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Evaluation of health hazards from radionuclides in soil and rocks of North Waziristan, Pakistan

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► ***Original article***

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ABSTRACT

Background: The aim & objective of this study is to evaluate the health hazards of the general public from the existence of the radioactive nuclides in soil of North Waziristan and to establish a reference line for future work on radioactivity concentration in this locality. ***Methods and Materials:*** A highly background radiation shielded HPGe system is used to measure radioactivity in the collected samples of the geographical study area. ***Results:*** The radioactivity concentration is found to be in the range 42.6–106.3 Bq/kg for Ra-226, 4.0–93.8 Bq/kg for Th-232, 49.9–645.5 Bq/kg for K-40, and 2.6–8.33 Bq/kg for Cs-137. The external and internal hazards indices (H_{ext} & H_{int}), absorbed γ- dose rate (ADR) and average effective dose (AED) are estimated from the computed activities and determined the average H_{ext} ~ 0.75 and H_{int} ~ 0.94. The mean absorbed dose rate was found to be 101 nGy/h. The Radium equivalent activity of Datta Khel (422.081 Bq/kg), Khaisur forest (407.938 Bq/kg), Jani Khel (379.213) specific regions and the corresponding outdoor annual effective dose rate was found to be 0.15 mSv/yr, which is higher than the world's mean value of 0.07 mSv/yr. ***Conclusion:*** A strong possitive correlation amongst the radioactive nuclides and, corresponding radiological variables at $P \leq 0.01$confirmed that Ra-226 and Th-232 were the significant contributors to γ-radiation emission. Radioactivity concentrations and corresponding radiometric parameters were found to be higher in the specific areas of North Waziristan compared to the world's mean values poses possible radiological risks for the residents of those areas.

Keywords: *HPGe detector, Background Radioactivity, radiological hazards, soil.*

INTRODUCTION

The background level radiation has been in existence since the earth formed. About eighty-seven percent of the overall doses encountered by the majority of humans are from natural sources, which can lead to deleterious health hazards such as cancer. The most commonly encountered radioactive nuclides are Radium, Thorium, Potassium, and Cesium. Radioactive Cesium possibly exists from fallout of nuclear testing and other nuclear activities. To control possible health effects from natural as well as artificial radioactive sources, it is essential to be able to accurately measure the radioactivity concentration. This paper is focused on making such measurements in a particular geographical, area, namely North Waziristan in Northern Pakistan(1–4). Radioactive emission can be from naturally occurring as well as from artificial radioactive substances (5, 6). A number of naturally occurring radioactive nuclides exists in the earth's crust, while others are generated as a result of cosmic interaction with the atmosphere (7). Radiation dose experienced by humankind is mostly from natural resources while man-made radioactivity emanated from nuclear reactors, nuclear weapon testing, industries, and research laboratories, the latter group forming only contributes a relatively small contribution to the total human exposure to radiation dose [2, 8–10]. In normal circumstances, these emissions are very small, but in extreme cases such in the case of a reactor accident, the environment may receive a significant amount of radioactivity e.g. (Chernobyl, Belarus in 1986 and Fukushima, Japan in 2011) [11, 12]. Uranium/Thorium series, K-40, and all of their daughter products are the most commonly encountered natural radioactive nuclides [10, 13–15]. These terrestrial & cosmogenic radioactive nuclides are ingested by humans routinely consumed usually by food stuff, for instance water, through inhalation of airborne particles. In the future, radioactive contamination is considered as one of the most serious threats to the global population and is therefore a major issue. Pollution due to radiation is caused due to the spreading of radioactive materials in soil, rocks, water, and air. Radiological surveys and different studies on soil, rocks, water, and air have enlarged, to study the radiation doses of living organisms [7, 15–19]. Soil is one of the most important naturally occurring environmental materials which is routinely used for many life sustaining purposes, including agriculture, but contains many natural radionuclides, contributing to indoor and outdoor exposure [19]. Therefore, for environmental protection, the measurement of natural radioactivity in rocks and soil is considered very important. To control possible health effects from such natural radioactive sources, the assessment and measurement of these nuclides in soil and rocks of the areas under investigation are important [8, 20].

