Spatial Distribution of Radionuclides and Major Elements in Soil of Murree and Kotli Sattian Punjab, Pakistan

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Abstract

The study is aimed to investigate the specific activity of radionuclides and major elements in soils of Subdivisions Murree and Kotli Sattian (Himalayan Mountain range) Pakistan as a part of the ongoing baseline data collection using gamma-ray spectrometer and wavelength dispersive X-ray fluorescence (WDXRF). The natural radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K showed inhomogenous distribution 20.0 to 29.5, 43.4 to 62.4 and 163.0 to 493.6 Bq kg⁻¹ respectively where as ¹³⁷Cs exhibited extreme variation from 1.3 to 54.1 Bqkg⁻¹. The dominant constitutes of soil were SiO₂, Al₂O₃ and Fe₂O₃. Weak to moderate correlation was observed between elemental composition and activity concentrations of radionuclides. The average annual effective absorbed dose rate measured at 1m above ground due to terrestrial sources was 72.9 ± 1.0 μ Svy⁻¹. The high altitude of study area causes the dominant doses to the public .The radium equivalent activity Ra_{eq}, outdoor and indoor hazard indices were lower than the safe limit of Organization for Economic Cooperation and Development report for general public.

Keywords: Gamma emitters, major elements, XRF, soil, Himalayan Mountain range, Gamma Spectrometry

1 Introduction

The assessment of natural (accounted for 50-80% of the gamma radiation flux at the soil surface) and anthropogenic radionuclides is necessary to evaluate the risks arising from radiations (Izquierdo et al., 2011). Naturally occurring radionuclides of terrestrial origin like members of the ²³⁸U and ²³²Th series, together with ⁴⁰K, and their concentrations in soil are related to the nature of the parent rock during soil genesis. Their natural radioactivity levels vary depending on the geological and geographical structure (Vukasinovic, DJORDJEVIC et al. 2010). It has been observed that granitic rocks contain higher amounts of thorium, uranium and light rare earth elements (REEs) compared to other igneous rocks such as basalt and andesites (SAHOO, HOSODA et al. 2011). Anthropogenic radionuclides are introduced into environment mainly from nuclear releases and nuclear weapon tests performed in early 1960's and dispersed through atmosphere.¹³⁷Cs a fission product with high fission yield and half-life of 30 years, is a prominent indicator of nuclear releases (Izquierdo et al., 2011).

Soil play a major role in the cycling of radionuclides and their physicochemical properties influence the mobility and bioavailability of these radionuclides in terrestrial ecosystems (Izquierdo et al., 2011). The concentration and composition of elements in the soil are also of primary importance in determining distribution of any radionuclide between soil particle and soil solution. Radionuclides are adsorbed onto soil components (organic matter, clays, carbonates, Fe/Mn oxides) and take part in biogeochemical processes.

The objective of this study was to determine spatial distribution of primordial (226 Ra, 232 Th and 40 K) and anthropogenic 137 Cs in the study area of Murree and Kotli sattian to provide a baseline data for future reference. In addition, the composition of parent materials in the study area, might explain the distribution of radionuclides in the landscape.

2 Materials and Methods

2.1 Geology of study area

The sampling was carried out in subdivisions Murree and Kotli sattian, hilly areas of District Rawalpindi of Punjab province. The study area is situated at $33^{0}35'-33^{0}54'$ latitude and $73^{0}26'-73^{0}27'$ longitude and is the one of the most popular tourists destination in Pakistan. The total area is approximately 762 km². The Murree is known as "Queen of Hills". The hills of Kotli sattian are at the edge of the Himalayan Mountain range of Miocene sand stone and Eocene MUMMULTIC lime stone (Satti 2000). The Murree formation at some places is purple to reddish brown, medium grained thin bedded hard sand stone contains of chert. The important rocks of this area include limestone, shale, sandstone, brick-red clay, silty clay stone and purple to dark brown sand stone (Kazmi and Jan, 1997). The average rainfall per annum is 1789 mm. The elevation of study area ranges from 2000 to 7200 feet above sea level.

2.2 Sample collection and Analysis

The sampling area was divided into grids $(5 \times 5 \text{ km}^2)$ on equidistant basis as shown in fig1. At each sampling site an area of dimensions $2m \times 2m$ was selected, after removing top vegetation cover, four subsamples were taken from corners of square and one from center. These samples were taken with the help of coring tool (2 cm diameter and 5cm coring length) at depth of (0-5) cm. These subsamples were pulverised manually to get a homogenized sample. Site locations were identified using a portable global positioning system (GPS) as presented in Table 1.

