major nuclear accident. A well known example is the Chernobyl nuclear reactor accident, which resulted in a release of large quantities of radioactive materials into the environment. The most significant radionuclides found in environmental samples were the isotopes of iodine (^{131}I, ^{132}I) and cesium (^{134}Cs, ^{137}Cs). Since half-life of ^{131}I is only 8 days, therefore, during first week of the accident it was of great concern. On the other hand ^{137}Cs is a long-lived radionuclide (half life = 30 years), hence its measurement in various environmental samples can be made even after several years.

The objective of the present study is to determine the natural and man-made radioactivity and associated radiation doses to general public. Similar studies have also been carried out by other researchers for different building materials and environmental samples of Pakistan [5-7]. Present investigation reports the measurement of specific activities of the decay products of ^{238}U and ^{232}Th series and of primordial radionuclides ^{40}K and a fission product ^{137}Cs in soil samples collected from different areas of Quaid-i-Azam University, Islamabad. Results were also compared with similar work carried out for other areas of Pakistan as well as for other countries of the world.

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2. Experimental

2.1. Sampling

Soil samples were collected from ten different sites of Quaid-i-Azam University, Islamabad and were marked as S-I to S-X. Sampling sites were chosen to be relatively flat, open and undisturbed. The sloppy areas where abnormal surface run-off may occur were avoided. The samples were collected by grabbing the top 5 cm layer of the soil. All the samples were packed in polythene bags and were properly labeled before further treatment.

2.2 Sample pretreatment

The soil samples were oven dried at 110 °C, until the sample weight became constant. After complete dryness, these were homogenized by mechanical treatment and passed through 2mm mesh sieve. The samples were properly labeled and packed in plastic container identical in shape and size to that of standard reference material (IAEA soil 375). These samples were properly sealed and stored for 40 days to enable an approach to an equilibrium of ^{226}Ra with its decay products in the uranium series and ^{228}Ra with its daughters in thorium series .[8,9]

2.3 Calibration of instrument

In order to measure the gamma-ray energies and absolute activities of samples under study, the spectrometer must be calibrated both in terms of energy as well as efficiency. Normally calibration is done with standard sources and reference material of known activity. In the present study, a standard reference material soil 375 from IAEA for gamma activity determination in soil was used.

2.4 Energy calibration

Energy calibration of the system is necessary to correctly identify the radionuclides within a spectrum. It is based on a computer analysis method which matches the energies of the principal gamma rays observed in the spectrum to the energies of gamma rays emitted by the known radionuclides. The ideal relation between gamma-ray energy in keV (E) and channel number (N) of multichannel analyzer (MCA) is given by the straight line equation [10].

$$E = 2.7+2.1e^{-1} x\ N \quad (1)$$

2.5 Efficiency calibration

An accurate efficiency calibration of the system is necessary to quantify radionuclides present in the sample, which must be known as a function of gamma-rays. The efficiency can be calculated only if radioactive source of the known strength emitting a spectrum of gamma-rays are available in the desired shape and geometry. For this purpose, soil-375 was used. Detection efficiencies of the system between 0-2000 keV energy were calculated by noting the areas under the photo peaks from the spectrum of soil-375. The software used third degree polynomials to fit the efficiency curve as [10].

$$\text{Ln}\ \eta = -\ 5e^{1} + 2.4e^{1} \text{ x ln } E - 3.9 \text{ x ln } E^{2} + 2e^{-1} \text{ x ln } E^{3} \quad (2)$$

2.6 Radiometric measurements

For the measurement of radioactivity, a Coaxial type High Purity Germanium (HPGe) detector, coupled with a PC based MCA (Accuspec-A, Canberra) was used. The relative efficiency of the detector was 30 % with 180 cm^3 active volume and the resolution of the system was 2.3 keV for 1332.5 keV gamma-ray of ^{60}Co. The detector shield had an internal cavity of 55 cm^3. The detector head was placed almost at the centre of the cavity. The shield consisted of 12 mm thick layer of copper and a 50 mm thick layer of lead. The function of copper is to absorb the X-rays of lead. The results were analyzed by Genie-2000 software. The measurement time for each sample was kept 65000 seconds. Soil-375 was used for efficiency calibration of detector. The analyses of ^{40}K and ^{137}Cs were based upon their single peaks of 1460.2 keV and 661 keV respectively. However, the analysis of ^{226}Ra and ^{232}Th depended upon the peaks of daughter products in equilibrium with their parent radionuclides. The values of lower limit of detection for all the detected radionuclides are cited in Table 1.