Among anthropogenic radionuclides, Cs-137 is a significant component of global radioactive fallout. Cs-137 could be considered up to 60 percent of the cumulative effective dose of external radioactivity accompanying previously conducted nuclear tests [21]. Also, as a result of a reactor accident, liberation of fission products may occur and, cesium isotopes are particularly important because of their volatility [22]. It is therefore desirable to accurately measure the Cs-137 level in the environment.

The mountainous region of North Waziristan in Pakistan is a potential geographical ‘hot-spot’ for such radio-active fall-out and therefore it is desirable to perform a study of this region. This is the first time that such a study has been undertaken. This article mainly deals with the quantitative assessment of radioactivity concentration of the $^{238}U/^{232}Th$ series and the primordial K-40 in clay samples obtained from various sites of North Waziristan. Radiological specifications, for instance Radium equivalent dose, rate of absorbed dose, H_{ext} & H_{int} hazards indices and, the corresponding outdoor annual effective dose rate have been calculated from the radioactivity concentration of the $^{238}U/^{232}Th$ series and, K-40. The main aim of this study is to evaluate the potential health hazards to the general public from the existence of these radioactive nuclides in the soil and to establish a reference line for further and future work on radioactivity concentration in this locality.

MATERIALS AND METHODS

Geology of the sampling area

North Waziristan is a former agency of the defunct FATA (Federally Administrated Tribal Area and recently merged in Bannu division of the Khyber Pakhtunkhwa province in Pakistan. It is a mountainous region of northwest Pakistan and is located in the northern part of Waziristan and covers an area of approximately 11,585 km^2 (4,473 mi^2). Its population according to the 2017 census is 543,254 [23]. It is currently subdivided into ten tehsils (Datta Khel, Dossali, Gharyum, Ghulam Khan, Mir Ali, Miran Shah, Razmak, She-wa, Spinwam, and Shawal) [24].

Collection and processing of samples

26 clay samples were collected which is in accordance with IAEA guidelines on the collection of soil samples for analysis [25]. These were obtained from specified areas of North Waziristan during January 2019 and the locations are shown in figure 1.

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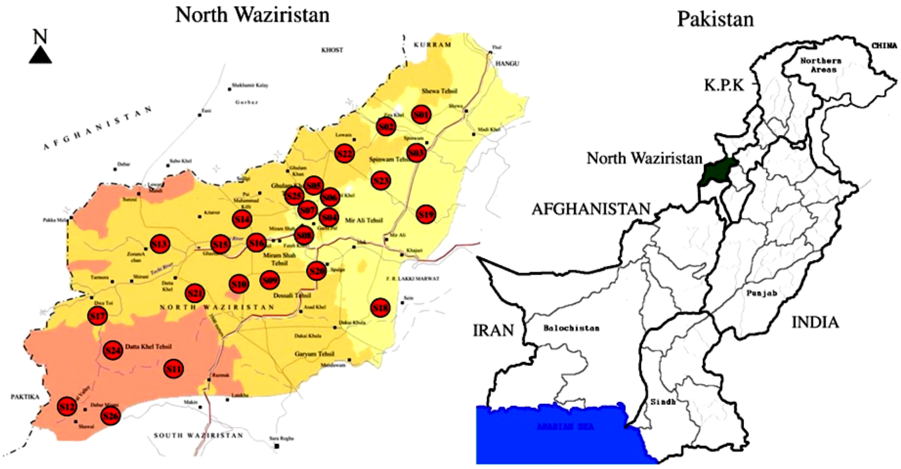

Figure 1. Samples collecting sites of North Waziristan (Detailed coordinates are shown in table 1)