The soil samples were dried, powdered, mixed and passed through 2 mm mesh sieve (IAEA, 1989). The samples were stored for more than 22 days to establish secular equilibrium of ²³⁸U with its progeny. The radiometric analysis of samples was carried out using a high-resolution coaxial high purity germanium detector (HPGe) Model GC2519 for 65000 seconds. The detector had graded shielding (15 cm thick lead having inner lining of 3 mm thick copper and 4 mm thick tin) to reduce the background by increasing backscattering (Knoll, 2010) . GENIE-2000 software was used for spectral analysis. The activity concentration of the ²²⁶Ra and ²³²Th were estimated using the photo-peaks of their daughters (²¹⁴Pb 351 keV and ²¹⁴Bi 609,1120 keV) and (²²⁸Ac, 911keV). Photo-peaks at 662 and 1460.8 keV were used for ¹³⁷Cs and ⁴⁰K activities respectively. The minimum detectable activity values for ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs were 3.60, 2.25, 6.70 and 1.10 Bqkg⁻¹ respectively. The quality control was assured using reference soils IAEA 375, IAEA 327 and soil 6.

For chemical composition, sieved samples were mixed with binder (4:1) and palletized in aluminum cap (diameter 37 mm) with help of 150 kN hydraulic press. Each sample was irradiated for 300 sec. The concentrations were measured against rock standard RG-1, RGN-1. The chemical analysis was performed using Axios Sequential XRF spectrometer. The instrument was operated at 4 kW power and equipped with a SST X-ray tube technology, Rh anode and LiF-200 crystal (for dispersion of wavelengths).

3 Results and discussion

3.1 Activity Concentrations

The measured concentrations of ²²⁶Ra, ²³²Th, ¹³⁷Cs and ⁴⁰K on dry weight basis with uncertainties at 1 sigma level are shown in Table 2. The mean specific activities of terrestrial radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K were 24.7 \pm 2.9, 52.5 \pm 4.9 and 368 \pm 75.0 Bq kg⁻¹ respectively with ranges (20-30), (43-63) and (163-493) Bq kg⁻¹ respectively. However, ¹³⁷Cs concentration showed large variation from 54.1 \pm 0.8 Bqkg⁻¹ in KotliRF to 1.3 \pm 0.1 Bq kg⁻¹ in Samli colony respectively. The quantitative analysis of data by Anderson Darling test showed that all the radionuclides ²²⁶Ra (p-value=0.4), ²³²Th (p-value=0.8), and ⁴⁰K (p=0.1) except Cs-137 (p- value < 0.05) were normally distributed about their mean with 95% confidence level.

The activity concentrations of primordial radionuclides were observed to follow order as: 226 Ra < 232 Th < 40 K. The average values of activity concentration in Bq kg⁻¹ were converted in units of part per million (ppm) for U and Th and % for 40 K using conversion factors reported in literature. The average values obtained were 2.0 \pm 0.01 ppm of uranium, 12.7 \pm 0.14 ppm of thorium and 1.2 \pm 0.01% of 40 K. These values are comparable to those for shales of sedimentary origin (Tyler, 1994).

The 232 Th/ 226 Ra activity ratio (i.e., progeny pair 228 Ac/ 214 Pb) can be used to assess the maintenance of the proportionality within the 232 Th and 226 Ra decay series, which in most environmental samples is about 1.1. In our study, 232 Th/ 226 Ra was found to vary from 1.6 to 2.6 with mean value 2.1. 232 Th can be easily mobilized in forms of various complex inorganic cations and organic compounds, <u>it</u> is expected that leaching and sorption might be attributed to different mobility of 232 Th and 226 Ra (Izquierdo et al., 2011) as reported for 238 U.

A comparison of present data with other hilly areas of Pakistan i.e. Hunza (Ali et al., 2013), Mirpur (AJK) (Rafique et al., 2011) and Swat (Jabbar et al., 2008) was also made through Box-Whisker plots as shown in Fig. 2 a-d. Hunza is located in the sedimentary belt of Karakorum mountains range. The rocks of Hunza are composed of limestone, conglomerates, quartzite, marble, schist, gneiss. The Mirpur formation consists of a conglomerate as a major unit with some mudstones or sand stones and lies on Himalayan mountain range as study area. Swat is located in Kohistan mountain range. The granite gneiss of swat consists of feldspar, biotite, garnet and chlorite (Kazmi and Jan, 1997).

