# **Radioactivity mapping of north western areas of Pakistan**

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(Received February 23, 2005)

A systematic study of natural and fall-out radionuclides was carried out with the environmental samples of soil, vegetation and water from some regions of North West Frontier Province (NWFP) of Pakistan. The pretreatment of the samples was performed in the laboratory using IAEA recommended methods. The analysis of gamma-emitters such as  ${}^{40}K$ ,  ${}^{226}Ra$ ,  ${}^{232}Th$  and  ${}^{137}Cs$  was performed with a high purity germanium detector (HPGe). For the determination of  $90$ Sr, a liquid scintillation counting system was used. The average specific activities of  $^{40}$ K,  $^{226}$ Ra,  $^{232}$ Th and  $^{137}$ Cs have been found to be 307±101 Bq·kg<sup>-1</sup>, 10.2±3 Bq·kg<sup>-1</sup>, 24±6 Bq·kg<sup>-1</sup> and 2.8±1.3 Bq·kg<sup>-1</sup>, respectively, in soil samples. Vegetation samples have smaller values of specific activities and even the analysis of water samples showed values less than LLD for earlier reported radionuclides. Other parameters like hazard indices, radium equivalent activities, absorbed dose rates and effective dose equivalents have also been determined. All these parameters have values less than their respective limiting values representing that the surveyed areas have no significant hazard from health point of view. Analysis of <sup>90</sup>Sr for all the samples showed results below LLD. The present study provides a general background of the detectable radionuclides for the surveyed areas that will be helpful in any radiological emergency.

### **Introduction**

Radiation of natural origin at the earth's surface consists of two components, namely cosmic rays and radiation of the radioactive nuclides in the earth's crust. The latter component, the terrestrial radiation, originates mainly from the so-called primordial radioactive nuclides originated in the early stage of the formation of the solar system. Uranium, thorium and potassium are the main elements contributing to natural terrestrial radioactivity. 1 Uranium has two primary isotopes 238U  $(T_{1/2} = 9.47 \cdot 10^{10} \text{ y})$  and  $^{235}$ U  $(T_{1/2} = 7.0 \cdot 10^{10} \text{ y})$  which, at present, occur in the proportion of 99.3% 238U and  $0.7\%$  <sup>235</sup>U. Both exhibit long and complex decay series. Thorium  $(T_{1/2} = 1.41 \cdot 10^{10} \text{ y})$  has only one isotope, while potassium has three isotopes  $(^{39}K, \frac{40}{K}, ^{41}K)$ , with  $^{40}K$  $(T_{1/2} = 1.28 \cdot 10^9 \text{ y})$  being the only radioactive isotope of natural abundance (0.012% of potassium). 2

 $137Cs$  and  $90Sr$  are two of the fission products released into the atmosphere as a result of nuclear tests carried out since 1945, and, due to their long half-lives, are considered the major contributors to the overall collective dose from artificial radiation. 3 The potential harmfulness of the former fission products is based on their chemical similarity to potassium and calcium, elements which are incorporated into human organisms and other vertebrates from both food and water. In particular, the determination of  $137Cs$  and  $90Sr$  in soils is of great importance owing to the fact that plant roots are one of the ways of incorporation into the trophic chain. 4

Knowledge about the radiation of natural origin is very important being the largest source of population dose and its assessment is important as a baseline with which radiation protection standards may be formulated. Additionally, natural radiation involves the entire global

population. 5 The exposure of men to these naturally occurring radionuclides is, and has been, an unavoidable consequence of their presence in the earth's crust, surface, soil, air, food and water.<sup>6</sup>

It is now widely accepted among experts that natural radiation accounts for the greatest part of public radiation exposure. Moreover, the common understanding of the exposure of man to the natural radiation environment in the 1960s was fairly simple compared with the current understanding.<sup>7</sup>

Due to the increased public concern and awareness about radioactive pollution, the aim of the present research work was to determine the radioactivity levels in a variety of environmental samples of some districts of North West Frontier Province (NWFP) of Pakistan. Present study is a joint collaborative work between Chemistry Department, Quaid-i-Azam University, Islamabad, Pakistan and HPD, PINSTECH, Nilore, Islamabad, Pakistan.