3. Results and Discussion

The specific activities (Bq kg^{-1}) of natural and anthropogenic radionuclides present in soil have been summarized in Table 2. The range of specific activities for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs was found to be from 23.4–67.5, 27.5–83.8, 183.6–562 and 0.51-17.2 respectively. The table shows that the trend of specific activities in individual samples is not uniform, but varies from sample to sample and location to location. This non-uniform behavior of radionuclides may be attributed to their un-even and irregular distribution in the earth crust and also to various topographical and agricultural activities [11]. Since concentration and distribution of

Table 1. Lower limit of detection (LLD) of the detected radionuclides.

S. No.	Parent Radionuclide	Daughter Radionuclide	Energy (Kev)	LLD (Bq)
1.	^{226}Ra	^{214}Pb	186	1.42
		^{214}Bi	351	0.35
			609	0.48
2.	^{232}Th	^{228}Ra	338	0.49
		^{228}Ac	911.1	0.83
		^{228}Ac	968.9	1.11
		^{208}Tl	583.1	0.19
		^{212}Pb	238	0.20
3.	^{40}K	–	1460	7.24
4.	^{137}Cs	–	661	0.14

Table 2. Specific activity of gamma-emitting radionuclides in soil samples of QAU campus, Islamabad.

Sr. No.	Sample Code No.	Specific Activity(Bqkg^{-1})			
		^{226}Ra	^{232}Th	^{40}K	^{137}Cs
1	S-I	49±0.8	52.5±0.6	388.4±8.3	12.5±0.3
2	S-II	58.2±0.9	63.1±0.6	302.4±12.2	5.4±0.3
3	S-III	67.5±0.9	95.9±0.8	562±13.2	3.5±0.3
4	S-IV	26.3±0.7	36.2±0.5	236.7±12	2.9±0.2
5	S-V	23.4±0.7	27.5±0.5	223.2±12	0.5±0.2
6	S-VI	44.2±0.8	55.6±2.3	523.7±13	7.7±0.3
7	S-VII	66.8±0.9	83.8±0.7	359.4±12	9.1±0.3
8	S-VIII	33.8±0.7	43.9±0.5	398.1±12	5.4±0.3
9	S-IX	35±0.8	46.9±0.6	183.6±11.8	6.9±0.3
10	S-X	51.±1.6	47.4±0.5	369.1±13	17.2±0.4
Average		45.5±16	55.2±21	355.7±123.8	7.1±4.9

radionuclides is not uniform throughout the world so uniformity in respect of exposure to radiation has been defined in terms of Radium equivalent activity (Ra_{eq}), internal hazard index (H_{in}), external hazard index (H_{ex}) and absorbed dose rate (D).

The term Ra_{eq} was introduced by Beretka and Mathew and is defined as [12,13]:

$$Ra_{eq} = C_{Ra} + 1.43\, C_{Th} + 0.077\, C_K \qquad (3)$$

where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K respectively.

While defining Ra_{eq} it has been assumed that 10 Bq kg^{-1} of ^{226}Ra, 7 Bq kg^{-1} of ^{232}Th and 130 Bq kg^{-1} of ^{40}K produce equal gamma-ray dose. The Ra_{eq} is related with external gamma-ray dose and internal dose due to radon and its daughters. Also, they defined a Ra_{eq} value of 370 Bqkg^{-1} as maximum allowed value for public dose consideration.

Table. 3. The Ra_{eq} activity, hazard indices and absorbed dose rate of soil sample.