From Fig. 2-a it can be seen that the average activity concentration of ²²⁶Ra was comparable to Mirpur but lower than values reported for Hunza and Swat. However, activity concentration in the study area covers a narrow range than all the other hilly stations compared. Quite similar to ²²⁶Ra, ²³²Th concentrations were also comparable to Mirpur and may be attributed to same geology. The average activity concentration of ⁴⁰K was lowest in the study area. Although concentration was comparable to Swat but data had still overlapping with Mirpur. Of the radionuclides detected, ¹³⁷Cs had the most heterogeneous distribution in study area contrary to other compared areas where it was below MDA for most of sampling sites.

3.2 Comparison with World's Data

A comparison of baseline data obtained in the present study with some countries in East Asia is shown in Table

3. It can be seen that average activity of terrestrial radionuclides are comparable to China, Thailand and Kazakhstan, lower those measured in Kauman Himalaya and Malaysia.¹³⁷Cs data was not available for most of countries compared except Kauman Himalaya. The average ¹³⁷Cs activity levels in present study area were higher than those found at Kauman Himalaya.

3.3 Chemical Composition

There are many physical and chemical factors that are not only responsible for the distribution of radionuclides in soil but also responsible for surface and sub-surface alterations. Therefore, few samples (grids size 10×10 km²) were analyzed by XRF for soil chemistry.

The chemical composition of soil samples was expressed as the function of eight major elements. Their concentration in terms of their respective oxides is represented by their weight percentage as shown in Table 4. The average values of major elements are given in terms of one standard deviation.

It s evident that the concentration of SiO₂ in the study area vary from 58.7 to 71.3 % with an average value of 66.2 ± 3.9 %. TiO₂ concentration had homogenous distribution in soil of study area and was found in range (0.8% -1.4%), consistent with the range value (0.5-1.5) reported by (Omoniyi et al., 2013). The average value of Al₂O₃ was estimated to be 16.2 ± 1.6 with the values ranging from 13.1-17.9%. Fe₂O₃ was found to be varying from 3.4 to 7.9%. Similarly, the mean concentrations of MnO, MgO, CaO and P₂O₅ were estimated to be 0.1, 2.93, 4.4, 0.2% respectively.

3.4 Correlation Study

Correlation study using linear regression model was carried out between radionuclide concentration and major stable elements as variables. Microsoft Excel 2007 was used to perform the correlation study. Weak to moderate correlations were observed. These moderate correlations include ¹³⁷Cs-Fe₂O₃ (r = 0.7), ⁴⁰K-Fe₂O₃ (r = 0.8), ⁴⁰K-Al₂O₃ (r = 0.8) see Fig. 3 a, b & c.

The data did not represent any significant correlation between silicates and terrestrial radionuclides in the study area. A Moderate correlation between ¹³⁷Cs and Fe₂O₃ revealed that major soil constituents responsible for the retention of cesium are clay, silt and Fe oxides. McKay and Baxter (1985) showed that the presence of oxides and organics was important in the soil thus preventing the release of ¹³⁷Cs from the clay mineral fractions. Therefore, clay soil acts as a sink for ¹³⁷Cs (Tyler, 1994).

3.5 Radiological parameters

Radium Equivalent activity and hazard indices

Most of the houses in the present study area are being constructed by using local rocks, walls and floors are labeled with mud and roofs are also covered by soil. So, people are exposed to radiation indoor as well as outdoors. The weighted sum of 226 Ra, 232 Th and 40 K activities, frequently used for the estimation of radiation hazards in building materials, is quantified by radium equivalent activity (Ra_{eq}) (Beretka and Mathew 1985). It is defined by following relation

 $Ra_{eq}(Bq kg^{-1}) = A_{226Ra+} 1.429 A_{232Th} + 0.077 A_{40K}$

(1)

Where, A_{226Ra} , A_{232Th} and A_{40K} are activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹ respectively. Any material is potentially radiotoxic if the value of Ra_{eq} exceeds 370 Bq kg⁻¹ corresponds to annual effective dose of 1.05 mSv. The brief statistics of radium equivalent activity values are shown in Table 5. The values vary from (107.3-147.8) with average value of 126.6 ± 9.5 Bqkg⁻¹ which is 66% lower than safe limit of 370 Bq kg⁻¹. The main contributor to Ra_{eq} in study area was ²³²Th with average 58% contribution.