The main objective of this study is to estimate the type and amount of natural and fallout radioactivity levels in the soil and other environmental samples and to establish base line data. A radiological environmental monitoring survey has already been accomplished for some other specific areas of Pakistan. The results are published elsewhere. $2,8-14$ 

#### **Experimental**

Twenty-five soil samples were collected from various locations of NWFP districts namely D. I. Khan, Tank, Lakki Marwatt, Karak and Bannu. These districts lie between the longitude of  $70^{\circ}$ -20<sup> $\prime$ </sup>N to  $71^{\circ}$ -02 $\prime$ N and the latitude of  $31^{\circ}$ -15<sup>'</sup>W to  $33^{\circ}$ -11'W as shown in Fig. 1. The sampling was carried out on equidistant basis

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at longitude and latitude, each line of the grid covered the distance of 27.5 km and 25 km, respectively. Vegetation and water samples were collected from D. I. Khan, Tank and Bannu. The sampling sites were chosen to be relatively flat, open and undisturbed. The soil samples were taken from the upper 5 cm layer with a coring tool. Vegetation was collected from different places in the selected site and was combined to make a single representative. In order to prevent contamination with soil, the vegetation sample was cut to a height of 5 cm from the ground. The soil and vegetation samples were collected in polyethylene bags and labeled properly (date and place). Water samples were collected in 10 liter cans. After collection, the samples were stored to avoid degradation, spoiling or other decomposition.

In the laboratory, after removing the roots and stones, soil samples were oven dried at 110 °C until the sample weight became constant and then they were ground and sieved. 5 About 200 g of soil samples were stored in air tight cylindrical plastic containers for at least 4 weeks before counting, so that secular equilibrium can be attained between 226Ra and its short lived progeny.<sup>15</sup> In order to remove all organic matter, the vegetation samples were ashed in a muffle furnace at a temperature not higher than 450 °C since at higher temperature loss of volatile radionuclides, i.e., radiocesium, may be significant. For ashing operation, as recommended, stainless steel trays were used.

In the case of water samples, evaporation was carried out on a hot plate for volume reduction from 1 liter to 100 ml. In order to maintain the homogeneity of the water samples and to avoid adsorption of radionuclides on the walls of the container, all the samples were acidified with  $0.1N$  HNO<sub>3</sub>. The temperature for evaporation was kept below the boiling point of water. Samples were transferred and stored in air tight containers for over 30 days to allow for Ra-Rn equilibrium before radiometric analysis. 6

To estimate the activity levels of 40K, 238U, 232Th and 137Cs, a high resolution gamma-ray spectrometer consisting of a HPGe detector (Model GC 3020 Canberra) coupled to PC based MCA card (Accuspec-A, Canberra) available at HPD, PINSTECH, Nilore, Islamabad, was used. The relative efficiency of the detector was 30% and the resolution 2.23 keV at 1332 gamma-rays of  ${}^{60}Co$ . The detector was equipped with 8192-channels and was shielded in an 8 cm lead chamber with an inner lining of 0.5 cm thick copper plate to reduce the background. The results were analyzed by using Geni-2000 software (Canberra). Efficiency calibration of the detection system was done with Soil-375, obtained from IAEA.<sup>16</sup> Every sample was counted for 65,000 seconds. A background sample was normally analyzed at the weekends and background

counts were then subtracted from the total counts to obtain sample counts during that week.  $^{40}$ K and  $^{137}$ Cs were analyzed by their single peaks of 1460.8 keV and 661.6 keV, respectively. However, the analysis of  $^{238}$ U and 232Th was based upon the peaks of progeny in equilibrium with their parent radionuclides.

Soil, vegetation and water samples were then prepared for  $90$ Sr measurement by  $90$ Y, as  $90$ Sr is a low energy-emitter. Ashed samples of soil and vegetation except water (i.e., in the form of liquid) were introduced for the estimation of  $90Sr$  by its daughter product  $90Y$  by solvent extraction that requires  $2-3$  days for analysis. To determine the  $90$ Sr, dry ashed samples were consecutively (a) boiled with 14.4M HNO<sub>3</sub> to bring  $90Sr$ into solution, (b) extracted with equilibrated TBP to separate  $90Y$  from  $90Sr$  and other interfering radionuclides, (c)  $90Y$  was precipitated as an oxalate to get a chemical yield (d) the Y-oxalate precipitate was dissolved in 2M HCl to obtain a liquid source, then it was counted (e) by a liquid scintillation counter.<sup>17</sup>

 $90Y$  activity was measured with the liquid scintillation counting system. In the present study, a tricarb system model 4530 with efficiency of 59.51% of a standard solution S6/11/142 was used. Background counts were determined by counting a blank vial containing all the chemicals used in the preparation of samples with the exception of radionuclides of interest. Counting time for each sample was 200 minutes.



