CHIN. PHYS. LETT. Vol. 28, No. 1 (2011) 019301

Radioactivity Levels and Gamma-Ray Dose Rate in Soil Samples from Kohistan (Pakistan) Using Gamma-Ray Spectrometry

Hasan M. Khan[1*], M. Ismail[1], K. Khan[2], P. Akhter[2]

[1]*National Center of Excellence in Physical Chemistry, University of Peshawar, Peshawar 25120, Pakistan*
[2]*Health Physics Division, PINSTECH, P. O. Nilore, Islamabad, Pakistan*

(Received 10 June 2010)

The analysis of naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) and an anthropogenic radionuclide ^{137}Cs is carried out in some soil samples collected from Kohistan district of N.W.F.P. (Pakistan), using gamma-ray spectrometry. The gamma spectrometry is operated using a high purity Germanium (HPGe) detector coupled with a computer based high resolution multi channel analyzer. The specific activity in soil ranges from 24.72 to 78.48 $Bq{\cdot}kg^{-1}$ for ^{226}Ra, 21.73 to 75.28 $Bq{\cdot}kg^{-1}$ for ^{232}Th, 7.06 to 14.9 $Bq{\cdot}kg^{-1}$ for ^{137}Cs and 298.46 to 570.77 $Bq{\cdot}kg^{-1}$ for ^{40}K with the mean values of 42.11, 43.27, 9.5 and 418.27 $Bq{\cdot}kg^{-1}$, respectively. The radium equivalent activity in all the soil samples is lower than the safe limit set in the OECD report (370 $Bq{\cdot}kg^{-1}$). Man-made radionuclide ^{137}Cs is also present in detectable amount in all soil samples. Presence of ^{137}Cs indicates that the samples in this remote area also receive some fallout from nuclear accident in Chernobyl power plant in 1986. The internal and external hazard indices have the mean values of 0.48 and 0.37 respectively. Absorbed dose rates and effective dose equivalents are also determined for the samples. The concentration of radionuclides found in the soil samples during the present study is nominal and does not pose any potential health hazard to the general public.

PACS: 93. 85. Np, 95. 85. Pw DOI: 10.1088/0256-307X/28/1/019301

Human beings are exposed to ionizing radiation from natural sources throughout their lifetime. Therefore, knowledge of radionuclide distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and cosmogenic sources. The planet Earth is a radioactive planet and natural radiations are present in every human environment; Earth's material, water, air, foods, and even human body contains naturally occurring radioactive materials.[1] Radionuclides present in the environment are normally found in low concentration. These may be naturally occurring (e.g. uranium-238, thorium-232, and their daughter products and potassium-40 or those produced by cosmic rays interaction in the biosphere) or man made sources (e.g. originating from nuclear fallouts or emanating from nuclear facilities).[2]

Naturally occurring radionuclides are not uniformly distributed and the knowledge of their distribution in soil and rocks play an important role in radiation protection and measurement.[3] Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world.[4]

The experimental demonstration of fission by Otto Frisch in 1939, during World War II and its conclusion at Hiroshima and Nagasaki in August 1945 generated potential risks of large-scale environmental contamination by artificial radioactivity.[5] Since then, several hundreds of various nuclear weapons have been tested, mainly by the USA and the former USSR and by some other countries, such as Britain, France and China. However, the subject of radioactive contamination has gained considerable public importance because of Chernobyl accident. It was the worst nuclear accident and released a large quantity of radioactive contamination throughout the world. ^{137}Cs and ^{90}Sr are two of the fission products released into the atmosphere as a result of nuclear tests carried out since 1945. Due to their long half-lives, these two radionuclides are considered to be the major contributors to the overall collective dose from artificial radiation. ^{137}Cs is an abundant fission product that decays slowly (half-life 30.17 years) and is distributed globally through the upper atmosphere. ^{137}Cs is primarily released by ground level nuclear weapons tests. However it is also emitted due to routine activities of nuclear power generation stations, fuel reprocessing plants and waste disposal sites.[6]

Several studies have been reported on natural and artificial radionuclides in the soil and other environmental samples as well as in food samples in different parts of the world. However, very few data have been reported in literature on the radiological conditions and the background level of different radionuclides in the environmental and food samples of Pakistan. Some studies have reported base line environment radioactivity data in a few areas of Pakistan,[3,7–12] how-

*To whom correspondence should be addressed. Email: hmkhan@upesh.edu.pk
© 2011 Chinese Physical Society and IOP Publishing Ltd

ever, no data for Kohistan area of Pakistan was available. Therefore, the primary aim of this work was to measure the radioactivity levels in soil samples from district Kohistan (N.W.F.P., Pakistan) for health risk assessment.