A minimume distance of 1km was maintained between two samples and, polythene bags were used for packing, taped up, marked as S01. S02, S03…………S26 according to the location with together with a designated site code and coordinates of the sample. During sample collection, the coordinates of the site of each sample were recorded utilizing a Garmin eTrex 309x global positioning system (GPS) unit and these are recorded in the 3rd and 4th column of table 1. These samples were categorized and subsequently brought to the laboratory for study. The collected soil samples were initially left to completely dry at room temperature by spreading on a paper sheet for one week. After the drying process was complete, the sample was crushed using a ball mill to reduce the particle size and subsequently further dried in an electric oven at temperature between 40 to 110 ±1°C for 20 hours to completely remove any remnant moisture so that the weight of the sample becomes constant. The oven dried samples were then stored in a desiccator until required for use. The dried samples were further ground to reduce the particle size. The moisture free samples were then homogenized, pulverized and sieved with a mesh size ~ 200µm to remove unwanted materials. The prepared samples were hermetically sealed in Marinelli beakers to avoid any leakage of radon as well as maintaining its geometrical dimension exactly identical to the IAEA provided reference soil-375. The beakers containing the prepared soil samples were placed in an uninterrupted situation for eight weeks to obtain equilibrium amongst ^{238}U and its progenies before counting.

Radiometric analysis

A spectrometry system comprising of a co-axial HPGe detector for the radiometric analyses of the collected soil samples was used [26]. The detector has a relative efficiency of 30% with an active volume 180 mm^3 and an operating voltage of 3000 V. The detector was housed in a lead shielding of well-type (thickness 5–15 cm) for the reduction of background radiation from its surrounding environment. It was continuously cooled using liquid nitrogen in order to minimize the detector thermal noise. The acquisition and analysis of the processed data were performed using a multichannel analyzer having 8196 channels. The system energy resolution at the 1332 keV from ^{60}Co is 2.3 keV FWHM. The system efficiency calibration was performed using reference material provided by IAEA. The spectrum for each 200 g dried soil sample was acquired continuously for about 13 hours to achieve adequate counts at the anticipated peaks. The lower Limits of Detection (LLD) for Ra-226, Th-232, K-40, and Cs-137 were obtained as 2.60, 4.16, 7.01 and 1.24 Bq/kg correspondingly with a ninety-five percent (95%) confidence level. Ra-226 was obtained from its progeny photopeaks of gamma lines ^{214}Pb (295.21 keV, 352 keV) and ^{214}Bi (609 KeV,

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1120.29 keV) and Th-232 was obtained from progeny photopeaks of γ- ^{228}Ac (338.32, 911.21, 968.97 keV) where the presence of gamma-ray peak of energy 1460.83 keV and 661.7 keV in the spectrum indicates K-40 and Cs-137 activity respectively. For the analysis of all of these spectra, the Genie 2000 (Canberra) application was used. Equation1 was used for the calculation of radioactivity accumulation of U - 238, Th-232 and K–40.

Measurement of activity concentration

Equation1 was used to calculate the level of radioactivity of the nuclides (Radium, Thorium, and Potassium) in the prepared samples for analysis and the results are presented in table1.

$$A_s = \frac{N_i}{\eta_i \times P_{\gamma_i} \times C_i \times t} \tag{1}$$

Where

C_i : Net counts

t : Time for data collection

$P_{\gamma i}$: Emission probability

η_i : Detector's efficiency.

Radium equivalent activity (Ra_{eq})

The Soil contained the radioactive nuclides, Ra-226, Th-232, and K-40 but these were not uniformly distributed. Uniformity in respect of exposure to radiation can be written as a Radium equivalent dose (Ra_{eq}), measured in the unit of Bq/kg for the comparison of the specific activity concentration level of constituents including Radium, Thorium, and Potassium as its constituent in varying amounts. It can be calculated using equation 2.

$$Ra_{eq} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_K \tag{2}$$

Where

A_{Ra} = Ra-226 activity concentration

A_{Th} = Th-232activity concentration

A_K = K-40 activity concentration

It is assumed in equation 2 that 10Bq/kg of Ra-226, 7 Bq/kg of Th-232, and 130 Bq/kg of K-40 yields an equivalent dose of gamma-ray radiation. In the above consideration, Ra-226 replaced U-238 as non-equilibrium conditions may exist between U-238 and its daughter nuclei Ra-226 [27, 28]. Radium equivalent activity (Ra_{eq}) for this study was calculated and is represented in the 2nd column of table 2.