Fig. 1. Map of the sampling sites

## **Results and discussion**

The results of specific activities of  ${}^{40}$ K,  ${}^{226}$ Ra,  ${}^{137}$ Cs and 232Th are reported in Table 1. The results indicate that  $40K$  is the only radionuclide present in a significant amount in all soil samples while the other radionuclides are only present in very nominal concentrations. The specific activities of  ${}^{40}$ K ranged from 196.90±7.80 to  $753.97\pm9.65$  with an average of 306.91 Bq·kg<sup>-1</sup>, that of  $137Cs$  from 0.60 $\pm$ 0.20 to 5.14 $\pm$ 0.20 with an average of 2.82 Bq·kg<sup>-1</sup>, that of <sup>226</sup>Ra from 5.78±0.35 to 21.44 $\pm$ 0.40 with an average of 10.18 Bq·kg<sup>-1</sup> and that of 232Th from 13.60±0.85 to 47.15±1.10 with an average specific activity of 24.0 Bq $kg^{-1}$ . The trend of specific activities is not uniform in individual samples, but varies from sample to sample and location to location as shown in Table 1. This non-uniform behavior of radionuclides may be attributed to their uneven and irregular distribution in the earth crust and also to various topographical and agricultural activities.<sup>18</sup>

The correlation between the specific activities of  $^{40}$ K,  $^{226}$ Ra and  $^{232}$ Th in soil samples is shown in Figs 2 to 4. As shown in Fig. 2, the correlation between  $226Ra$ 

and 232Th is high, with a correlation coefficient 0.84. The relation between  $232$ Th and  $40$ K as well as between  $226$ Ra and  $40$ K (Figs 3 and 4) also represents the positive trend between the specific activities of these radioactive elements. In the case of grass and water samples, no distinct trend was observed between any radionuclides.

As the concentration and distribution of  ${}^{40}$ K,  ${}^{226}$ Ra, 232Th and 137Cs in soils is not uniform throughout the world, so uniformity in respect of exposure to radiation has been defined in terms of the radium equivalent activity given by:<sup>19</sup>

$$
Ra_{\text{eq}} = A_{\text{Ra}} + 10/7 A_{\text{Th}} + 10/130 A_{\text{K}} \tag{1}
$$

where  $A_{\text{Ra}}$ ,  $A_{\text{Th}}$  and  $A_{\text{K}}$  are the specific activities of  $^{226}$ Ra,  $^{232}$ Th and  $^{40}$ K, respectively.

Defining the above equation, it has been assumed that  $10 \text{ Bq·kg}^{-1}$  of  $226\text{Ra}$ ,  $7 \text{Bq·kg}^{-1}$  of  $232\text{Th}$  and 130 Bq·kg<sup>-1</sup> of <sup>40</sup>K produce the same gamma-dose. The maximum  $Ra_{\text{eq}}$  must be less than 370 Bq·kg<sup>-1</sup> for safe use (OECD limit). In the present study, the value of  $Ra_{eq}$ for all samples was less than the permissible limit as shown in Table 2.