The word Kohistan is a Persian word, literally means Country of the Hills. Kohistan is a sparsely populated district of the North West Frontier Province of Pakistan. This area lies between the longitude of 72°50′ E to 73°57′ E and the latitude of 34°55′ N to 35°48′ N. The soil samples were collected from various places of Kohistan district (N.W.F.P.) of Pakistan. The sampling was carried out on equidistant basis at longitude and latitude, each line of grid covered the distance about 20 km to 30 km. The sampling sites were chosen to be relatively flat, open and undisturbed. The samples were taken from upper 5 cm layer of land. Each sample was taken with a coring tool within area of 1 m^2. Five cores were taken for each sample, one in the middle and four cores from the corner of 1 m^2 area. These samples were then mixed to make a single sample and were collected in labeled plastic bags. In the laboratory, after removing the roots and stones, soil samples were oven dried at 100 ± 5°C for 24 h, so that the sample weight was constant and then they were ground and sieved through 1–2 mm mesh sieve. We made 200 g soil samples stored in air tight cylindrical plastic containers for at least four weeks before counting, so that secular equilibrium can be attained between ^{226}Ra and its short lived progeny.[13]

Table 1. Radionuclides found in soil samples collected from Kohistan.

Sample No.	Ra-226 Bq·kg^{-1}	Th-232 Bq·kg^{-1}	Cs-137 Bq·kg^{-1}	K-40 Bq·kg^{-1}
S-1	24.72 ± 0.95	25.77 ± 1.62	11.36 ± 0.64	407.38 ± 15.66
S-2	31.16 ± 0.99	21.73 ± 1.59	14.90 ± 0.66	352.92 ± 15.47
S-3	35.38 ± 1.01	32.59 ± 1.68	9.27 ± 0.64	363.81 ± 15.50
S-4	78.48 ± 1.24	73.76 ± 1.96	12.14 ± 0.65	548.98 ± 16.14
S-5	35.61 ± 1.01	41.94 ± 1.75	7.84 ± 0.63	385.60 ± 15.58
S-6	65.72 ± 1.78	74.52 ± 1.97	9.45 ± 0.64	570.77 ± 16.22
S-7	27.76 ± 0.97	29.05 ± 1.65	7.95 ± 0.63	374.70 ± 15.54
S-8	30.22 ± 0.98	35.62 ± 1.70	7.06 ± 0.62	331.13 ± 15.39
S-9	24.72 ± 0.95	22.48 ± 1.60	7.88 ± 0.63	298.46 ± 15.27
S-10	67.36 ± 1.18	75.28 ± 1.97	7.06 ± 0.62	548.98 ± 16.14
Min	24.72 ± 0.95	21.73 ± 1.59	7.06 ± 0.62	298.46 ± 15.27
Max	78.48 ± 1.24	75.28 ± 1.97	14.90 ± 0.66	570.77 ± 16.22
Average	42.11 ± 1.11	43.27 ± 1.72	9.49 ± 0.64	418.27 ± 15.69

To estimate the activity levels of different radionuclides in the soil samples, a high purity germanium (HPGe) detector based gamma-ray spectrometer was used. Analysis of samples was carried out in the Environmental Monitoring Laboratory of Health Physics Division (HPD), Pakistan Institute of Nuclear Sciences and Technology (PINSTECH), Nilore, Islamabad. At HPD, a closed-end coaxial high purity germanium (HPGe) detector (Model GC 3020 Canberra) with 8192 channels, coupled with computer-based MCA acquisition board (Accuspec-A, Canberra) was used. The relative efficiency of the detector was 30% and its resolution (full width at half maximum (FWHM)) for full energy peak of 1332.5 keV gamma ray of Co-60 was 2 keV and peak to Compton ratio was 54 : 1. The results were analyzed by using Windows based Genie-2000 software (Canberra). The HPGe system was calibrated with a standard reference material, Soil 327, from IAEA (International Atomic Energy Agency) for gamma ray activity determination in soil samples. The counting time for calibration, background measurement and measurement of activity during this work was 65000 s (18.055 h). The activity concentration of radionuclide was determined by applying the equation:[14]

$$A_C(\text{Bq/sample}) = (C_S - C_B)/t \times E\gamma \times P\gamma,$$

where C_S is cps for samples, C_B is cps for background, $E\gamma$ and $P\gamma$ are detection efficiency and emission probability of γ-ray, t is counting time.