Sample Code No.	Radium Eq. Activity Ra_{eq} (Bq kg^{-1})	Hazard Indices					Absorbed Dose Rate ($nGyh^{-1}$)
		H_{ex}	$H_{in(I)}$	$H_{in(II)}$	$H_{in(III)}$	$H_{in(IV)}$	
S-I	154	0.41	0.54	0.61	0.16	0.55	12.4
S-II	171.7	0.46	0.62	0.62	0.18	0.61	79.6
S-III	248	0.65	0.83	0.92	0.25	0.87	113.4
S-IV	96.3	0.26	0.33	0.36	0.10	0.35	45.4
S-V	80	0.21	0.28	0.31	0.08	0.29	37.8
S-VI	164	0.44	0.56	0.62	0.18	0.59	78.2
S-VII	214.3	0.58	0.76	0.84	0.22	0.78	99.4
S-VIII	127.2	0.34	0.43	0.47	0.134	0.46	60.6
S-IX	116.2	0.31	0.41	0.45	0.12	0.41	53.9
S-X	147.3	0.39	0.54	0.60	0.16	0.53	69.1
Mean	151.9±51.5	0.40±0.14	0.53±0.17	0.58±0.19	0.15±0.05	0.54±0.18	65.0±29.8

The external and internal hazard indices are defined as [5, 14-16]

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1 \qquad (4)$$

$$H_{in(I)} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1 \qquad (5)$$

$$H_{in(II)} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1 \qquad (6)$$

$$H_{in(III)} = \frac{A_{Ra}}{1000} + \frac{A_{Th}}{700} + \frac{A_K}{10000} \le 1 \qquad (7)$$

$$H_{in(IV)} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \le 1 \qquad (8)$$

For safe levels of these radionuclides the values of external as well as internal hazard indices must be less than unity.

The gamma dose rate "D" ($nGyh^{-1}$) in outdoor air at 1m above the ground is calculated using the conversion factor reported by Selvasekarapandian [17] and is given as,

$$D = 0.662C_{Th} + 0.427C_{Ra} + 0.043C_K \qquad (9)$$

where C_{Th}, C_{Ra} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K respectively.

The values of Ra_{eq} concentration, external and internal hazard indices and absorbed dose rate were also calculated in the present study and are shown in Table 3. The Ra_{eq} results ranged from 80 to 248 with an average value of 151.9±51.5, which is less than the limiting value of 370 $Bqkg^{-1}$. The internal and external hazard indices are also less than unity, showing that there is no danger of any health hazard from soil of Quaid-i-Azam University, Islamabad. The values of absorbed dose rate are also given in Table 3. The results ranged from 37.8 to 113.4 with an average value of 65±29.8 $nGyh^{-1}$.

Table 4. Comparison of natural radionuclides in soil with other countries of the world

Country	Average specific activities($BqKg^{-1}$)				References
	^{40}K	^{226}Ra	^{232}Th	^{137}Cs	
Algeria	370	50	25	20	18,32
Arab Republic	270	20	20	---	18
Bangladesh	350	34	---	---	18
Belgium	380	26	27	---	18
Bulgaria	400	45	30	---	18
China	440	32	41	---	20,21
Denmark	460	17	19	---	23
Egypt	320	17	18	6	18,31
Greece	360	25	21	---	18
Hungary	370	33	28	---	18
India	400	29	64	---	18
Iran	640	28	22	---	18
Ireland	350	60	26	---	24
Japan	310	33	28	---	22
Korea	670	---	---	33	18,34
Malaysia	310	67	82	---	18
Netherlands	---	23	---	---	25
Nile Delta (Egypt)	---	---	---	5	33
Norway	850	50	45	---	18
Poland	410	26	21	---	27
Romania	490	32	38	---	28
Spain	470	32	33	27	18,29
Sweden	780	42	42	---	18
Switzerland	370	40	25	---	18
Taiwan	---	---	---	8	30
Thailand	230	48	51	---	18
UK	---	37	---	---	26
United States	370	40	35	---	19
World average	420	33	45	---	18
Pakistan (QAU)	354.7	45.5	55.2	7	Present study

The average specific activities due to ^{226}Ra, ^{232}Th and ^{40}K were compared with the similar work reported for the other countries of the world and are given in Table 4. This table shows that the measured value of the specific activity due to ^{40}K is less than China, India, Korea, Iran, Denmark,

Table 5. Comparison of present study with other areas of Pakistan.