	Specific activity, Bq·kg <sup>-1</sup>				
Place	40 <sub>K</sub>	$\overline{^{137}}$ Cs	$226$ Ra	$\overline{^{232}}$ Th	
Mahrah	$296.3 \pm 8.2$	$3 \pm 0.2$	$10.7 \pm 0.3$	$26.4 \pm 0.9$	
Bukhara	$267.1 \pm 8.1$	$0.8 \pm 0.2$	$11.1 \pm 0.3$	$23.2 \pm 0.9$	
Lakhani	$284.6 \pm 8.1$	$1.7 \pm 0.2$	$10.4 \pm 0.3$	$21.5 \pm 0.9$	
Matt	$302.2 \pm 8.2$	$5.1 \pm 0.2$	$12.3 \pm 0.4$	$25.7 \pm 0.9$	
Khutti	$401.5 \pm 8.5$	$4.2 \pm 0.2$	$10.4 \pm 0.3$	$26.8 \pm 0.9$	
Suggu southern	$267.0 \pm 8.1$	$3.3 \pm 0.2$	$11.8 \pm 0.4$	$29.3 \pm 0.9$	
Daraban	$308.0 \pm 8.2$	$2.6 \pm 0.2$	$11.9 \pm 0.4$	$29.9 \pm 0.9$	
Dajal	$290.5 \pm 8.2$	$2.7 \pm 0.2$	$11.1 \pm 0.3$	$24.4 \pm 0.9$	
Rangpur	$302.2 \pm 8.2$	$3.4 \pm 0.2$	$10.8 \pm 0.3$	$25.7 \pm 0.9$	
Suggu northern	$313.9 \pm 8.2$	$3.5 \pm 0.2$	$10.8 \pm 0.3$	$25.4 \pm 0.9$	
Paniala	$319.7 \pm 8.2$	$3.2 \pm 0.2$	$10.4 \pm 0.3$	$26 \pm 0.9$	
Chunda	$243.7 \pm 8$	$2.7 \pm 0.2$	$7.6 \pm 0.3$	$19 \pm 0.9$	
<b>Tittar Khel</b>	$319.7 \pm 8.2$	$2.7 \pm 0.2$	$11.4 \pm 0.3$	$25.8 \pm 0.9$	
Hathala	$208.6 \pm 7.8$	$0.7 \pm 0.2$	$5.8 \pm 0.3$	$13.6 \pm 0.8$	
Tank	$197 \pm 7.8$	$4.8 \pm 0.2$	$6 \pm 0.3$	$17.3 \pm 0.9$	
Gul Imam	$302.2 \pm 8.2$	$0.6 \pm 0.2$	$8.2 \pm 0.3$	$23.4 \pm 0.9$	
Bain	$278.8 \pm 8$	< 0.113	$7.5 \pm 0.3$	$23 \pm 0.9$	
Sarai Gambila	$284.6 \pm 8$	< 0.113	$8.8 \pm 0.3$	$22.2 \pm 0.9$	
Peen	$308 \pm 8.2$	$3.4 \pm 0.2$	$8.2 \pm 0.3$	$22.9 \pm 0.9$	
Nurar	$255.4 \pm 8$	$3 \pm 0.2$	$8.5 \pm 0.3$	$20.3 \pm 0.9$	
Surani	$278.8 \pm 8$	$4.2 \pm 0.2$	$9.5 \pm 0.3$	$23.4 \pm 0.9$	
Bannu	$284.6 \pm 8$	$0.8 \pm 0.2$	$10.8 \pm 0.3$	$21.1 \pm 0.9$	
Domail	$273 \pm 8$	$1.5 \pm 0.2$	$9.6 \pm 0.3$	$18.7 \pm 0.9$	
Land Kamar	$754 \pm 9.6$	$4.6 \pm 0.2$	$21.4 \pm 0.4$	$47.2 \pm 1$	
Landiwah	$331.4 \pm 8.3$	$2.3 \pm 0.2$	$9.8 \pm 0.3$	$20.4 \pm 0.9$	
Minimum:	$197 \pm 7.8$	$0.6 \pm 0.2$	$5.8 \pm 0.3$	$13.6 \pm 0.8$	
Maximum:	$754 \pm 9.6$	$4.8 \pm 0.2$	$21.4 \pm 0.4$	$47.2 \pm 1$	
$A.M. \pm S.D.$	$307 \pm 101$	$2.8 \pm 1.3$	$10.2 \pm 3$	$24 \pm 6$	
$G.M. \pm S.D.$	$296.8 \pm 101.8$	$2.4 \pm 1.4$	$9.8 \pm 3$	$23.5 \pm 6$	
Median:	$290 \pm 8$	$3 \pm 0.2$	$10.4 \pm 0.3$	$23.4 \pm 0.9$	

Table 1. Gamma-spectrometric measurements of soil samples



Fig. 2. Scatter plot of soil radium versus soil thorium with linear regression line showing positive correlation



Fig. 3. Scatter plot of soil potassium versus soil thorium with linear regression line showing positive correlation



Fig. 4. Scatter plot of soil potassium versus soil radium with linear regression line showing positive correlation

