Assessment of radiological hazards due to soil and building materials used in Mirpur Azad Kashmir; Pakistan

M. Rafique^{1*}, H. Rehman¹, Matiullah², F. Malik², M.U. Rajput², S.U. Rahman³, M.H. Rathore⁴

¹Department of Physics, University of Azad Jammu & Kashmir Muzaffarbad, Azad Kashmir, Pakistan ²Physics Division, PINSTECH, P.O. Nilore, Islamabad, Pakistan ³Department of Physics, COMSATS Institute of Information Technology, Islamabad, Pakistan ⁴AKMIDC, Muzaffarabad, Azad Kashmir, Pakistan

Background: Health hazards associated with exposure due to the natural radioactivity which is a part of our physical environment are of great concern. In order to assess the risks associated with exposure due to the natural radioactivity in soil and building materials, extensive studies have been carried out all over the world. The most commonly encountered radionuclide's are 238U, 232Th, their decay products and 40K. Materials and Methods: In order to study the concentration of these radionuclides soil, sand, gravel aggregates, bricks and marble, samples were collected from different sites and local suppliers of the district Mirpur, Azad Kashmir (Pakistan). 226Ra, 232Th, and 40K activities in the collected samples were measured using HPGe detector. The measured specific radioactivity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K in the studied samples ranged from 10 ± 1 to 47 \pm 2, 18 \pm 1 to 75 \pm 4 and 40 \pm 3 to 683 \pm 3 Bq.kg⁻¹, respectively. Results: From the measured activity concentration, radium equivalent activity, external and internal hazard indices, gamma and alpha indices, terrestrial absorbed dose and annual effective dose were calculated. Maximum value of radium equivalent activity of 197.1 ± 9 Bq.kg-1 was observed in soil sample whereas minimum value of 45.9 ± 2 Bq.kg-1 was found in gravel aggregates. Relatively higher mean values of hazard indices were found in brick samples. Annual effective dose varied from 0.06 ± 0.01 to 0.47 \pm 0.02 mSv.y⁻¹. Conclusion: Current values of annual effective dose, radium equivalent activity and hazard indices have been found to be within the recommended limits. Iran. J. Radiat. Res., 2011; 9(2): 77-87

Keywords: Building materials, radiological hazards, radium equivalent activity, annual effective dose, external and internal hazard indices.

INTRODUCTION

It is a well known fact that rocks and soil contain trace amounts of uranium and

thorium with their progenies which makes most of the naturally occurring materials slightly radioactive. In some concentration of radioactive elements in building materials (made up of earth's strata) may be high enough to result in significant exposure to the population. Presence of these naturally occurring radionuclides in earth crust strongly depends upon geological and geographical locations (1-4) (e.g., radionuclide concentrations in granite locations are higher relative to sand stones and limestone regions). Building materials (granite, gravel aggregates, bricks, marble, sand, soil) contain few parts per million of uranium and thorium, together with their radioactive progenies and ⁴⁰K. These radionuclides are considered as direct source of external and internal radiation exposure. 40K considered as a principal internal source of terrestrial radiations. From a total of 130 g of potassium in an average person weighing 70 kg, there is about 0.0157 g of ⁴⁰K. Therefore, total activity due to 40K in the body is approximately 0.11 µCi (4.07×10³ Bq) (5).

Soil and Building materials are known to contribute towards the radiation doses received by the general population ⁽⁶⁻⁴¹⁾. Therefore, knowledge of natural radioactivity levels is a pre-requisite in soil and building materials in order to set the standards and national guidelines in the

*Corresponding author:

Dr. Muhammad Rafique,

Department of Physics University of Azad Jammu & Kashmir, Muzaffarabad, 13100 Azad Kashmir, Pakistan.

Fax: +92 5822 960402 E-mail: mrafique@gmail.com

Muhammad Rafique, Habib-ur-Rehman, Matiullah, et al.

light of international recommendations. Hence, the prime objective of this work was to develop a base line reference data of natural radioactive elements present in the soil/building materials of the district Mirpur of the Azad Kashmir; Pakistan and to evaluate their radiological consequence if used in construction of buildings.

Geology of Mirpur

The studied area is underlain by sedimentary rocks of Siwalik Group of non-marine origin, ranging in age from Late Miocene to Pleistocene (figure 1). It is postulated that the sediments of the group were deposited in a slowly sinking basin under fresh water conditions. The total thickness of the sequence is about 5000 meters and constitutes a major portion of the whole Siwalik Group.

Nagri formation

The Nagri Formation is exposed in the north and northeastern part of the mapped area where, it is about 100 to 1450 meters

thick and consists of inter-bedded sandstone and clays. It consists of about 70% sandstone and 30% clays. The sandstones are greenish gray to light gray, massive, medium to coarse grained sandstones with subordinate intercalations of reddish brown mudstone, gritty and conglomeratic beds are found mat places. The formation has yielded fairly rich assemblage of vertebrate remains which indicates late middle Miocene to late Miocene age (42, 43).

Dhok pathan formation

The Dhok Pathan Formation is about 1500 to 2250 meters thick in the mapped area. The formation is represented by monotonous cyclic deposition of alternate sandstone and clay beds, exposed towards northern parts of the mapped area. The sandstones are gray, light gray, reddish brown and gleaming white, fine to medium grained, medium to thick bedded, with alternations of orange brown and dull red silty clay. Dhok Patan formation has conglomerate in the form of layers and lenses

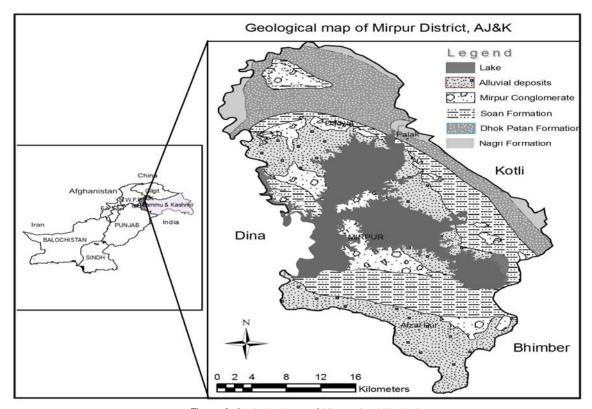


Figure 1. Geological map of Mirpur, Azad Kashmir

and it is the essential character of the upper part. The fauna present in this formation indicates as early to middle Pliocene age (44).