Area	Average Sp. Activity($BqKg_{-1}$)				References
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	
Islamabad	42.4	56.2	565.3	-	35
Charsaddah	57.1	60.1	387.4	7.1	36
Nowshera	-	-	864	18.4	37
Peshawar	-	-	907	19.6	37
D.I.Khan	39	61	657	-	37
Eastern Salt Range (Punjab)	51.3	58.4	678.4	12	1
QAU	45.5	55.2	354.7	7.1	Present study

Norway, Sweden, Poland, Romania, Span and from world average, while it is slightly higher than Egypt, Japan, Malaysia and Arab Republic. However, for rest of the countries this value is comparable. For ^{226}Ra values, all the countries have comparable results except Egypt and Denmark, where the activities are found to be less than those reported for Pakistan as well as for other countries. In case of ^{232}Th, the specific activity of Pakistani samples is slightly higher than Algeria, Egypt, Japan, Iran, Arab Republic, Denmark, Belgium, Ireland, Hungary, Poland and Greece, while for rest of the countries results are comparable. Comparison of ^{137}Cs in soil with other countries of the world is also given in Table 4. This comparison shows that the specific activity of ^{137}Cs in the soil of QAU is less than Spain, Algeria and Korea while it is comparable with Taiwan, Egypt and Nile Delta.

Comparison of the present study with similar work carried out for other areas of Pakistan shows that the concentrations of ^{226}Ra and ^{232}Th are in good agreement with other cities, however, the specific activities of ^{40}K and ^{137}Cs indicate some variations for different areas of Pakistan as shown in Table 5. Comparisons of the radium equivalent activity, external hazard index, internal hazard index and absorbed dose rate are also made with the reported values for other countries of the world and are given in histograms in Figures 1-4 respectively.

4. Conclusions

The average values of the specific activities due ^{226}Ra, ^{232}Th ^{40}K and ^{137}Cs were well compared, with slight variations, with the data available in the literature for other countries of the world as well as that reported for other areas of Pakistan. The activities of natural radionuclides were also well compared with the world average values. The Ra_{eq} activities and internal and external hazard indices were less than their respective limiting values showing that the surveyed area is safe from health hazard point of view.

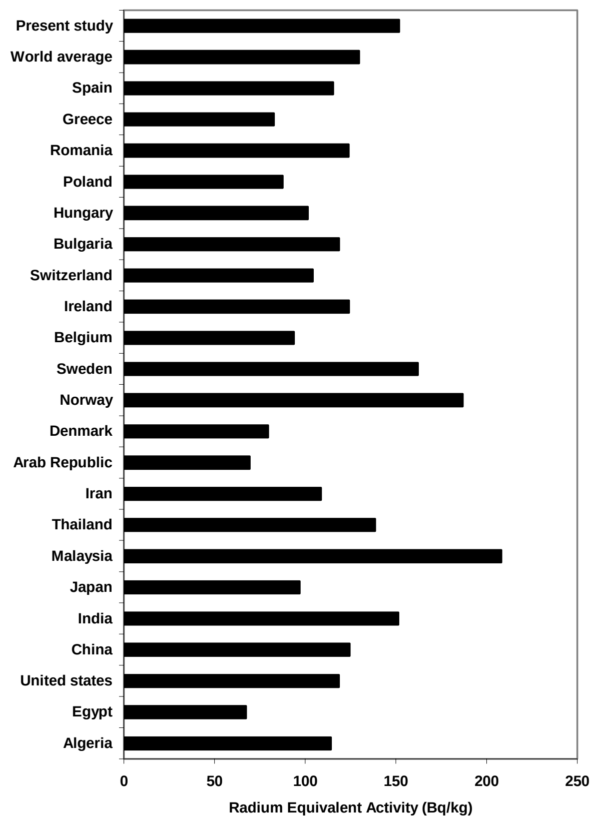

Figure 1. Comparison of radium equivalent activities.