Estimation of the radiological hazards

Since multiple radioactive nuclides contribute to the γ- dose, radiological hazards were therefore presented as a single quantity known as the hazards index. External & internal hazards indices were computed using equations 3 and 4 [29–31] for the radiological health risks assessment of this locality.

$$H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{3}$$

$$H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{4}$$

It is recommended that $H_{ext} \leq 1$ to control the external dose of γ-radiation from materials lower than 1.5 mGy/yr.

Absorbed dose estimation

The estimated radioactivity of the radionuclides of interest was changed into absorbed γ- dose expressed in the unit of nGy/h. The absorbed γ-dose rate (D_r) in air at one hundred centimeter over the ground-level is determined using equation5 below.

$$D_r(^{nGy}\!/_h) = (0.043C_K + 0.427C_U + 0.662C_{Th}) \tag{5}$$

C_K = specific activity concentrations of potassium

C_U = specific activity concentrations of uranium

C_{Th} = specific activity concentrations of thorium

The decay products of Ra-226 and Th-232 are assumed to be in radioactive equilibrium with their predecessor.

Rate of effective dose

The annual effective dose rate are measured in unit of mSv/yr and are received by the general public as a result of radioactivity concentrations that exist in soil and construction material and is estimated by using equation 6

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and 7 [29].

$$E_{in} = Q_c \times T \times O_f \times D_r \times 10^{-6} \quad (6)$$

$$E_{out} = Q_c \times T \times O_f \times D_r \times 10^{-6} \quad (7)$$

Where Q_c is the conversion coefficient to the effective dose (0.7 Sv G/yr) for the absorbed dose in the air, T is time in hours for one year duration (365×24 h), O_f is an occupancy factor being 0.8 & 0.2 for indoors and outdoors respectively as proposed by UNSCEAR-2000 and D_r is the dose rate in the unit of nG/yr given by equation 5. This calculation assumes that the general public spends most of their time, about 80 percent (80%) outdoors and 20 percent (20%) indoors. Where E_{in} is the indoor annual effective dose rate (IAEDR) and E_{out} is the outdoor annual effective dose rate(OAEDR).

Statistical analysis

IBM SPSS version-25 and MS-Excel 2013 software packages for the statistical analysis were used in order to find the correlation that exists amongst the measured radioactivity and the corresponding radiological variables.

Table 1. Measured activity concentrations in the collected samples from North Waziristan.