Soan formation

The formation comprised of yellowish gray clay and claystones, greenish gray, soft, massive sandstone and reddish brown clays. The unit consists of interbeds of conglomerate beds and clays. The conglomerate bands mainly consist of pebbles and boulders of limestone, quartzite, porphyritic rocks, sandstone, gneisses, schist, diabase etc. The pebbles and boulders range in size from 5 cm to 30 cm. The age of this formation is late Pliocene to early Pleistocene (44).

Mirpur conglomerates

The Mirpur Conglomerates are more than 250 meters thick and mainly comprised of unsorted conglomerates and mudstones having no bedding pattern. The conglomerates are composed of, unsorted, and well rounded pebbles and boulders of limestone, sandstone, quartzite and volcanic material, which range in size from 5 mm to 200 mm in diameter.

MATERIALS AND METHODS

In order to carry out this study, a total of 44 samples were collected from different locations of the district Mirpur, Azad Kashmir, Pakistan. These included 20 samples of soil, 6 samples of sand, 6 samples of gravel aggregates, 6 samples of bricks and 6 samples of marbles. The collected samples were crushed, sieved and dried at 110 ±1 °C for 20 h. These samples were then placed in Marinelli beakers and hermitically sealed. These beakers were then placed in an undisturbed position for a period of 60 days in order to attain equilibrium between ²²⁶Ra and ²²²Rn. To measure gamma spectra, a P-type HPGe detector was used. The resolution and relative efficiency of the detector for 1332 keV (60Co) was 2.5 keV and 90 %, respectively. The detector and preamplifier

were placed inside a low-background well-type lead shielding and cooled by liquid nitrogen from vertical dipstick cryostat. Energy and efficiency calibration of the detector were performed using standard point sources and mined nuclides gamma reference source (from AEA technology QSA GmH, Germany). Background signals were periodically recorded for the same time period and subtracted from each result obtained. Spectra were collected for about 12 hours in order to get sufficient counts at the desired peaks. ²²⁶Ra concentration was determined by means of its progeny photo peaks of gamma-ray lines: 214Pb (295.21 keV, 352 keV) and 214Bi (609 KeV, 1120.29 keV), ²³²Th was determined through its progeny photo peaks of gamma-ray lines: ²²⁸Ac (338.32 keV, 911.21 keV, 968.97 keV) whereas 40K activity was measured directly through its gamma-ray energy peak of 1460.83 keV. The software Personnel Computer Analyzer (PCA-II) was used for the collection of the spectra. The lowest limits of detection (LLD) for ²²⁶Ra, ²³²Th and $^{40}\mathrm{K}$ are found 1.2, 2.1 and 10 Bq kg⁻¹ respectively.

RESULTS AND DISCUSSION

Activity concentrations of various radio nuclides ²²⁶Ra, ²³²Th, and ⁴⁰K were calculated by using the following relation,

$$A = \frac{N_i}{\eta_i \times P_{ji} \times C_i \times t} \tag{1}$$

Where C_i the net counts, t is is the data collection time, $P_{\gamma i}$ is the emission probability and η_i is the efficiency of the detector for the corresponding peak.

The most important aspect is to determine the energy delivered by a radionuclide per unit mass of a substance which is known as specific activity (A_s).

$$A_s = \frac{A}{m} \tag{2}$$

Where "A" is activity concentration (Bq) and m is mass of the sample in kg.

Table's 1-5 shows measured values of specific activities of $^{26}Ra,\ ^{232}Th$ and $^{40}K,$ radium equivalent, external and internal hazard indices, $I_{\gamma},\ I_{\alpha}$, absorbed dose and annual effective dose along with Geometric Mean (GM) \pm Geometric Standard Deviation (STDEV) in soil, sand, gravel aggregates, bricks and marble samples.

As may be seen in table 1, activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the studied soil samples varied from 14 ± 1 to 47 ± 2 Bq kg⁻¹ , 26 ± 1 to 75 ± 4 Bqkg⁻¹ and

268±12 to 679±30 Bqkg⁻¹ with mean values of 27.4±8.4, 52±13, 478±111 Bqkg⁻¹ respectively. Maximum activity concentration of the ²²⁶Ra and ²³²Th has been observed in Soil-3 which was collected from the Mirpur University of Sciences and Technology (MUST) site whereas minimum concentration was observed in Soil-7 that was collected from the Chaichyan Mirpur site. Minimum concentration value of ⁴⁰K was also noted in Soil-7 whereas maximum value was observed in Soil-1 collected from the Industrial Area of the Mirpur Azad Kashmir.

Table 1. Measured values of the specific activity concentrations of 226 Ra, 232 Th and 40 K in the studied soil samples collected from the district Mirpur. R_{eq} , H_{ex} , H_{in} , I_{v} , I_{α} , absorbed dose and Annual effective dose values are also given in this table.