The hazards associated with outdoor or indoor occupancy are represented by outdoor radiation hazard index H_{out} and H_{in} respectively(Beretka and Mathew, 1985). These indices are calculated by using following formulae

| $H_{out} = (A_{226Ra} / 370) + (A_{232Th} / 259) + (A_{40K} / 4810)$ | (2) |
|--|-----|
| $H_{in} = (A_{226Ra} / 185) + (A_{232Th} / 259) + (A_{40K} / 4810)$ | (3) |

Its value also should be less than one. It can be seen from Table 5 that maximum values of H_{in} and H_{out} were found at DewalRF but still these values are less than unity.

Annual effective dose

The annual effective dose due to background radiations may be regarded as the sum of doses from terrestrial radionuclides (226 Ra, 232 Th and 40 K), anthropogenic radionuclides (137 Cs) and cosmic radiations. The air absorbed dose rate at one meter above ground due to terrestrial radiations is given by (Singh et al., 2009) D (nGyh⁻¹) = 0.461A_{226Ra} + 0.623 A_{232Th} + 0.0414 A_{40K} (4) The outdoor annual effective dose equivalent can be calculated by using following relation (Khan et al., 2012)

The outdoor annual effective dose equivalent can be calculated by using following relation (Khan et al., 2012) $E_{out} (\mu Svy^{-1}) = 0.7 \times D (nGyhr^{-1}) \times 0.2 \times 8760 \times 10^{-3}$ (5)

Where, E out is outdoor effective dose equivalent, 0.7 (SvGy⁻¹) is Sievert to Grey conversion factor, 0.2

is outdoor occupancy factor. Since atomic weapon testing in early sixties and the accident of Chernobyl and Fukushima, ¹³⁷Cs is unevenly distributed throughout the world and its higher levels may produce significant exposures to mankind. The effective dose rate at one meter above ground contributed by ¹³⁷Cs, neglecting air attenuation, can be calculated by using equation 6 as given in (Cember and Johnson, 2000)

 $D_{137Cs} (\mu Svh^{-1}) = 0.576 E^* \Phi^* (\mu_a/\rho)^{tissue} \times 10^{-3}$

Where, E is gamma ray energy of ¹³⁷Cs photon, $(\mu_a/\rho)^{\text{tissue}}$ is energy dependent mass attenuation coefficient of soft tissue and has value $0.0316(\text{cm}^2/\text{gm})$. Φ is gamma ray flux at one meter above ground and is given by

$$\Phi = \frac{A * Y}{2 * (\mu a / \rho)^{\text{soil}}}$$

(7)

Where, A is activity of 137 Cs in Bqkg⁻¹, Y is gamma ray emission probability per disintegration of 137 Cs, 2 in the denominator is due to fact that one half of the total emitted photons form soil probably move upward and (μ_a/ρ) soil is mass attenuation of soil. The value of mass attenuation coefficient of at 661 keV of ¹³⁷Cs was measured using gamma ray transmission method (Cutshall et al., 1983). The average value of mass attenuation coefficient was found to be $0.0780 \text{ cm}^2/\text{ g}$. The outdoor annual effective dose rate was calculated by equation 8 as.

$$E_{137Cs} = D_{137Cs} (\mu \text{Svhr}^{-1}) \times 8760 \times 0.2$$

(8)

(6)

The average value of the outdoor annual effective dose rate due to 137 Cs in soil came out to be 1.9 ± 0.02 which is 63% of world's average reported value of 3µSvy⁻¹. Table 6 represents the brief statistics. The dose contribution from ground deposition of ¹³⁷Cs was insignificant. The component of annual effective dose from directly ionizing cosmic radiation and that from neutron part as the function of altitude above sea level can be expressed by equations 9 and 10 respectively (UNSCEAR, 2000). exp(-1.649z)] (9)

$$E_{DI} = E(0) [0.79 \exp(0.4528z) + 0.21]$$

 $\dot{E}_{N} = 1.98 \dot{E}_{N}(0) \exp(0.698z)$

(10)Where, $\dot{E}(0)$ is annual effective dose from comic radiations at sea level, $\dot{E}_{N}(0)$ is annual effective dose from neutron part at sea level and z is altitude above sea level in kilometers. The cumulative dose that a person may receive during a year is the sum of all these contributions