			Hazard indices				
Place	$Ra_{\text{eq}}$ , Bq·kg <sup>-1</sup>	$H_{\text{in}}(I)^*$	$H_{\text{in}}(\text{II})$	$H_{\text{in}}(III)$	$H_{\text{in}}(VI)$	$H_{\rm ex}$ **	
Mahrah	$71.3 \pm 5.3$	0.2	0.2	0.1	0.3	0.2	
<b>Bukhara</b>	$64.8 \pm 4.8$	0.2	0.2	0.1	0.2	0.2	
Lakhani	$63 \pm 4.8$	0.2	0.2	0.1	0.2	0.2	
Matt	$72.3 \pm 5.8$	0.2	0.2	0.1	0.2	0.2	
Khutti	$79.6 \pm 6.8$	0.2	0.3	0.1	0.3	0.2	
Suggu southern	$74.3 \pm 5.5$	0.2	0.3	0.1	0.3	0.2	
Daraban	$78.3 \pm 6$	0.2	0.2	0.1	0.3	0.2	
Dajal	$68.4 \pm 5$	0.2	0.2	0.1	0.3	0.2	
Rangpur	$70.8 \pm 5$	0.2	0.2	0.1	0.3	0.2	
Suggu northern	$71.3 \pm 5$	0.2	0.2	0.1	0.3	0.2	
Paniala	$72.1 \pm 5.2$	0.2	0.2	0.1	0.1	0.2	
Chunda	$53.5 \pm 4.2$	0.2	0.2	0.1	0.2	0.1	
<b>Tittar Khel</b>	$72.9 \pm 5.2$	0.2	0.2	0.1	0.3	0.2	
Hathala	$41.3 \pm 3.4$	0.1	0.1	0.05	0.2	0.1	
Tank	$45.8 \pm 3.4$	0.1	0.2	0.05	0.2	0.1	
Gul Imam	$64.9 \pm 5.2$	0.2	0.2	0.1	0.2	0.2	
Bain	$61.9 \pm 5.4$	0.2	0.2	0.1	0.2	0.2	
Sarai Gambila	$60.4 \pm 5.4$	0.2	0.2	0.1	0.2	0.2	
Peen	$64.6 \pm 5.4$	0.2	0.2	0.1	0.2	0.2	
Nurar	$57.3 \pm 5.2$	0.2	0.2	0.1	0.2	0.1	
Surani	$64.4 \pm 5.4$	0.2	0.2	0.1	0.2	0.2	
Bannu	$62.8 \pm 6$	0.2	0.2	0.1	0.2	0.2	
Domail	57.3 $\pm$ 6	0.2	0.2	0.1	0.2	0.1	
Land Kamar	$146.9 \pm 8$	0.5	0.5	0.2	0.6	0.4	
Landiwah	$64.5 \pm 5.2$	0.2	0.2	0.1	0.2	0.2	
Minimum:	$41.3 \pm 3.4$	0.1	0.1	0.05	0.2	0.1	
Maximum:	$146.9 \pm 8$	0.5	0.5	0.2	0.6	0.4	
$A.M. \pm S.D.$	$68.2 \pm 18.8$	$0.2 \pm 0.1$	$0.2 \pm 0.1$	$0.1 \pm 0.04$	$0.2 \pm 0.1$	$0.2 \pm 0.1$	
$G.M. \pm S.D.$	$66.36 \pm 18.86$	$0.2 \pm 0.1$	$0.2 \pm 0.1$	$0.1 \pm 0.04$	$0.2 \pm 0.1$	$0.2 \pm 0.1$	
Median:	64.80	0.20	0.21	0.1	0.2	0.2	

Table 2.  $Ra_{eq}$  activities and internal and external hazard indices of soil samples

 $*$  H<sub>in(I-IV)</sub> are internal hazard indices.

\*\*  $H_{\text{ex}}$  is external hazard index.

In order to limit the radiation dose from soil, a number of internal and external hazard indices have been suggested by some workers $19,20$  and are given below:

Internal hazard indices:

$$
H_{\rm in}(I) = A_{\rm Ra}/185 + A_{\rm Th}/259 + A_{\rm K}/4810
$$
 (2a)

$$
H_{\text{in}}(\text{II}) = A_{\text{Ra}}/150 + A_{\text{Th}}/259 + A_{\text{K}}/4810 \tag{2b}
$$
  
H. (III) = A<sub>2</sub>, (1000 + A<sub>27</sub>, (700) + A<sub>37</sub>, (10000) (2c)

$$
H_{\rm in}(III) = A_{\rm Ra}/1000 + A_{\rm Th}/700 + A_{\rm K}/10000 \qquad (2c)
$$

$$
H_{\rm in}(IV) = A_{\rm Ra}/300 + A_{\rm Th}/200 + A_{\rm K}/3000 \tag{2d}
$$

External hazard index:

$$
H_{\rm ex} = A_{\rm Ra}/370 + A_{\rm Th}/259 + A_{\rm K}/4810\tag{3}
$$

For radiologically safe materials, the maximum recommended values of these hazard indices must be less than or equal to 1. In the present study, these hazard indices are found to be less than unity for all samples (Table 2), which are in accordance with the recommended values.