| Sample Code | ²²⁶ Ra (Bq.kg ⁻¹) | ²³² Th (Bq.kg ⁻¹) | ⁴⁰ K (Bq.kg ⁻¹) | Ra _{eq} | H _{ex} | H _{in} | lγ | Iα | D _{air} nGyh ⁻¹ | Annual effective dose (mSv) |
|----------------|---|---|---|------------------|-----------------|-----------------|-----------|-----------|--|-----------------------------------|
| Soil-1 | 39±2 | 74±4 | 679±30 | 197.10±9 | 0.53±0.03 | 0.64±0.03 | 0.72±0.04 | 0.19±0.01 | 94.8±5 | 0.47±0.025 |
| Soil-2 | 37±2 | 66±3 | 650±28 | 181.43±9 | 0.49±0.02 | 0.59±0.03 | 0.66±0.03 | 0.18±0.01 | 87.4±4 | 0.43±0.020 |
| Soil-3 | 47±2 | 75±4 | 539±24 | 195.75±9 | 0.53±0.03 | 0.66±0.03 | 0.70±0.04 | 0.23±0.01 | 92.9±5 | 0.46±0.025 |
| Soil-4 | 29±1 | 51±3 | 494±22 | 139.97±7 | 0.38±0.02 | 0.46±0.02 | 0.51±0.03 | 0.14±0.01 | 67.4±3 | 0.33±0.015 |
| Soil-5 | 23±1 | 40±2 | 460±20 | 115.62±6 | 0.31±0.01 | 0.37±0.02 | 0.42±0.02 | 0.11±0.01 | 56.1±3 | 0.28±0.015 |
| Soil-6 | 25±1 | 58±3 | 398±17 | 138.59±7 | 0.37±0.02 | 0.44±0.02 | 0.50±0.02 | 0.12±0.01 | 66.2±3 | 0.33±0.015 |
| Soil-7 | 14±1 | 26±1 | 268±12 | 71.82±4 | 0.19±0.01 | 0.23±0.01 | 0.26±0.01 | 0.07±0.01 | 34.7±2 | 0.17±0.010 |
| Soil-8 | 21±1 | 40±2 | 390±17 | 108.23±5 | 0.29±0.01 | 0.35±0.02 | 0.40±0.02 | 0.10±0.01 | 52.2±3 | 0.26±0.015 |
| Soil-9 | 24±1 | 47±2 | 405±18 | 122.40±6 | 0.33±0.01 | 0.40±0.02 | 0.45±0.02 | 0.12±0.01 | 58.8±3 | 0.29±0.015 |
| Soil-10 | 26±1 | 59±3 | 589±26 | 155.72±8 | 0.42±0.02 | 0.49±0.02 | 0.57±0.03 | 0.12±0.01 | 75.5±4 | 0.37±0.020 |
| Soil-11 | 22±1 | 39±2 | 342±15 | 104.10±5 | 0.28±0.01 | 0.34±0.02 | 0.37±0.02 | 0.10±0.01 | 49.9±2 | 0.25±0.010 |
| Soil-12 | 22±1 | 42±2 | 352±15 | 109.16±5 | 0.29±0.01 | 0.35±0.02 | 0.39±0.02 | 0.10±0.01 | 52.3±3 | 0.26±0.015 |
| Soil-13 | 32±2 | 52±3 | 503±22 | 145.09±7 | 0.39±0.02 | 0.48±0.03 | 0.53±0.03 | 0.15±0.01 | 69.7±3 | 0.34±0.015 |
| Soil-14 | 41±2 | 73±4 | 474±21 | 181.89±9 | 0.49±0.02 | 0.60±0.03 | 0.66±0.03 | 0.20±0.01 | 86.2±4 | 0.42±0.020 |
| Soil-15 | 23±1 | 54±3 | 572±25 | 144.26±7 | 0.39±0.02 | 0.45±0.02 | 0.53±0.03 | 0.11±0.01 | 70.2±3 | 0.34±0.015 |
| Soil-16 | 25±1 | 57±3 | 518±23 | 146.40±7 | 0.40±0.02 | 0.46±0.02 | 0.54±0.03 | 0.12±0.01 | 70.7±3 | 0.35±0.015 |
| Soil-17 | 34±2 | 64±3 | 562±25 | 168.79±8 | 0.46±0.02 | 0.55±0.03 | 0.62±0.03 | 0.17±0.01 | 81.1±4 | 0.40±0.020 |
| Soil-18 | 20±1 | 41±2 | 417±18 | 110.74±6 | 0.30±0.01 | 0.35±0.02 | 0.41±0.02 | 0.10±0.01 | 53.6±3 | 0.26±0.015 |
| Soil-19 | 38±2 | 61±3 | 596±26 | 171.12±8 | 0.46±0.02 | 0.56±0.03 | 0.62±0.03 | 0.19±0.01 | 82.2±4 | 0.40±0.020 |
| Soil-20 | 28±1 | 55±3 | 610±27 | 153.62±7 | 0.41±0.02 | 0.49±0.02 | 0.57±0.03 | 0.14±0.01 | 74.6±4 | 0.37±0.020 |
| GM± STDEV | 27.4±8.4 | 52±13 | 478±111 | 139±33.7 | 0.38±0.09 | 0.45±0.11 | 0.51±0.12 | 0.14±0.04 | 66.9±16.04 | 0.33±0.08 |
| average | 28.50 | 53.70 | 490.90 | 143.09 | 0.39 | 0.46 | 0.53 | 0.14 | 68.83 | 0.34 |

In sand samples (table 2), specific activity concentration of 226Ra, 232Th and ⁴⁰K has been found to range from 18± 1 to 29 ± 1 Bq kg⁻¹, 30 ± 2 to 48 ± 2 Bq.kg⁻¹ and 345 ± 15 to 545 ± 24 Bq.kg⁻¹ with mean values of 2.6 ±4.7, 39.6±7, 423.3±82.9 Bq.kg⁻¹, respectively. Maximum activity concentration of the ²²⁶Ra was found in Sand-2 which was collected from the Wazirabad site whereas minimum concentration observed in Sand-3 and 5 that were collected from the Jehlum city and Jatlaan. For ²³²Th, maximum activity concentration was noted in the sand sample which was collected from the Khokhran Mirpur (Sand-4) and a minimum value was found in sample collected from the Jatlaan Mirpur (Sand -5). Minimum concentration value of ⁴⁰K was also noted in sand-5 whereas maximum value was observed in sand-2, collected from the Industrial Area of the Mirpur Azad Kashmir.

In gravel aggregates (see table 3), specific activities of ²⁶Ra, ²³²Th and ⁴⁰K range from 10± 1 to 36±2 Bq kg-1, 18±1to 29±2 Bq.kg⁻¹ and 45±2 to 223±1Bq.kg⁻¹ with mean values of 15.8 ± 9.4 , 24.2 ±3.8 , 132.9±58.5 Bq.kg⁻¹, respectively. Maximum activity concentration of the ²²⁶Ra was found in sample of gravel aggregates (A-5) collected from the Mangla site whereas minimum concentration was observed in A-6 that was collected from the Lehrdi Mirpur. Minimum concentration values of ²³²Th and ⁴⁰K was found in A-6 and A-5 which were collected from Mangla and Lehrdi respectively, whereas maximum value was observed in A-4 which was collected from the Phalot Mirpur.