 $E_{net} = E_{terr} + E_{cos} + E_{Cs-137}$ (11)It can be seen from Table 6 that average annual effective dose $72.0 \pm 1.0 \ \mu Svy^{-1}$ is comparable to world's average. However, outdoor dose contribution from cesium part came out to be 46% lower than world's average value. The most dominate term to annual effective dose rate came from cosmic radiations which was 87% of total annual effective dose thus 32% higher than world's average value. Clearly, this is attributed to higher altitudes of the study area.

4 Conclusions

The data obtained revealed that the concentration of radionuclides and major stable elements vary considerably from one site to the other . The activity concentrations of primordial radionuclides showed that soil of study area might have originated from sedimentary shale. The ²³²Th/²²⁶Ra activity ratios as a measure of disequilibrium caused by the different mobility of the radioisotopes provided insights into the intensity of soil processes. The chemical analysis confirmed that the study area was rich in silicates and alumina. Weak to moderate correlation was found between major elements and radionuclide concentrations. The outdoor and indoor radiation hazard indices exhibited that the soil of study area was hazard free. The annual effective dose rate due to terrestrial radionuclides was comparable to world's average value. The major dose contribution to total annual effective dose came from cosmic radiations due to high altitudes of study area and was 32% higher than World's average value. The dose contribution from ¹³⁷Cs part may be neglected. Nevertheless, two sampling locations KotliRF and Bariyan were identified to have relatively higher values of ¹³⁷Cs concentration.

5 References

Ali, M., Iqbal, S., Wasim, M., Arif, M., Saif, F., 2013. Soil radioactivity levels and radiological risk assessment in the highlands of Hunza, Pakistan. Rad. Prot. Dosim. 153 (3), 390-399.

Beretka, J., Mathew, P., 1985. Natural radioactivity of Australian building materials, industrial wastes and byproducts. Health Phys. 48 (1), 87-95

Cember, H., Johnson, T., 2000. Introduction to Health Physics. 4th Edition, McGraw-Hill New York.

Cutshall, N., Larsen, J.L., Olsen, C.R., 1983. Direct analysis of Pb-210 in sediment samples: a self-absorption corrections. Nucl. Instr. and Meth. A 206, 309-312.

International Atomic Energy Agency IAEA, 1989. Measurement of radionuclides in food and environment. Technical Report Series No. 295, Vienna.

Izquierdo, N.A., Gaspar, L., López-Vicente, M., Machín, J., 2011. Spatial distribution of natural and artificial radionuclides at the catchment scale (South Central Pyrenees). Rad. Meas. 46 (2), 261-269.

Jabbar, T., Khan, K., Subhani, M., Akhter, P., Jabbar, A., 2008. Environmental gamma radiation measurement in District Swat, Pakistan. Rad. Prot. Dosim. 132 (1), 88-93

Kazmi, A.H., Jan, M.Q., 1997. Geology and tectonics of Pakistan, Graphic publishers Karachi

Khan, K., Khalid, M.R., Jabbar, A., Akhter, P., 2012. Appraisal of radioactivity and associated radiation hazards in sand samples of four rivers of Punjab province, Pakistan. Isot.Environ. Healt. S. 48 (2), 286-294

Knoll, G.F., 2010. Radiation detection and measurement, John Wiley & Sons

- Omoniyi, I.M., Oludare, S.M., Oluwaseyi, M., 2013. Determination of radionuclides and elemental composition of clay soils by gamma-and X-ray spectrometry. SpringerPlus 2 (1), 1-11
- Rafique, M., Rehman, H., Malik, F., Rajput, M., Rahman, S., Rathore, M., 2011. Assessment of radiological hazards due to soil and building materials used in Mirpur Azad Kashmir; Pakistan Iran. J. Radiat. Res.9 (2), 77-87
- Sahoo, S.K., Hosoda, M., Kamagata, S., Sorimachi, A., Ishikawa, T., Tokonami, S., Uchida, S., 2011. Thorium, uranium and rare earth elements concentration in weathered Japanese soil samples. Nucl. Sci.Technol. 1, 416-419.