Codes	Sample location	Latitude	Longitude	K-40 (Bq/kg)	Ra-226 (Bq/kg)	Th-232 (Bq/kg)	Cs-137 (Bq/kg)
S01	Shewa	33° 15' 20.34'' N	70° 29' 51.972'' E	416.7	73.3	150.4	Not detected
S02	Spin Wam	33° 10' 45.228'' N	70° 23' 38.328'' E	485.3	79.9	170.7	Not detected
S03	Datta Khel	33° 8' 58.272'' N	70° 26' 0.384'' E	553.9	106.3	191.0	Not detected
S04	Mir Ali	32° 58' 10.992'' N	70° 16' 30.144'' E	416.7	67.3	84.8	Not detected
S05	Idak	32° 58' 45.084'' N	70° 12' 5.616'' E	485.3	51.4	161.5	Not detected
S06	Hurmaz	32° 58' 16.32'' N	70° 15' 41.112'' E	553.9	55.2	82.1	Not detected
S07	Miran Sha	32° 57' 17.46'' N	70° 10' 9.696'' E	576.7	77.1	70.1	Not detected
S08	Spalga	32° 55' 13.656'' N	70° 8' 22.992'' E	348.1	54.1	149.5	Not detected
S09	Musaki	32° 49' 23.844'' N	70° 3' 52.308'' E	645.5	65.1	69.1	Not detected
S10	Dosali	32° 48' 44.64'' N	69° 57' 57.492'' E	531.0	81.0	74.7	Not detected
S11	Cadet College Razmak	32° 40' 58.404'' N	69° 49' 54.408'' E	462.4	53.6	156.8	Not detected
S12	Shawal	32° 36' 49.428'' N	69° 30' 36.144'' E	462.4	42.6	28.0	Not detected
S13	Datta Khel Post	32° 54' 23.58'' N	69° 45' 54.756'' E	462.4	90.3	155.0	Not detected
S14	Hamzoni	32° 58' 22.908'' N	69° 59' 13.632'' E	508.1	83.7	4.0	Not detected
S15	Boyya	32° 56' 35.376'' N	69° 54' 6.588'' E	393.9	47.0	149.5	Not detected
S16	Sample16	32° 55' 19.416'' N	70° 1' 16.716'' E	393.9	52.5	75.6	Not detected
S17	Chaprai	32° 48' 4.32'' N	69° 34' 21.216'' E	393.9	57.4	152.2	Not detected
S18	Jani khel	32° 47' 21.984'' N	70° 30' 44.28'' E	622.4	85.9	171.6	Not detected
S19	Baka Khel	32° 56' 58.128'' N	70° 31' 48.612'' E	439.6	56.3	122.4	Not detected
S20	Khaisur Forest	32° 52' 5.52'' N	70° 14' 10.176'' E	348.1	104.0	193.8	4.13
S21	Razmak Road	32° 48' 1.44'' N	69° 49' 34.716'' E	553.9	61.3	164.2	Not detected
S22	Dandy	33° 6' 26.964'' N	70° 15' 41.904'' E	508.1	79.9	147.7	Not detected
S23	Mandi Khel Dam	33° 1' 55.92'' N	70° 22' 22.296'' E	49.9	72.8	139.3	Not detected
S24	Shawal Road	32° 43' 15.888'' N	69° 38' 46.032'' E	348.1	53.6	48.2	Not detected
S25	Bannu Miran-sha road	33° 0' 11.196'' N	70° 7' 51.204'' E	508.1	81.0	151.3	3.8
S26	Shawal	32° 33' 53.892'' N	69° 40' 38.28'' E	325.5	73.9	152.2	Not detected
Minimum				49.9	42.6	4.00	
Maximum				645.5	106.3	193.8	
Mean±SE				453.60±23.34	69.5±3.33	123.68±10.17	
World's average*				420	33	45	

Where, Standard Error (SE) = $\sigma/\sqrt{N}$ is the standard deviation and N is the number of observation. *UNSCEAR-2000

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Table 2. Radiological parameters linked with the measured radioactivity concentration in the collected sample from North Waziristan.