In brick samples (table 4), ²²⁶Ra, ²³²Th

Table 2. Measured values of the specific activity concentrations of 226 Ra, 232 Th and 40 K in the studied sand samples collected from the district Mirpur. R_{eq} , H_{ex} , H_{in} , I_{γ} , I_{α} , absorbed dose and Annual effective dose values are also given in this table.

| Sample Code | ²²⁶ Ra (Bq.kg ⁻¹) | ²³² Th (Bq.kg ⁻¹) | ⁴⁰ K (Bq.kg ⁻¹) | Req | H _{ex} | H _{in} | lγ | lα | Dair nGyh ⁻¹ | Annual effective dose mSv |
|----------------|---|---|---|----------|-----------------|-----------------|-----------|-----------|----------------------------|---------------------------------|
| Sand-1 | 25±1 | 43±2 | 417±18 | 115.68±6 | 0.32±0.01 | 0.39±0.02 | 0.44±0.02 | 0.13±0.01 | 57.1±3 | 0.28±0.015 |
| Sand-2 | 29±1 | 44±2 | 517±23 | 128.11±6 | 0.36±0.02 | 0.43±0.02 | 0.49±0.02 | 0.15±0.01 | 63.7±3 | 0.31±0.015 |
| Sand-3 | 18±1 | 43±2 | 393±17 | 107.00±5 | 0.30±0.01 | 0.35±0.02 | 0.41±0.02 | 0.09±0.01 | 53.1±2 | 0.26±0.010 |
| Sand-4 | 27±1 | 48±2 | 545±24 | 133.79±7 | 0.37±0.02 | 0.44±0.02 | 0.51±0.02 | 0.14±0.01 | 66.7±3 | 0.33±0.015 |
| Sand-5 | 18±1 | 30±2 | 345±15 | 85.05±4 | 0.24±0.01 | 0.28±0.02 | 0.33±0.02 | 0.09±0.01 | 42.4±2 | 0.21±0.010 |
| Sand-6 | 21±1 | 33±2 | 361±16 | 93.46±5 | 0.26±0.01 | 0.32±0.02 | 0.36±0.02 | 0.11±0.01 | 46.3±2 | 0.23±0.010 |
| GM± STDEV | 22.6±4.7 | 39.6±7 | 423.3±82.9 | 109±19 | 0.30±0.05 | 0.36±0.06 | 0.42±0.07 | 0.11±0.02 | 54.2±9.6 | 0.27±0.05 |
| average | 23 | 40.17 | 429.7 | 110.5 | 0.31 | 0.37 | 0.42 | 0.12 | 54.9 | 0.27 |

Table 3. Measured values of the specific activity concentrations of 226 Ra, 232 Th and 40 K in the studied gravel aggregate samples collected from the district Mirpur. R_{eq}, H_{ex}, H_{in}, I_y, I_{\alpha}, absorbed dose and Annual effective dose values are also given in this table.

| Sample Code | ²²⁶ Ra (Bq.kg ⁻¹) | ²³² Th (Bq.kg ⁻¹) | ⁴⁰ K (Bq.kg ⁻¹) | Req | H _{ex} | H _{in} | lγ | lα | Dair nGyh ⁻¹ | Annual effective dose mSv | | |
|-------------|---|---|---|-----------|-----------------|-----------------|-----------|-----------|----------------------------|---------------------------------|--|--|
| A-1 | 13±1 | 23±1 | 169±7 | 58.90±3 | 0.16±0.01 | 0.19±0.01 | 0.21±0.01 | 0.07±0.01 | 28.0±1 | 0.14±0.015 | | |
| A-2 | 14±1 | 25±1 | 157±7 | 61.84±3 | 0.17±0.01 | 0.20±0.01 | 0.22±0.01 | 0.07±0.01 | 29.3±1 | 0.14±0.015 | | |
| A-3 | 14±1 | 27±1 | 157±7 | 64.70±3 | 0.17±0.01 | 0.21±0.01 | 0.23±0.01 | 0.07±0.01 | 30.6±1 | 0.15±0.010 | | |
| A-4 | 17±1 | 29±2 | 223±1 | 75.64±4 | 0.20±0.01 | 0.25±0.01 | 0.28±0.01 | 0.09±0.01 | 36.0±2 | 0.18±0.015 | | |
| A-5 | 36±2 | 25±1 | 45±2 | 75.22±4 | 0.20±0.01 | 0.30±0.02 | 0.26±0.01 | 0.18±0.01 | 33.9±1 | 0.17±0.010 | | |
| A-6 | 10±1 | 18±1 | 132±6 | 45.90±2 | 0.12±0.01 | 0.15±0.01 | 0.17±0.01 | 0.05±0.01 | 21.9±1 | 0.11±0.010 | | |
| GM± STDEV | 15.81±9.4 | 24.2±3.8 | 132.9±58.5 | 62.8±11.1 | 0.17±0.03 | 0. 21±0.05 | 0.23±0.04 | 0.08±0.05 | 29.6±4.9 | 0.15±0.02 | | |
| average | 17.3 | 24.5 | 147.17 | 63.7 | 0.17 | 0.22 | 0.23 | 0.09 | 29.9 | 0.15 | | |

and ⁴⁰K activities varied from 26±1 to 46±2 Bq kg⁻¹, 52±3 to 63±3 Bqkg⁻¹ and 554±2 to 683±3 Bqkg⁻¹ with mean values of 38.9±8.2, 59.4±3.9, 630.8±44.9 Bqkg⁻¹ respectively. Maximum value of specific activity concentration of ²²⁶Ra was found B-3 which was collected from the M.A Mirpur site whereas minimum value was found in B-5 collected from the A.G Mirpur site. Minimum specific activity concentrations of ²³²Th and ⁴⁰K were found in B-4 and B-5 which were collected from the A.A.P Mirpur and A.G Mirpur sites, respectively.

In marble samples (see, table 5), ²²⁶Ra, ²³²Th and ⁴⁰K activities varied from 18±1 to 41±3 Bq kg⁻¹, 18±1 to 33±2 Bqkg⁻¹ and 40±3to 82±4 BqKg⁻¹ with mean values of 27.8±8.9, 24.4±6, 59.3±16.4 BqKg⁻¹ respectively. Maximum value of specific activity concentration of ²²⁶Ra was found M-4 which was collected from the M.A Mirpur site

whereas minimum value was found in M-3 collected from the A.G Mirpur site. Minimum specific activity concentrations of ²³²Th and ⁴⁰K were found in M-6 and M-5 and maximum were found in M-5 and M-1 respectively, which were collected from the A.A.P Mirpur and A.G Mirpur sites, respectively.