Satti, S. 2000. A Hand Book of Kotli Sattian. Nanopathy

- Singh, J., Singh, H., Singh, S., Bajwa, B., Sonkawade, R., 2009. Comparative study of natural radioactivity levels in soil samples from the Upper Siwaliks and Punjab, India using gamma-ray spectrometry. J. Environ. Radioac. 100 (1), 94-98
- Tyler, A N., 1994. Environmental influences on gamma ray spectrometry, Ph. D. Thesis, The Science Faculty, University of Glasgow
- UNSCEAR 2000. Sources and effects of ionizing radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York.
- Vukasinovi, I., Dordevic, A., Rajkovic, M.B., Todorovic, D., Pavlovic, V.B., 2010. Distribution of natural radionuclides in anthrosol-type soil. Turk. J. Agric. For. 34, 539-54

| | | Table 1 Geographical | data of sampling points | |
|--------|------------|----------------------|-------------------------|---------------|
| Sr.No. | Sample ID. | Sample Location | Coordinates | Altitude (ft) |
| 1. | I. | Newala | 33.88 N ,73.58 E | 2287 |
| 2. | II. | Bahndi | 33.88 N ,73.54 E | 3438 |
| 3. | III. | Kamra | 33.73 N, 73.50 E | 4089 |
| 4. | IV. | Bhattian | 33.75 N, 73.49 E | 3782 |
| 5. | V. | Bariyan | 33.96 N, 73.39 E | 6980 |
| 6. | VI. | Dhar RF | 33.93 N, 73.39 E | 5890 |
| 7. | VII. | Sambli colony | 33.72 N, 73.35 E | 2431 |
| 8. | VIII. | Nanjan | 33.74 N,73.39 E | 3755 |
| 9. | IX. | Patriata | 33.85 N, 73.48 E | 7280 |
| 10. | Χ. | Messyari | 33.87 N,73.44 E | 5843 |
| 11. | XI. | Tarmina | 33.80 N ,73.23E | 2282 |
| 12. | XII. | Arokas | 33.84 N,73.29 E | 2926 |
| 13. | XIII. | Kotli RF | 33.82 N ,73.52 E | 3960 |
| 14. | XIV. | Barhad | 33.80 N,73.55 E | 2574 |
| 15. | XV. | DewalRF | 33.98 N, 73.46E | 5940 |
| 16. | XVI. | Talut | 34.03 N,73.85 E | 3630 |
| 17. | XVII. | Suleha | 33.78 N ,73.40E | 2546 |
| 18. | XVIII. | Balkh | 33.80 N,73.33 E | 3585 |
| 19. | XIX. | Lawrence college | 33.88 N, 73.36E | 5730 |
| 20. | XX. | Ghora Gali | 33.81 N,73.33 E | 4505 |
| 21. | XXI. | Charra pani | 33.85 N,73.32E | 3610 |
| 22. | XXII. | Sambli Behra mal | 33.83 N,73.37 E | 3774 |
| 23. | XXIII. | Sanj | 33.84 N,73.45 E | 4889 |
| 24. | XXIV. | Durbhandi | 33.79 N,73.45 E | 4935 |
| 25. | XXV. | Chapprian | 33.93 N,73.55 E | 2206 |
| 26. | XXVI. | Gohi Rf | 33.93 N,73.50 E | 2896 |
| 27. | XXVII. | Kashmiri Bazar | 33.94 N,73.45 E | 6386 |
| 28. | XXVIII. | Kuthian | 33.84 N,73.54 E | 3979 |
| 29. | XXIX. | Sarmandal | 33.84 N,73.49 E | 6596 |
| 30. | XXX. | Behl | 33.75 N,73.43 E | 3436 |
| 31. | XXXI. | Channeri | 33.74 N,73.33E | 2934 |