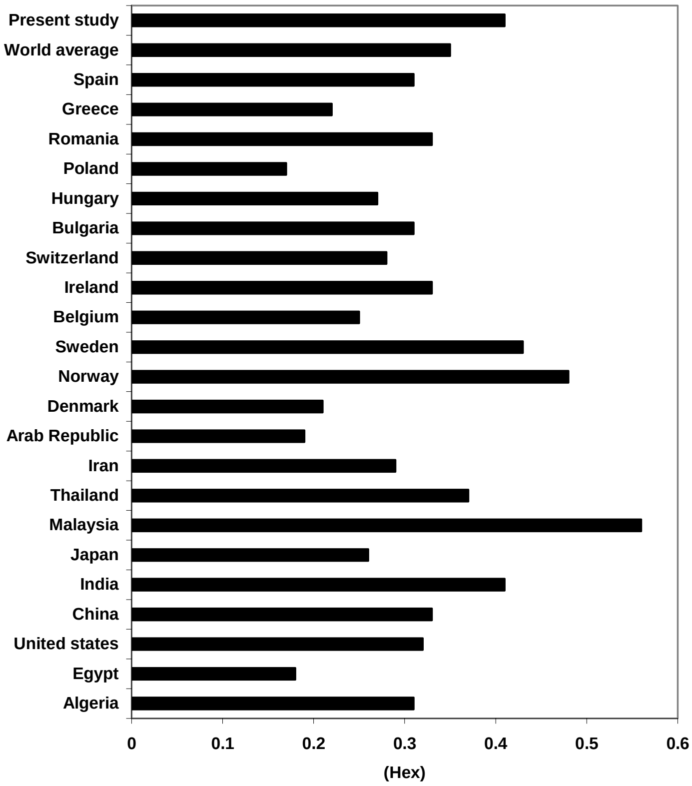

Figure 2. Comparison of external hazard index.

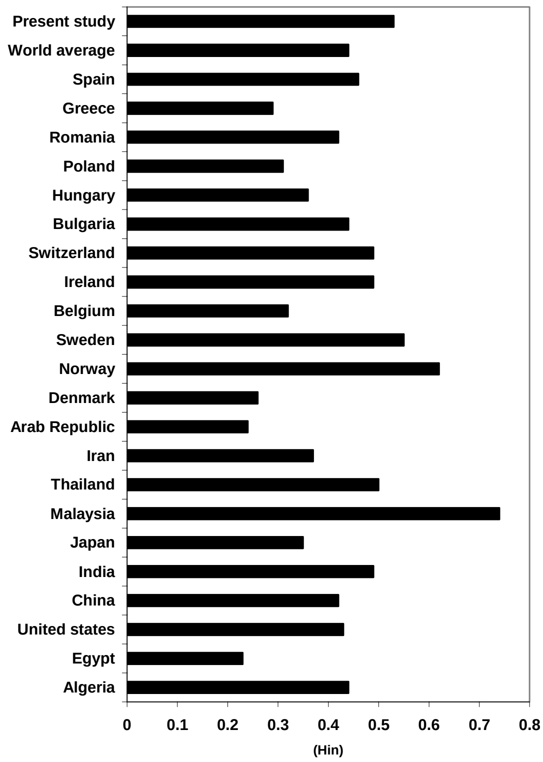

Figure 3. Comparison of internal hazard index.

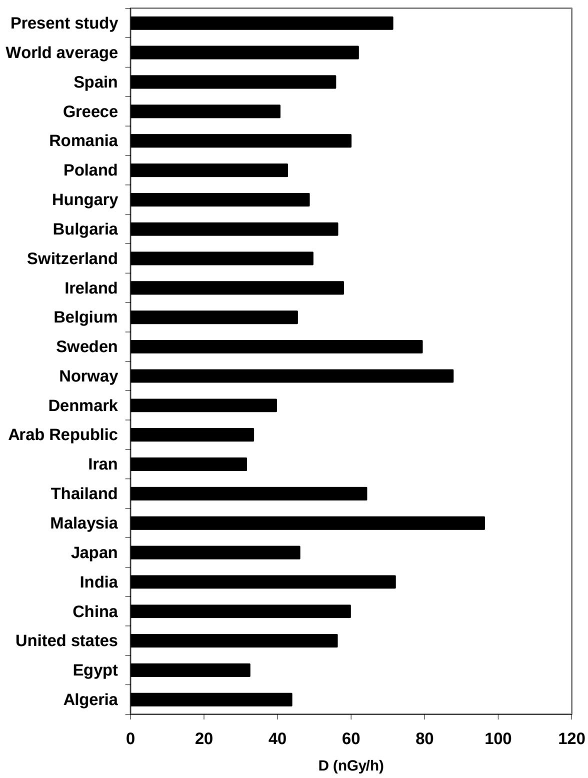

Figure 4. Comparison of absorbed dose rate.

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