Assessment of radiological hazards

The measured specific activity concentrations were used to assess the radiological hazards associated with soil, sand, gravel aggregates, bricks and marbles of the studied area. As more than one radionuclide contributes towards the gamma doses (i.e. ²²⁶Ra, ²³²Th and ⁴⁰K), therefore radiological hazards are presented in terms of a single quantity called 'hazard index'.

| Table 4. Measured values of the specific activity concentrations of ²²⁶ Ra, ²³² Th and ⁴⁰ K in the studied brick samples collected from |
|---|
| the district Mirpur. R_{eq} , H_{ex} , H_{in} , I_y , I_α , absorbed dose and Annual effective dose values are also given in this table. |

| Sample Code | ²²⁶ Ra (Bq.kg ⁻¹) | ²³² Th (Bq.kg ⁻¹) | ⁴⁰ K (Bq.kg ⁻¹) | Req | H _{ex} | H _{in} | lγ | lα | Dair nGyh ⁻¹ | Annual effective dose mSv |
|----------------|---|---|---|-----------|-----------------|-----------------|-----------|------------|----------------------------|---------------------------------|
| B-1 | 44±2 | 61±3 | 619±3 | 178.89±9 | 0.48±0.03 | 0.60±0.02 | 0.66±0.02 | 0.22±0.01 | 45.41±4 | 0.22±0.020 |
| B-2 | 45±2 | 59±3 | 632±3 | 178.03±9 | 0.48±0.03 | 0.60±0.02 | 0.66±0.02 | 0.23±±0.01 | 86.77±3 | 0.43±0.015 |
| B-3 | 46±2 | 62±3 | 639±3 | 183.86±9 | 0.50±0.03 | 0.62±0.02 | 0.68±0.02 | 0.23±±0.01 | 86.18±3 | 0.42±0.015 |
| B-4 | 33±2 | 52±3 | 554±2 | 150.02±7 | 0.41±0.03 | 0.49±0.02 | 0.55±0.02 | 0.17±0.01 | 78.96±3 | 0.39±0.015 |
| B-5 | 26±1 | 63±3 | 683±3 | 168.68±8 | 0.46±0.03 | 0.53±0.02 | 0.63±0.02 | 0.13±0.01 | 74.89±3 | 0.37±0.015 |
| B-6 | 44±2 | 60±3 | 666±3 | 181.08±9 | 0.49±0.03 | 0.61±0.02 | 0.67±0.02 | 0.22±0.01 | 89.13±3 | 0.44±0.015 |
| GM± STDEV | 38.9±8.2 | 59.4±3.9 | 630.8±44.9 | 173 ±12.6 | 0.47±0.03 | 0.57±0.05 | 0.64±0.04 | 0.19±0.04 | 75.1±16.3 | 0.37±0.08 |
| average | 39.67 | 59.50 | 632.17 | 173.43 | 0.47 | 0.58 | 0.64 | 0.20 | 76.89 | 0.38 |

Table 5. Measured values of the specific activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in the studied Marble samples collected from the district Mirpur. R_{eq}, H_{ex}, H_{in}, I_y, I_a, absorbed dose and Annual effective dose values are also given in this table.

| Sample Code | ²²⁶ Ra (Bq.kg ⁻¹) | ²³² Th (Bq.kg ⁻¹) | ⁴⁰ K (Bq.kg ⁻¹) | Req | H _{ex} | H _{in} | lγ | lα | Dair nGyh ⁻¹ | Annual effective dose mSv |
|----------------|---|---|---|------------|-----------------|-----------------|------------|-----------|----------------------------|---------------------------------|
| Marble-1 | 21±1 | 19±1 | 82±4 | 54.48±4 | 0.15±0.01 | 0.20±0.01 | 0.19±0.01 | 0.11±0.01 | 12.5±1 | 0.06±0.005 |
| Marble-2 | 36±2 | 31±2 | 78±4 | 86.34±4 | 0.23±0.01 | 0.33±0.02 | 0.30±0.01 | 0.18±0.01 | 31.3±2 | 0.15±0.01 |
| Marble-3 | 18±1 | 23±1 | 62±4 | 55.66±4 | 0.15±0.01 | 0.20±0.01 | 0.20±0.01 | 0.09±0.01 | 30.9±1 | 0.15±0.005 |
| Marble-4 | 41±3 | 26±1 | 56±4 | 82.49±4 | 0.22±0.01 | 0.33±0.02 | 0.29±0.02 | 0.21±0.01 | 35.1±2 | 0.17±0.01 |
| Marble-5 | 32±2 | 33±2 | 40±3 | 82.27±4 | 0.22±0.01 | 0.31±0.02 | 0.29±0.02 | 0.16±0.01 | 32.6±2 | 0.16±0.01 |
| Marble-6 | 26±1 | 18±1 | 49±4 | 55.51±4 | 0.15±0.01 | 0.22±0.01 | 0.19±0.01 | 0.13±0.01 | 35.1±2 | 0.17±0.01 |
| GM± STDEV | 27.8±8.9 | 24.4±6 | 59.3±16.4 | 67.97±15.7 | 0.18±0.04 | 0.26±0.06 | 0.24± 0.05 | 0.14±0.04 | 28 ±8.6 | 0.14± 0.04 |
| average | 29 | 25 | 61.2 | 69.5 | 0.19 | 0.27 | 0.24 | 0.15 | 29.60 | 0.15 |

$$Ra_{eq} = A_{Ra} + (A_{Th} \times 1.43) + (A_k \times 0.077)$$
 (5)

Where, A_{Ra}, A_{Th} and A_k are ²²⁶Ra, ²³²Th ⁴⁰K specific activity concentrations. From the radiological point of view, Ra_{eq}≤370BqKg⁻¹. In the case when Ra_{eq} \leq 370BqKg⁻¹ then external gamma dose will be less than 1.5 mGy y⁻¹ (28, 29). Radium equivalent activity (Ra_{eq}) has been calculated for the studied soil and building material samples and results obtained are listed in tables 1-5. For soil samples Ra_{eq} varies from 71.82±4 to 197.10±9 Bqkg⁻¹, with mean value of 139.09±33.7 Bq kg⁻¹. For sand, gravel aggregates, bricks and marble samples Raea activity varied from 85.05±4 to 128.11±6, 45.90±2 to 75.64±4, 150.02±7 and 54.48±4 to 86.34±4 Bq kg⁻¹, with mean values of 109±19, 62.8±11.1, 173±12.6 and 67.97±15.7 Bakg⁻¹ respectively.