The specific activities of natural radionuclides present in the soil samples of the selected areas of Kohistan district measured by direct gamma ray spectrometry are given in Table 1. The results for natural radionuclides are reported in Bq·kg^{-1} on dry weight basis. For ^{226}Ra, the specific activity ranged from 24.72 ± 0.95 to 78.48 ± 1.24 Bq·kg^{-1} with an average of 42.11 ± 1.11 Bq·kg^{-1}. For ^{232}Th, the specific activity ranged from 21.73 ± 1.59 to 75.28 ± 1.67 Bq·kg^{-1} with an average of 43.27 ± 1.72 Bq·kg^{-1} and for ^{40}K, it ranged from 298.46 ± 15.27 to 570.77 ± 16.22 Bq·kg^{-1} with an average of 418.27 ± 15.69 Bq·kg^{-1}.

Therefore, as far as the radioactivity from natural radionuclides is concerned, the results clearly indicate that ^{40}K is the only major radionuclide detected in significant amount in all soil samples. All other naturally occurring radionuclides (^{226}Ra and ^{232}Th) determined were in nominal concentrations. The specific activities of these radionuclides in the soil were not uniform and even but varied from soil to soil and location-to-location depending upon the geological and topographic characters of the area under study.

Cesium-137 is an anthropogenic radionuclide and among the class of artificial isotopes released as fission products, it is the most prominent isotope detected by its gamma radiation on Earth's surface. The specific activities of ^{137}Cs found in the soil samples of selected areas are summarized in Table 1. These values varied from 7.06 ± 0.62 to 14.90 ± 0.66 Bq·kg^{-1} with an average value of 9.49 ± 0.64 Bq·kg^{-1}. The presence of ^{137}Cs in all the soil samples is very important and it clearly indicates that the areas under study have received some fallout. It is very difficult to point out the source of this contamination. However, it may be due to Chernobyl accident and minor component, may be due to deposition from different atmospheric nuclear

weapons tests in the past.

The concentration and distribution of ^{40}K, ^{226}Ra, and ^{232}Th in soil is not uniform through out the world, so to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, taking into account the radiation hazard associated with them, a common index, radium equivalent activity (Ra_{eq}), is often calculated.[15] Radium equivalent activity can be defined as

$$Ra_{eq} = A_{Ra} + \frac{10}{7}A_{Th} + \frac{10}{130}A_{K},$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra,^{232}Th and ^{40}K, respectively. It may be noted that ^{238}U has been replaced by a decay product ^{226}Ra. Defining Ra_{eq} by the above equation, 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 the same gamma dose. The Ra_{eq} is related with external gamma ray dose and internal gamma ray dose due to radon and its daughters. The maximum Ra_{eq} should be less than 370 Bq·kg^{-1} for safe use as recommended by the Organization for Economic Cooperation and Development.[16] Radium equivalent activity calculated for different soil samples is given in Table 2.

Table 2. Radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}) and internal hazard index (H_{in}) calculated for soil samples.

Samples No.	Ra_{eq} Activity (Bq·kg^{-1})	H_{ex}	H_{in}
S-1	92.86	0.25	0.32
S-2	89.34	0.24	0.33
S-3	109.92	0.30	0.39
S-4	226.09	0.61	0.82
S-5	125.18	0.34	0.44
S-6	216.08	0.58	0.76
S-7	98.09	0.26	0.34
S-8	106.58	0.29	0.37
S-9	79.79	0.22	0.28
S-10	217.13	0.59	0.77
Minimum	79.79	0.22	0.28
Maximum	226.09	0.61	0.82
Average	136.11	0.37	0.48

In addition, external and internal hazard indices were also calculated for the selected area.[15] The external hazard index H_{ex} can be defined as

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810.$$

It must be less than unity for radiation hazard to be negligible, i.e. the radiation exposure due to radioactive substances from any material to be limited to 1.5 mGy/year. For the maximum value of H_{ex} to be less than unity, the maximum value of Ra_{eq} must be < 370 Bq·kg^{-1}.

Internal hazard index H_{in} is given by[17]

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_k/4810.$$

Here H_{in} should be less than one for safe use for the soil samples. The values calculated for internal and external hazard indices calculated for the soil samples are shown in Table 2. These results show that the value of radium equivalent activity ranges from 79.79 to 226.09 Bq·kg^{-1} with a mean value of 136.11 Bq·kg^{-1}, which is less than the safe limit (370 Bq·kg^{-1}) recommended by the Organization for Economic Cooperation and Development.[16] The internal hazard index ranges from 0.28 to 0.82 with a mean value of 0.48, while external hazard index ranges from 0.22 to 0.61 with an average of 0.37. The highest values of internal and external hazard indices have been found to be less than one, as shown in Table 2. Therefore, based on these results of radium equivalent activity, internal and external hazard indices, one can conclude that there is no health hazard from the soil of district Kohistan as far as gamma radioactivity is concerned. The values of all radiation parameters studied are well within the permissible limits.