| Table2: Activity concentration of Radionuclides in soil samples | | | | | |
|---|--------|--|-------------------|-------------------|---------------------------|
| Sr .No. | Sample | Activity Concentration (Bqkg ⁻¹) | | | |
| | ID | ²²⁶ Ra | ²³² Th | ¹³⁷ Cs | ⁴⁰ K |
| 1. | Ι | 24.3 ± 1.5 | 53.7 ± 5.8 | 6.3 ± 0.4 | 377.4 ± 23.3 |
| 2. | II | 26.2 ± 1.3 | 52.3 ± 4.6 | 9.7 ± 0.4 | 421.1 ± 20.5 |
| 3. | III | 27.7 ± 1.0 | 57.9 ± 2.6 | 10.3 ± 0.3 | 472.4±22.8 |
| 4. | IV | 25.9 ± 1.4 | 54.7 ± 3.2 | 13.6 ± 0.4 | 435 ± 23.1 |
| 5. | V | 28.9 ± 1.1 | 49.3 ± 2.0 | 38.3 ± 0.7 | 424.8±23.2 |
| 6. | VI | 27.8 ±1.0 | 48.6 ± 2.1 | 23.6 ± 0.3 | 349 ± 18.6 |
| 7. | VII | 29.5±0.9 | 51.9 ± 3.1 | 1.3±0.1 | 163.1±16.2 |
| 8. | VIII | 25.4 ± 1.1 | 55.3 ± 4.1 | 4.7 ± 0.1 | 456 ± 25.7 |
| 9. | IX | 20.0±0.8 | 43.4 ± 3.1 | 3.6±0.2 | 374.4±17.5 |
| 10. | Х | 22.7 ± 0.9 | 48.4 ± 3.7 | 2.7 ± 0.1 | 409 ± 20.8 |
| 11. | XI | 21.5±0.9 | 45.1 ± 3.4 | 2.3±0.1 | 276.8±19.9 |
| 12. | XII | 23.4 ± 1.3 | 60.4 ± 4.8 | 16.6 ± 0.3 | 344 ± 19.1 |
| 13. | XIII | 20.6±0.9 | 45.5 ± 3.7 | 54.1±0.8 | 493.6±21.1 |
| 14. | XIV | 28.5 ± 1.5 | 49.9 ± 3.1 | 30.6 ± 0.7 | 417 ± 22.3 |
| 15. | XV | 27.0±1.0 | 62.4 ± 4.1 | 26.4±0.5 | 410.8±22.9 |
| 16. | XVI | 29.4 ± 1.3 | 47.7 ± 2.8 | 10.6 ± 0.4 | 234 ± 17.6 |
| 17. | XVII | 25.4±1.0 | 54.6 ± 4.0 | 3.8±0.2 | 288.1±19.0 |
| 18. | XVIII | 22.2 ± 0.8 | 55.8 ± 4.2 | 24.34 ± 0.5 | 376 ± 20.3 |
| 19. | XIX | 23.8±0.9 | 50.6 ± 3.9 | 20.8±0.5 | 397.9±21.4 |
| 20. | XX | 24.1 ± 0.9 | 48.9 ± 3.2 | 11.7 ± 0.3 | 387 ± 21.2 |
| 21. | XXI | 27.6 ± 1.2 | 44.3 ± 2.9 | 14.4 ± 0.3 | 287 ± 18.7 |
| 22. | XXII | 26.5 ± 1.3 | 57.2 ± 4.1 | 8.3 ± 0.2 | 295 ± 19.3 |
| 23. | XXIII | 21.3 ± 0.9 | 48.5 ± 3.1 | 15.9 ± 0.4 | 411 ± 22.1 |
| 24. | XXIV | 23.9 ± 1.2 | 44.6 ± 2.7 | 3.9 ± 0.2 | 348 ± 19.7 |
| 25. | XXV | 29.1 ± 1.2 | 50.7 ± 3.3 | 12.8 ± 0.2 | 435 ± 23.7 |
| 26. | XXVI | 20.5 ± 0.8 | 47.8 ± 2.8 | 17.6 ± 0.3 | 408 ± 22.6 |
| 27. | XXVII | 23.2 ± 0.9 | 60.5 ± 4.0 | 4.8 ± 0.2 | 312 ± 16.6 |
| 28. | XXVIII | 25.1 ± 1.3 | 56.4 ± 3.7 | 4.2 ± 0.3 | 457 ± 25.3 |
| 29. | XXIX | 20.5 ± 1.1 | 43.9 ± 2.7 | 9.3 ± 0.4 | 421 ± 23.4 |
| 30. | XXX | $\overline{23.6 \pm 1.4}$ | 45.7 ± 3.4 | 2.8 ± 0.1 | $3\overline{42 \pm 22.3}$ |
| 31. | XXXI | $\overline{21.5 \pm 0.9}$ | 53.8 ± 3.6 | 13.7 ± 0.5 | $2\overline{87 \pm 18.3}$ |