External hazard index

In order to assess the external radiological hazards from building materials, external hazard index (H_{ex}) is calculated using the following expression $^{(30)}$,

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{6}$$

To limit the external gamma radiation dose from building materials below 1.5 mGy y-1 the external hazard index, Hex should obey the following relation $H_{ex} \le 1$ (29). External hazard indices for the soil, sand, gravel aggregate, bricks and marble samples varied from 0.19±0.01 to 0.53±0.03, 0.24 ± 0.01 to 0.37 ± 0.02 , 0.12 ± 0.01 0.20 ± 0.01 , 0.41 ± 0.03 to 0.50 ± 0.03 and 0.15 ± 0.01 to 0.23 ± 0.01 with mean values of 0.38 ± 0.09 , 0.30 ± 0.05 , 0.17 ± 0.03 , 0.47 ± 0.03 and 0.18±0.04. The external hazard index for the studied samples is less than unity and therefore these building materials are safe to be used for construction.

Internal hazard index

The internal radiation hazards due to the inhalation of radon and its short-lived products are assessed using (30),

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \tag{7}$$

Like external hazard index. the construction materials would be safe if H_{in}≤1. Internal Hazard indices for the soil, sand, gravel aggregate, bricks and marble samples in current study varied from 0.23 ± 0.01 to 0.66 ± 0.03 , 0.28 ± 0.02 0.44 ± 0.02 , 0.15 ± 0.01 to $0.30 \pm$ 0.02, 0.49 ± 0.02 to 0.62 ± 0.02 and 0.20 ± 0.01 to 0.33 ± 0.02 , with mean values of 0.45 ± 0.11 , 0.36 ± 0.06 , 0.21 ± 0.05 , 0.57 ± 0.05 0.26±0.06 respectively. The internal hazard index for the studied samples of building materials is less than unity which indicates that the studied building materials are safe to be used for construction in the Mirpur.

Gamma index (ly)

European Commission has suggested a gamma activity concentration index, I_{γ} for defining radiation risk from excessive gamma exposure by the following relation (31, 32).

$$I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_{K}}{3000} \tag{8}$$

Values of index I_y≤2 corresponds to a dose rate criterion of 0.3 mSv y⁻¹, whereas 2≤ I_y≤6 corresponds to a criterion of 1 mSvy⁻¹ (31, 33). Thus the material with Iy>6 should be avoided to use as building material, since these values correspond to the dose rates higher than 1 mSv y-1 which is highest then recommended values (34). In the current study, I_{ν} was calculated using equation (8). The gamma index value ranged from 0.72 ± 0.04 , 0.33 ± 0.02 0.26 ± 0.01 to 0.51 ± 0.02 , 0.17 ± 0.01 to 0.28 ± 0.01 , 0.55 ± 0.02 to 0.68±0.02 and 0.19±0.01 to 0.30±0.01 for soil, sand, gravel aggregates, bricks and marbles samples, respectively, with mean values of 0.51 ± 0.12 , 0.42 ± 0.07 , 0.23 ± 0.04 , 0.64 ± 0.04 and 0.24 ± 0.05 respectively. All the current 'I_γ' values of the studied samples follow the criterion $(I_{\nu} \leq 2)$ therefore it may be concluded that the construction material used in the district Mirpur is safe

and does not pose any significant health hazards.

Alpha index (Ia)

The excess alpha radiation due to the radon inhalation originating from the building materials is assessed through the alpha index (I_{g}) which is defined as follows ⁽³²⁾.

$$I_{\gamma} = \frac{A_{Ra}}{200} \tag{9}$$

The recommended exemption and levels ofrecommended upper concentrations in building materials are 100 Bq kg⁻¹ and 200 Bq kg⁻¹ (35). When the ²²⁶Ra activity concentration of building materials exceeds the value of 200 Bq.kg-1, it is possible that radon exhalation from this material may cause indoor radon concentration greater than 200 Bq.m⁻³. On the other hand, if ²²⁶Ra concentration is less than 100 Bq.kg⁻¹, than resulting indoor radon concentration is less than 200 Bq.m⁻³ (35). These considerations are reflected in the alpha index. The recommended limit concentration of 226 Ra is 200 Bq.kg⁻¹, for which $I_a = 1$. Using equation (9), alpha index was calculated. The values of I_{α} ranged from to 0.23 ± 0.01 , 0.09 ± 0.01 0.07 ± 0.01 0.15 ± 0.01 , 0.05 ± 0.01 to 0.18 ± 0.01 , 0.13 ± 0.01 to 0.23±±0.01 and 0.09±0.01 to 0.21±0.01 for soil, sand, gravel aggregates, bricks and marbles samples with mean values of 0.14±0.04, 0.11±0.02, 0.08±0.05, 0.19±0.04 and 0.14±0.04, respectively. These observed values are less than unity showing that construction materials are safe from the point of view of environmental radiation hazards.

Radiation doses

In order to calculate the dose rate in air from the samples collected from the area under study, the following relation was used (36)

$$\dot{D} = \sum_{x} A_{x} \times C_{x} \tag{10}$$

Where $A_x(Bq.kg^{-1})$ is the mean activity of ^{226}Ra , ^{232}Th and ^{40}K and C_x (nGyh⁻¹ per Bq kg⁻¹) is the corresponding dose conversion factor. The dose conversion factors used in the calculation for ^{226}Ra , ^{232}Th and ^{40}K were 0.427, 0.662 and 0.043, respectively $^{(36)}$.

As may be seen from table 1-6, absorbed dose rates for soil, sand, gravel aggregates, bricks and marbles varies from 34.7 ± 2 to 94.8 ± 5 , 42.4 ± 2 to 66.7 ± 3 , 21.9 ± 1 to 36.0 ± 2 , 45.41 ± 4 to 89.13 ± 3 and 12.5 ± 1 to 35.1 ± 2 nGyh⁻¹ with mean values 66.9 ± 16.04 , 54.2 ± 9.6 , 29.6 ± 4.9 , 75.1 ± 16.3 and 28 ± 8.6 nGyh⁻¹.