The outdoor absorbed dose rate is calculated usually in the height of 1 m above the ground surface using the computer programme INGRE based on the volume integral method using the conversion factors as reported by UNSCEAR:<sup>21</sup>

$$
D_{\text{outdoor}} = (4.27 C_{\text{Ra}} + 6.62 C_{\text{Th}} + 0.43 C_{\text{K}}) \times
$$
  
×10<sup>-1</sup>nGy·h<sup>-1</sup> (4)

where  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$  and  $C_{\text{K}}$  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 dose and is given as: $21$ 

$$
D_{\text{indoor}} = 1.2 \times D_{\text{outdoor}} \tag{5}
$$

The annual effective dose equivalent  $D_{\text{eff}}$  from outdoor terrestrial gamma-radiation is given as: $2^{\frac{2}{2}}$ 

$$
D(eff)_{\text{outdoor}} = [D_{\text{outdoor}} (nGy \cdot h^{-1}) \times
$$
  
×0.7 (Sv·Gy<sup>-1</sup>)×8760 (h·y<sup>-1</sup>)×0.3]×10<sup>-6</sup> (6)

where 0.3 is the occupancy factor for Pakistan, 8760 is the number of hours in one year,  $0.7 \text{ Sv-Gy}^{-1}$  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: <sup>22</sup>

$$
D(eff)_{\text{indoor}} = [D_{\text{indoor}} (n\text{Gy} \cdot \text{h}^{-1}) \times 0.7 \, (\text{Sv} \cdot \text{Gy}^{-1}) \times
$$
  
×8760 (h·y<sup>-1</sup>)×0.7]×10<sup>-6</sup> (7)

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

The results of outdoor, indoor and annual effective dose equivalents are shown in Table 3. The total (outdoor plus indoor) annual effective dose equivalent from terrestrial radiation is found to be 0.234 mSv of which 0.172 mSv comes from indoor and 0.062 mSv from outdoor, the corresponding world average value is 0.41 mSv of which 0.34 mSv is from indoor and 0.07 mSv from outdoor. 21

Specific activities of natural and fall-out radionuclides have been obtained in vegetation and water samples. Table 4 indicates the minor presence of radionuclides in vegetation samples. The data in Table 5 shows the specific activities below LLD for all water

samples, thus these radionuclides in the aquatic environment do not pose any significant health risk to the public.

In present study, very low concentration of  $90Sr$  has been observed, i.e., below LLD (Tables 6 to 8) showing that the study area might have received an ignorable concentration of this fall-out radionuclide. Even for this small amount, it can be assumed that it may originate due to the past nuclear tests. In the case of  $90Sr$ , it is less firmly fixed to the soil matrix and so it is more available for root uptake than cesium. The higher mobility of  $90Sr$ also means that this radionuclide migrates faster than  $137Cs$  in the soil column thus results in the lower concentration of the 90Sr in surface layer as compared to  $137Cs$ ,  $23$  For the analysis of undisturbed soil, as in the present study, it may be assumed that radionuclides will be retained in the upper 2 cm layer until ploughing.<sup>24</sup> 90Sr has a greater resemblance with Ca. The exchangeable Ca in the soil is the most important factor in determining the extent of  $90Sr$  in the soil. Thus, the increase in the concentration of Ca in the soil may lead to a decrease in 90Sr concentration.