Table 3 Comparison with World's Data

| Country | ²²⁶ Ra | ²³² Th | ⁴⁰ K | ¹³⁷ Cs |
|------------------------|-------------------|-------------------|-----------------|-------------------|
| China | 32 | 41 | 440 | |
| India | 29 | 64 | 400 | |
| Bangladesh | 34 | | 350 | |
| Malaysia | 67 | 82 | 310 | |
| Kazakstan | 35 | 60 | 300 | |
| Thailand | 48 | 51 | 230 | |
| Japan | 33 | 28 | 310 | |
| Kauman Himalaya, India | 67 | 79 | 887 | 2.8 |
| Punjab, Pakistan | 35 | 41 | 615 | 2.8 |
| Gilgit | 25 | 29 | 115 | 3.6 |
| Hunza | 60 | 59 | 766 | 2.2 |
| Ghanger Valley | 56 | 58 | 852 | 13.4 |
| Sawat | | | | |
| World's Median | 400 | 35 | 30 | |
| Present study | 25 | 53 | 368 | 13.6 |

| Table 4 Concentration of major elements in soil samples | | | | | | | | |
|---|------|------|-------|-------|-----|-----|------|------|
| Sample | SiO2 | TiO2 | Al2O3 | Fe2O3 | MnO | MgO | CaO | P2O5 |
| ID | | | | | | _ | | |
| Ι | 69.1 | 1.2 | 16.9 | 6.5 | 0.1 | 2.4 | 2.6 | 0.1 |
| III | 67.0 | 1.2 | 17.6 | 6.2 | 0.1 | 2.7 | 2.9 | 0.1 |
| V | 64.0 | 1.3 | 16.0 | 7.4 | 0.2 | 8.5 | 2.3 | 0.1 |
| VII | 71.3 | 1.4 | 14.4 | 4.3 | 0.1 | 1.1 | 3.5 | 0.8 |
| IX | 70.0 | 1.2 | 17.4 | 7.0 | 0.1 | 1.7 | 1.4 | 0.6 |
| XI | 58.7 | 0.8 | 13.1 | 3.4 | 0.1 | 2.0 | 18.5 | 0.1 |
| XIII | 62.0 | 1.3 | 17.7 | 7.9 | 0.2 | 3.9 | 3.0 | 0.1 |
| XV | 69.5 | 1.3 | 16.5 | 6.3 | 0.1 | 2.4 | 1.0 | 0.2 |
| XVII | 66.2 | 1.0 | 14.7 | 3.9 | 0.1 | 2.3 | 7.1 | 0.1 |
| XIX | 64.0 | 1.4 | 17.9 | 7.2 | 0.2 | 2.2 | 2.0 | 0.1 |

Table 5 Radiological parameters estimated from soil of study area

| Statistics | Ra _{eq} (Bqkg ⁻¹) | H _{in} | H _{out} |
|---|--|-----------------|------------------|
| Minimum | 107.3 | 0.3 | 0.3 |
| Maximum | 147.8 | 0.5 | 0.4 |
| Mean | 126.6 | 0.4 | 0.3 |
| Geometric Mean Median Permissible value | 127.9 128.5 | 0.4 0.4 | 0.3 0.3 |
| | 370 | 1 | 1 |

| Table 6 Annual effective dose rate components | | | | |
|---|--------------------------|-------------------------|----------------------------|--|
| Statistics | $E_{terr}(\mu Svy^{-1})$ | $E_{cos}(\mu Svy^{-1})$ | $E_{Cs-137}(\mu Svy^{-1})$ | |
| Minimum | | 368.7 | 0.1 | |
| | 60.7 | | | |
| Maximum | | 798.9 | 6.2 | |
| | 83.9 | | | |
| Mean | 72.0 | 503.8 | 1.6 | |
| Geometric Mean | 71.8 | 490.7 | 1.2 | |
| Median | 71.5 | 459.4 | 1.1 | |
| World's average | 70.0 | 380 | 3.0 | |

List of Figures

Fig. 1 Study area.

Fig. 2 Correlation between ²²⁶Ra and ²³²Th activity concentrations (a) (b)

Fig. 3 Correlation between major elements and radionuclides (a) FeO and 40 K (b) MnO and 137 Cs

Fig. 4 Box-Whisker plots for comparison of activity concentration in the soil of different hilly stations (a) 226 Ra (b) 232 Th (c) 40 K and (d) 137 Cs



Fig.1 Study area



Fig. 2