The annual effective dose equivalent to be received by the public due to the activity

Table 6. Comparison of mean radium equivalent activity observed in the current study with some of the data available in the literature (Bq kg⁻¹).

| Country | Soil | Sand | Gravel /Aggregates | Clay Bricks | Marble/ limestone | Reference |
|----------------------|-------|-------|-----------------------|-------------|----------------------|---------------|
| Algeria | - | 28 | 58 | 130 | 37 | (13) |
| Australia | - | 70 | 115 | - | - | (30) |
| China | - | 96 | 82 | - | - | (24) |
| Germany | - | 59 | 322 | - | - | (29) |
| India | - | 170.8 | - | 151.7 | - | (22) |
| Pakistan Punjab | 149 | 91 | - | 106 | 82 | (38) |
| Pakistan (Punjab) | 141 | - | - | - | - | (25) |
| Punjab | 158.5 | - | - | - | - | (23) |
| Punjab | 122 | - | - | - | - | (14) |
| Isl/Rwp | 211 | - | - | - | - | (34) |
| Lahore | 252 | - | - | - | - | (34) |
| MirPur, Azad Kashmir | 143±7 | 113±5 | 45±2 | 174±8 | 69±4 | Present study |

84

in soil and building materials has been calculated using the following relation (36).

$$E = T \times Q \times D \times O_f \times 10^{-6} \tag{11}$$

Where the value of Q is 0.7 SvGy⁻¹ y⁻¹ for environmental exposure to gamma rays of moderate energy, T is time in hours in one year, i.e., 8760h, O_f is the occupancy factor (0.8), and "D" is the dose rate given in equation (3). The mean annual effective dose equivalent in the six districts of Punjab, as calculated by equation (4), was about 0.34 mSvy⁻¹, which is within the permissible dose equivalent limit (i.e., 1 mSvy⁻¹) (37). Having determined the mean annual effective dose equivalent,

The annual effective dose (see table 1-6) for soil, sand, gravel aggregates, bricks and marbles varies from 0.170 ± 0.010 to 0.47 ± 0.025 , 0.21 ± 0.01 to 0.33 ± 0.015 , 0.11 ± 0.010 to 0.18 ± 0.015 , 0.22 ± 0.020 to 0.44 ± 0.015 and 0.06 ± 0.005 to 0.17 ± 0.010 mSv with mean values 0.33 ± 0.08 , 0.27 ± 0.05 , 0.15 ± 0.02 , 0.37 ± 0.08 and 0.14 ± 0.04 mSv respectively.

Figure 2 gives geometric mean (G.M) values of specific activities due to ²²⁶Ra, ²³²Th, ⁴⁰K and Ra_{eq}. As may be seen from figure 2, bricks are major source of activities followed by soil and sand samples. In table 6. mean radium equivalent activities of present study have been compared with those published by the other groups. Current values of radium equivalent, specific activity for soil, sand, gravel aggregates, bricks and marble samples are greater than the values reported for some countries like Algeria, Australia, and china. For soil samples, radium equivalent activity observed in current study is less than the values reported for other parts of the country like Islamabad/Rawalpindi, Lahore and other cities of the Punjab. From table 1-6, it may be seen that activities due to ²²⁶Ra, ²³²Th and ⁴⁰K for the sample collected from district Mirpur of Azad Kashmir are within the world range reported by UNSCEAR 2000 (for ²²⁶Ra 17-60 Bq kg⁻¹, for ²³²Th 11 64 Bq kg^{-1} and for $^{40}\text{K}\ 140\text{-}850 \text{ Bq kg}^{-1}$).

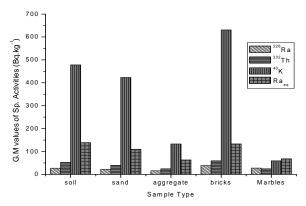


Figure 2. Variations in geometric mean of specific activities due to 226 Ra, 232 Th, 40 K and Ra_{eq}.

CONCLUSION

Specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K in soil and building materials collected from different sites of the district Mirpur have been measured using HPGe based gamma spectroscopy technique. The radium equivalent activity, external and internal hazard indices, gamma and alpha indices, terrestrial absorbed dose and effective dose have been determined to radiological assess the hazards from building materials used in studied area.

Radium equivalent activity, all the hazard indices and Mean annual effective dose have been found to be within recommended limits. In view of the current study, studied soil and building material would not pose any significant source of radiation hazard if used in the construction of buildings.

ACKNOWLEDGEMENTS

We are thankful to Higher Education Commission of Pakistan and the University of Azad Jammu & Kashmir for providing funds and Director PINSTECH for providing us research facilities at their laboratories.

REFERENCES

1. Durrani SA and Iliç R (1997) Radon Measurements by

Muhammad Rafique, Habib-ur-Rehman, Matiullah, et al.

- etched track detectors: Applications in radiation protection, earth sciences and the environment. Singapore: World Scientific Publishing Co. Pte Ltd. ISBN 9810226667.
- Durrani SA and Bull RK (1987) Solid State Nuclear Track Detection. Principles, Methods and Applications, Pergamon Press.
- Miller KM and Shebell P (1993) In situ gamma ray spectrometry. A tutorial for environmental scientists. US-DOSE publication, EML-557 (New York: Environmental Measurement Laboratory).
- Martin A and Harbison S (2006) An introduction to radiation protection, 5th edition ISBN-10 0 340 885 432, ISBN-13 978 0 340 885 437.
- Lamarsh JR and Barrata AJ (2001) Introduction to Nuclear Engineering, 3rd edition. Prentice Hall.
- Khan E, Tahseen R, Din NA, Matiullah, Ansari F, Hao HX, Wang YL, Guo SL (1991) Environmental radioactivity in D.I. Khan and its adjacent areas—Pakistan. *Nucl Tracks & Radiat Meas*, 19: 761–764.
- Ahmad N, Matiullah, Khatibeh AJAH (1997) Indoor Radon Levels and Natural Radioactivity in Jordanian Soil. Radiat. Protect. Dosim. 71: 231-233.
- Khatibeh AJAH, Maly A, Ahmad N, Matiullah (1997): Natural Radioactivity in Jordanian Construction Materials. Radiat Protect Dosim, 69: 143-147.
- Quindos LS, Newton GJ, Fernandez PL, Soto J (1988) Natural radioactivity of some Spanish building materials. Sci Total Environ, 68: 181-185.
- 10. Khatibeh AJAH, Ahmad N, Matiullah, Kenawy MA (1997) Natural radioactivity in marble stones Jordan. *Radiat Meas*, **28**: 345-348.
- 11. Nasir A, Matiullah, Hussein AJA (1998) Determination of natural radioactivity in Jordanian soil and Building materials and the Associated Radiation Hazards. Jr Environ Radioactivity, 39: 9-22.
- 12. Rahman S, Mati N, Matiullah, Ghauri B (2007) Radon exhalation rate from the soil, Sand and brick samples collected from NWFP and FATA, Pakistan. Radiation Protection Dosimetry 124: 392-399.
- 13. Amrani D and Tahtat M (2001) Natural radioactivity in Algerian building materials. *Appl Radiat Isot,* **54:** 687-690
- 14. Rahman S, Matiullah, Mujahid SA, Hussain S (2008) Assessment of radiological hazards due to the presence of natural radionuclides in samples of building materials collected from the northwestern areas of Pakistan. J Radiol Prot, 28: 205-212.
- Zikovsky L and Kennedy G (1992) Radioactivity of building materials available in Canada. Health Phys, 63: 449 -452.
- Rahman S, Faheem M, Matiullah ? (2008) Natural Radioactivity Measurements in Pakistan An Overview. J Radiol Prot, 28: 443-452.
- 17.Ali S, Tufail M, Jamil K, Ahmad A, Khan HA (1996) Gamma ray activity and dose rate of brick samples from areas of North West Frontier Province (NWFP), Pakistan. Sci Total Environ, 187: 247-252.
- 18. Rahman SU, Matiullah, Anwar J (2009) Assessment of the dose received by students and staff in schools in the Rawalpindi region of Pakistan due to indoor radon. *Jr. Radiological Protection*, **29**: 273-277.
- 19. Chowdhury IM, Alam MN, Ahmed AKS (1998) Concen-