The absorbed dose rate in air from radioactivity in soil is also determined. Absorbed dose rate in air is the dose that is received in the open air from the radiation emitted from the radionuclides present in the soil. It was calculated at a height of 1 m above the surface of the ground using a computer programme (INGRE) based on the volume integral method using the conversion factors as follows:[18]

$$D_{outdoor} = (4.27C_{Ra} + 6.62C_{Th} + 0.43C_K) \times 10^{-1}\,\mathrm{nGy \cdot h^{-1}},$$

where $D_{outdoor}$ is the outdoor absorbed dose rate in air, C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

The indoor contribution is assumed to be 1.2 times higher than the outdoor absorbed dose and is given as[19]

$$D_{indoor} = 1.2 \times D_{outdoor}.$$

The annual effective dose equivalent, D_{eff}, from outdoor terrestrial gamma radiation is given as[19]

$$D(\mathrm{eff})_{outdoor} = (D_{outdoor}(\mathrm{nGy \cdot h^{-1}}) \times 0.7(\mathrm{Sv \cdot Gy^{-1}}) \times 8760(\mathrm{h \cdot y^{-1}}) \times 0.3) \times 10^{-6},$$

where 0.3 is the occupancy factor for Pakistan, 8760 is the number of hours in one year, 0.7 Sv·Gy^{-1} is the quotient of effective dose equivalent rate to absorbed dose rate in air and 10^{-6} is the conversion factor between nano and milli.

For indoor exposure, the annual effective dose equivalent is given as[19]

$$D(\mathrm{eff})_{indoor} = (D_{indoor}(\mathrm{nGy \cdot h^{-1}}) \times 0.7(\mathrm{Sv \cdot Gy^{-1}}) \times 8760(\mathrm{h \cdot y^{-1}}) \times 0.7) \times 10^{-6},$$

where 0.7 is the occupancy factor for Pakistan and 0.7 Sv·Gy^{-1} is the conversion factor between effective dose equivalent rate and gamma dose rate in the indoor.

Table 3. Total radioactivity, outdoor and indoor absorbed dose rates in air and annual effective dose equivalents in soil samples.

Sample No.	Total activity ($Bq{\cdot}kg^{-1}$) ($^{226}Ra+^{232}Th+^{40}K$)	$D_{outdoor}$ $nGy{\cdot}h^{-1}$	D_{indoor} $nGy{\cdot}h^{-1}$	$D(eff)_{outdoor}$ $mSv{\cdot}y^{-1}$	$D(eff)_{indoor}$ $mSv{\cdot}y^{-1}$
S-1	457.87	45.13	54.16	0.08	0.23
S-2	405.8	42.86	51.44	0.08	0.22
S-3	431.78	52.32	62.79	0.10	0.27
S-4	701.22	105.95	127.14	0.20	0.55
S-5	463.15	59.55	71.46	0.11	0.31
S-6	711.01	101.94	122.33	0.19	0.52
S-7	431.51	47.20	56.64	0.09	0.24
S-8	396.87	50.72	60.87	0.09	0.26
S-9	345.66	38.27	45.93	0.07	0.20
S-10	691.62	102.20	122.64	0.19	0.53
Minimum	345.66	38.27	45.53	0.07	0.20
Maximum	711.01	105.9 5	127.14	0.20	0.55
Average	503.65	64.61	77.54	0.12	0.33

These factors for the soil samples studied in the present study have been calculated, and the results are summarized in Table 3. Total specific activity from natural terrestrial radionuclides (^{40}K, ^{226}Ra, and ^{232}Th) is given in Table 3 and total terrestrial activity ranges from 345.66 to 711.01 $Bq{\cdot}kg^{-1}$ with an average of 503.65 $Bq{\cdot}kg^{-1}$.