Place	$D_{\text{outdoor}}$	$D_{\text{indoor}}$	$D(\text{eff})_{\text{outdoor}}$	$D(\text{eff})_{\text{indoor}}$
	$nGy h^{-1}$	$nGy h^{-1}$	$mSvy$ <sup>-1</sup>	$mSv·y^{-1}$
Mahrah	34.8	41.7	0.1	0.2
Bukhara	31.5	37.8	0.1	0.2
Lakhani	30.9	37.1	0.1	0.1
Matt	35.2	42.2	0.1	0.2
Khutti	39.4	47.3	0.1	0.2
Suggu southern	35.9	43.1	0.1	0.2
Daraban	38.1	45.7	0.1	0.2
Dajal	33.4	40.1	0.1	0.2
Rangpur	35.6	42.7	0.1	0.2
Suggu northern	34.9	41.9	0.1	0.2
Paniala	35.3	42.4	0.1	0.2
Chunda	26.3	31.5	0.04	0.1
<b>Tittar Khel</b>	35.7	42.8	0.1	0.2
Hathala	20.4	24.5	0.04	0.1
Tank	22.4	26.9	0.04	0.1
Gul Imam	32	38.3	0.1	0.2
Bain	30.4	36.5	0.1	0.1
Sarai Gambila	30.6	36.8	0.1	0.1
Peen	31.9	38.2	0.1	0.2
Nurar	28.1	33.7	0.05	0.1
Surani	31.5	37.8	0.1	0.2
Bannu	30.7	36.8	0.05	0.1
Domail	28.2	33.8	0.05	0.1
Land Kamar	72.8	87.3	0.1	0.4
Landiwah	31.9	38.3	0.1	0.2
Minimum:	20.4	24.5	0.04	0.1
Maximum:	72.8	87.3	0.1	0.4
$A.M. \pm S.D.$	$33.5 \pm 9.3$	$40.2 \pm 11.2$	$0.1 \pm 0.02$	$0.2 \pm 0.04$
$G.M. \pm S.D.$	$32.6 \pm 9.3$	39.1±11.2	$0.1 \pm 0.02$	$0.2 \pm 0.04$
Median:	31.9	38.2	0.1	0.2
World average:	61.9	74.3	0.1	0.3

Table 3. Absorbed dose rates and effective dose equivalents in soil samples

Table 4. Gamma-spectrometric measurements of vegetation samples

	Specific activity, $Bq \cdot kg^{-1}$			
Place	$40\nu$	137 $C_{\rm c}$	$^{226}Ra$	<sup>232</sup> Th
Paniala	70.1	< 0.1	1.4	2.7
Tank	26.7	< 0.05	<0.1	0.3
Bannu	141 5	<0 04	04	05

Table 5. Gamma-spectrometric measurements of water samples



The detection limits of these radionuclides are  ${}^{40}$ K = 4.6·10<sup>-3</sup>,  $137$ Cs = 1.1·10<sup>-4</sup>,  $226$ Ra = 2·10<sup>-4</sup> and  $232$ Th = 4.8·10<sup>-4</sup> Bq·kg<sup>-1</sup>.

The comparison of present results with the corresponding world average values is presented in Table 9. The mean activities of  $2^{26}Ra$ ,  $2^{32}Th$  and  $40K$ are, respectively, 0.31, 0.54 and 0.73 times that of the world average values. Hazard indices, radium equivalent activity, absorbed dose rates and effective dose equivalents result in less than the world average indicating that the surveyed areas are beyond any risk from health point of view.

## **Conclusions**

The average values of specific activities of all natural and fall-out radionuclides in the studied soil, vegetation and water samples are found to be normal when compared to the world average values and data reported in the literature. The radium equivalent activities, hazard indices, indoor and outdoor absorbed dose rates and effective dose equivalents have been found to be less than their respective limiting values showing that the surveyed area has no significant hazard from health point of view. The present study provides a general background of the detectable radionuclides for the surveyed area and will be helpful in any radiological emergency.



Table 6. <sup>90</sup>Sr concentrations in soil samples

\* Yield, % is the percent chemical recovery of the radiotracer after completion of all the radiochemical steps.

Table 7. <sup>90</sup>Sr concentrations in vegetation samples

Place	Yield, %	Specific activity, $Bq \, kg^{-1}$
Paniala	97	< 0.04
Tank	92	< 0.04
Bannu	85	<0.04