- tration of radionuclide's in building and ceramic materials of Bangladesh and evaluation of radiation hazard. *J Radioanal Nucl Chem*, **231**: 117-122.
- Rahman SU, Faheem M, Anwar J, Ziafat M, Nasir T, Matiullah (2009) External dose assessment from the measured radioactivity in soil samples collected from the Islamabad capital territory, Pakistan. *Jr Radiol Prot*, 29: 499-505.
- 21. Kovler K, Haquin G, Manasherov V, Neeman E, Lavi N (2002) Natural radionuclide's in building materials available in Israel. *Building and Environment*, 37: 531-537
- 22. Kumar V, Ramachandran TV, Prasad R (1999) Natural radioactivity of Indian building materials and by-products. Applied Radiation and Isotopes, 51: 93-96.
- 23.Matiullah, Ahad A, Rehman S, Rehman S, Faheem M (2004) Measurement of radioactivity in the soil of Bahawalpur division, Pakistan. Radiat Prot Dosim, 112: 443 –447.
- 24.Xinwei L (2005) Natural radioactivity in some building materials of Xi'an, China. *Radiat Meas*, **40**: 94-97.
- 25.Tahir SNA, Jamil K, Zaidi J H, Arif M, Ahmed N, Ahmed S A (2005) Measurements of activity concentrations of naturally occurring radionuclides in soil samples from Punjab province of Pakistan and assessment of radiological hazards. Rad Prot Dosim, 113: 421–427.
- 26.Roy S, Alam MS, Begum M, Alam B (2005) Radioactivity in building materials used in and around Dhaka city. *Radiat Protect Dosimetry,* **114:** 527-532.
- 27.0ECD (1979) Exposure to radiation from natural radioactivity in building materials. Report by a group of experts of the OECD Nuclear Energy Agency, Paris, France.
- 28.Krisiuk EM, Tarasov SI, Shamov VP, Shalak NI, Lisa CEP, Gomelsky LG (1971) A Study on Radioactivity in Building Materials. Research Institute for Radiation Hygiene, Leningrad.
- 29.Krieger VR (1981) Radioactivity of construction materials. Betonwerk Fertigteil Tech, 47: 468-473.
- 30.Beretka J and Mathew PJ (1985) Natural radioactivity of Australian building materials, industrial wastes and byproducts. Health Phys, 48: 87-95.
- 31.European Commission (EC) (1999) Radiation Protection 112-radiological protection principles concerning the natural radioactivity of building materials Directorate-General Environment. Nuclear safety and civil Protection.
- 32.Righi S and Bruzzi L (2006) Natural radioactivity and radon exhalation in building materials used in Italian dwellings. J Environ Radioact, 88: 158-170.
- 33.Anjos RM, Veiga R, Soares T, Santos AMA, Aguiar JG, Frasca MHBO, Brage JAP, Uzeda D, Mangia L, Facure A, Mosquera B, Carvalho C, Gomes PRS (2005) Natural radionuclide distribution in Brazilian commercial granites. Radiation measurement, 39: 245-253.
- 34.UNSCEAR (2000) Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations Publication, New York, USA.
- 35. The Radiation Protection in Denmark, Iceland, Norway and Sweden (2000) Naturally occurring Radiation in the Nordic Countries- recommendations The Flag-Book Series (Reykjavik).
- 36.United Nations Scientific Committee on the Effect of

- Atomic Radiation. Sources, effects and risks of ionizing radiation, (NY: UNSCEAR) (1988).
- 37.ICRP 60 (1990) Recommendations of the International Commission on Radiological Protection. ICRP Publication 60. Annals of the ICRP, Pergamon Press, Oxford.
- 38. Faheem M, Mujahid SA, Matiullah (2008) Assessment of radiological hazards due to the natural radioactivity in soil and building material samples collected from six districts of the Punjab province-Pakistan. *Radiation Measurements*, **43**: 1443–144.
- 39.Tufail M (1992) Radon and gamma activity measurements for determination of radiation doses, assessment of cancer risks and application to geology / geophysics; Ph. D Thesis, Department of Physics, University of the Punjab, Lahore, Pakistan.
- 40.Changizi V, Jafarpoor Z, Naseri M (2010) Measurement of ²²⁶Ra, ²²⁸Ra, ¹³⁷Cs and ⁴⁰K in edible parts of two

- types of leafy vegetables cultivated in Tehran Province-Iran and resultant annual ingestion radiation dose. *Iran J Radiat Res, 8*: 103-110.
- 41. Rahman SU, Matiullah, Malik F, Rafique M, Anwar J, Ziafat M, Jabbar A (2010) Measurement of naturally occurring/fallout radioactive elements and assessment of annual effective dose in soil samples collected from four districts of the Punjab Province, Pakistan. Journal of radioanalytical and nuclear chemistry, 287: 647-655
- 42. Lewis (1937) A new Siwalik correlation (India). American Journal of Scientific Series. 5 (195), 33: 191-204.
- 43. Pilgrim GE (1913) The correlation of the Siwaliks with mammal horizon of Europe. Geological Survey of India Records, 43: 137-160.
- 44. Pascoe EH (1963) A Manual of Geology of India and Burma. (vol. III) Calcutta.