The outdoor and indoor absorbed doses in air due to these terrestrial radionuclides are also calculated, as shown in Table 3. The mean value of outdoor absorbed dose rate in air is 64.61 $nGy{\cdot}h^{-1}$ with a range of 38.27 $nGy{\cdot}h^{-1}$ to 105.95 $nGy{\cdot}h^{-1}$. However, indoor absorbed dose rate ranges from 45.53 $nGy{\cdot}h^{-1}$ to 127.14 $nGy{\cdot}h^{-1}$ with a mean value of 77.54 $nGy{\cdot}h^{-1}$. The large variations in the dose rate, as shown in Table 3, are due to large variations in the specific activity of different primordial radionuclides in the soil samples, which vary from soil to soil. The annual effective dose equivalent from outdoor terrestrial gamma-radiation ranges from 0.07 $mSv{\cdot}y^{-1}$ to 0.20 $mSv{\cdot}y^{-1}$ with a mean value of 0.12 $mSv{\cdot}y^{-1}$. For indoor exposure, the annual effective dose equivalent has a range from 0.20 $mSv{\cdot}y^{-1}$ to 0.55 $mSv{\cdot}y^{-1}$ with a mean value of 0.33 $mSv{\cdot}y^{-1}$.

The total (outdoor plus indoor) annual effective dose equivalent from terrestrial radiation is found to be 0.45 $mSv{\cdot}y^{-1}$, of which 0.12 $mSv{\cdot}y^{-1}$ comes from outdoor and 0.33 $mSv{\cdot}y^{-1}$ from indoor exposure. The corresponding world average value is 0.41 $mSv{\cdot}y^{-1}$ of which 0.07 $mSv{\cdot}y^{-1}$ is from outdoor and 0.34 $mSv{\cdot}y^{-1}$ from indoor exposure.[18]

The soil samples analyzed in the present study show all four types of radioactive contaminants (^{40}K, ^{137}Cs, ^{226}Ra and ^{232}Th). Among all the radionuclide, only ^{40}K is found in appreciable amount in all the samples, and other radionuclides (^{226}Ra, ^{232}Th and ^{137}Cs) are in nominal concentration. ^{137}Cs is an artificially produced radionuclide and was also detected in the soil samples. The origin of this artificially produced radioactive contaminant in soil is expected to be from Chernobyl accident fallout or from nuclear tests conducted by different countries in the past. The values of radium equivalent activity and hazard indices are found to be lower than the safe limit. The values of absorbed dose rates and effective dose equivalents show that the radioactivity of radionuclides found in the surveyed area is nominal and does not pose any potential health hazard to the general public. However, these data may provide a general background level for the area studied and may also serve as a guideline for future measurement and assessment of radionuclides in the case of any radiological emergency and will play a major role in radioactivity mapping of the entire country.

References

[1] Hutchison S G and Hutchison F I 1997 *J. Chem. Edu.* **74** 501
[2] Khan H M et al 1997 *J. Nucl. Sci.* **34** 209
[3] Khan H M et al 1994 *J. Chem. Soci. Pak.* **16** 183
[4] UNSCEAR 2000 *Sources and Effects of Ionizing Radiation* (New York: United Nations Scientific Committee on the Effect of Atomic Radiation)
[5] Vandecasteele C M 2004 *J. Environ. Radioactivity* **72** 17
[6] Rowan D J et al 1998 *J. Environ. Radioactivity.* **40** 15
[7] Khan H M et al 1995 *J. Nucl. Sci.* **32** 249
[8] Tahir et al 2006 *Radiat Prot. Dosim.* **118** 345
[9] Khan H M et al 2010 *Water Air Soil Pollut.* **213** 353
[10] Khan K et al 2003 *International Conference on Isotopic and Nuclear Analytical Techniques for Health and Environment* (Vienna, Austria 10–13 June 2003) IAEA-CN-103/005P
[11] Khan K et al 2010 *Chin. Phys. Lett.* **27** 039301
[12] Chaudhry Z S et al 2002 *J. Radioanal. Nucl. Chem.* **253** 497
[13] IAEA 1989 *Measurement of Radionuclide in Food and Environment* (Vienna: International Atomic Energy Agency) p 27
[14] Fredj A B et al 2005 *Radiat Prot. Dosim.* **117** 419
[15] Beretka J and Mathew P J 1995 *Health Phys.* **48** 87
[16] OECD 1979 *Exposure to radiation from natural radioactivity in building materials* (Paris: Organization for Economic Cooperation and Development Nuclear Energy Agency)
[17] Gallyas M and Torok I 1984 *Radiat Prot. Dosim.* **7** 69
[18] UNSCEAR 1988 *Sources and Effects of Ionizing Radiation* (New York: United Nations Scientific Committee on the Effect of Atomic Radiation)
[19] Hamid B N et al 2002 *Radiat Prot. Dosim.* **98** 227