## Table 8. <sup>90</sup>Sr concentrations in water samples



Radiological parameters	Present results	World average	Ratio of the
	(Average)		present average/world average
$226$ Ra. Bq·kg <sup>-1</sup>	10.2	33	0.3
$^{232}$ Th, Bq·kg <sup>-1</sup>	24	45	0.5
${}^{40}K$ , Bq·kg <sup>-1</sup>	307	420	0.7
Internal hazard index, Bq·kg <sup>-1</sup>	0.2	0.5	0.4
External hazard index, Bq·kg <sup>-1</sup>	0.2	0.5	0.4
Radium equivalent activity, Bq·kg <sup>-1</sup>	68.2	129.7	0.5
Outdoor dose, $nGy \cdot h^{-1}$	33.5	61.9	0.5
Indoor dose, $nGy \cdot h^{-1}$	40.2	74.3	0.5
Outdoor effective dose, $mSv y^{-1}$	0.1	0.1	
Indoor effective dose, $mSv-y^{-1}$	0.2	0.3	0.6
Total annual effective dose, $mSv \cdot y^{-1}$	0.3	0.4	0.8

Table 9. Comparison of present results with the corresponding world average values

#### **References**

- 1. UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, Report to the General Assembly, United Nations, New York, 1993.
- 2. T. JABBAR, M. S. SUBHANI, K. KHAN, A. RASHID, S. D. ORFI, A. Y. KHAN, J. Radioanal. Nucl. Chem., 258 (2003) 143.
- 3. E. GOMEZ, F. GARCIAS, M. CASAS, V. CERDA, J. Appl. Radiation Isotopes, 48 (1997) 699.
- 4. UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, Ionizing Radiation Source and Biological Effects, Report to the General Assembly, United Nations, New York, 1982.
- 5. J. YEBOAH, M. BOADU, E. O. DARKO, J. Radioanal. Nucl. Chem., 249 (2001) 629.
- 6. K. KHAN, P. AKHTER, S. D. ORFI, G. M. MALIK, M. TUFAIL, J. Radioanal. Nucl. Chem., 256 (2003) 289.
- 7. B. N. HAMID, M. I. CHOWDHURY, M. N. ALAM, M. N. ISLAM, Radiat. Prot. Dosim., 98 (2002) 227.
- 8. M. ASLAM, S. D. ORFI, K. KHAN, A. JABBAR, J. Radioanal. Nucl. Chem., 253 (2002) 483.
- 9. K. KHAN, H. M. KHAN, J. Pak. Chem. Soc., 16 (1994) 183.
- 10. H. M. KHAN, K. KHAN, M. A. ATTA, F. JAN, J. Nucl. Sci., 32 (1995) 249.
- 11. K. KHAN, H. M. KHAN, M. TUFAIL, N. AHMED, J. Environ. Radioact., 38 (1998) 77.
- 12. K. KHAN, H. M. KHAN, J. Appl. Radiation Isotopes, 54 (2001) 861.
- 13. K. KHAN, M. ASLAM, S. D. ORFI, H. M. KHAN, Radiat. Prot. Dosim., 95 (2001) 263.
- 14. K. KHAN, M. ASLAM, S. D. ORFI, H. M. KHAN, J. Environ. Radioact., 58 (2002) 59.
- 15. S. SADASIVAN, V. K. SHUKLA, S. CHINNAESAKKI, S. J. SARTANDEL, J. Radioanal. Nucl. Chem., 256 (2003) 603.
- 16. AQCS/IAEA, Analytical Quality Control Services, International Atomic Energy Agency, Vienna, Austria, 1998.
- 17. A. RASHID, M. ASLAM, S. D. ORFI, A Rapid Method for the Estimation of 90Sr in Dietary Samples, Report HPD-234, June, 2001.
- 18. A. BRODSKY, Handbook of Environmental Radiation, CRC Press, Inc., Florida, 1982, ch. 1.
- 19. J. BERETKA, P. J. MATHEW, Health Phys., 48 (1985) 87.
- 20. L. S. QUINDOS, P. L. FERNANDEZ, J. SOTO, Building materials as source of exposure in houses, in: Indoor Air, 87, 2, B. SEIFERT, H. ESDORN (Eds), Institute of Water, Soil and Air Hygiene, Berlin, 1987, p. 365.
- 21. UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, Report to the General Assembly, United Nations, New York, 1988.
- 22. B. N. HAMID, M. I. CHOWDHURY, M. N. ALAM, M. N. ISLAM, Radiat. Prot. Dosim., 98 (2002) 227.
- 23. T. E. MYRICH, B. A. BERVEN, F. F. HAYWOOD, Health Phys., 45 (1983) 631.
- 24. H. FRIBERG, R. VESANEN, Appl. Radiation Isotopes, 51 (1999) 229.