Sample location	Ra_{eq} activity (Bq/kg)	Absorbed dose rate $D_r(^{nGy}/_h)$	External hazard index H_{ext}	Internal Hazard index H_{int}	Annual effective dose (mSv/yr) Indoor	Outdoor
Shewa	320.458	112.002	0.865435	1.063543	0.54943	0.187 68
Spin Wam	361.369	126.113	0.975913	1.191859	0.61866	0.211 91
Datta Khel	422.08	149.372	1.13990	1.427202	0.73271	0.243 75
Mir Ali	220.65	82.728	0.595937	0.777828	0.40583	0.129 67
Idak	319.713	109.229	0.863365	1.002283	0.53583	0.188 87
Hurmaz	215.253	81.768	0.581333	0.730522	0.4011	0.128 34
Miran Sha	221.749	87.988	0.598930	0.807309	0.43189	0.131 35
Spalga	294.689	99.907	0.795806	0.942022	0.49018	0.172 87
Musaki	213.617	84.909	0.576940	0.752886	0.41653	0.128 43
Dosali	228.708	89.743	0.617730	0.836649	0.44024	0.134 81
C C Razmak	313.429	107.392	0.846403	0.991268	0.52682	0.184 89
Shawal	118.245	50.275	0.319376	0.434511	0.24664	0.060 82
Datta Khel Post	347.555	123.620	0.938642	1.182696	0.60643	0.203 16
Hamzoni	128.544	61.473	0.347294	0.573510	0.30156	0.075 98
Boyya	291.115	98.537	0.786138	0.913166	0.48338	0.171 53
Sample16	190.938	71.223	0.515675	0.657567	0.34939	0.112 78
Chaprai	305.376	104.433	0.824671	0.97980	0.51230	0.179 38
Jani khel	379.213	134.966	1.024107	1.256266	0.66209	0.223 33
Baka Khel	265.181	93.791	0.716141	0.868328	0.46010	0.156 38
Khaisur Forest	407.938	140.858	1.101713	1.382773	0.69099	0.236 74
Razmak Road	338.756	117.755	0.914808	1.08411	0.57765	0.200 18
Dandy	330.235	117.772	0.891850	1.196258	0.57774	0.193 94
Mandi Khel Dam	275.841	91.991	0.744968	0.925572	0.45127	0.155 54
Shawal Road	149.33	58.751	0.403335	0.548200	0.28821	0.088 29
Banu Miransha road	336.483	119.734	0.908722	1.127648	0.58737	0.197 54
Shawal	316.61	109.203	0.855046	1.054765	0.53571	0.184 55
Minimum	118.24	50.271	0.319376	0.434511	0.24664	0.060 82
Maximum	422.08	149.371	1.139981	1.427202	0.73271	0.243 75
Mean ± SE	281.272±15.97	100.982±4.90	0.759±0.04	0.947±0.05	0.495±0.02	0.150 ±0.01
World's average*	370	59	1≤	1≤	0.42	0.07

Where, Standard Error (SE) = $\sigma/\sqrt{N}$ is the standard deviation and N is the number of observation. *UNSCEAR-2000

RESULTS

The radioactivity concentration of Ra-226, Th-232, and K-40 in the soil samples collected from 26 various parts of North Waziristan were calculated using equation 1. Table 1 summarizes the computed values for activity concentration that ranges from 42.6 (Shawal) - 106.3 (Datta Khel) Bq/kg for Radium, 4.00 (Hamzoni) - 193.80 (Khaisur Forest) Bq/kg for Thorium, and 49.90 (Mandi Khel Dam) - 645.50 (Musaki) Bq/kg for Potassium respectively. The mean activity concentrations of Ra-226, Th-232 and, K-40 of the 26 collected samples from various locations in North Waziristan were found to be 69.5, 123.68 and 453.60 Bq/kg respectively. The concentration of the radioactivity showing that the mean value of Potassium is higher than Radium and Thorium in the collected samples. Equation2 was used to calculate Ra_{eq} as one index in order to illustrate the γ-output from the varying amount of Radium, Thorium and Potassium were present in the collected samples and their specific activities are included in the

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2nd column of table 2. It shows the minimum level of radium equivalent activity for Shawal (113.91 Bq/kg) and maximum for Datta Khel (418.10 Bq/kg) with mean value being 281.272 Bq/kg. The 3rd column of table 2 includes the γ-absorbed dose rate due to radium, thorium and, potassium 100 centimeters above ground level in the samples as calculated using equation5. The absorbed γ-dose rate of the mentioned radionuclides at each of the 26 different sampling sites was calculated [32–34]. The estimated absorbed γ- dose rate lies in the range of 50.271(Shawal) to 149.371 nGy/h (Datta Khel) (with a mean value of 101 nGy/h) which is almost twice of the world's average value 59 nGy/h [35], indicating that the measured absorbed dose rate due to natural radioactive nuclides in air for the studied area is1.7 times the internationally recommended value. The corresponding indoor and outdoor effective dose rates estimated by equation 6&7 shown in the last column of table 2 varies in the range 0.24 - 0.73 mSv/yr (with a mean value of 0.49 mSv/yr) and 0.06 - 0.25 mSv/yr (Mean value 0.15 mSv/yr). The mean outdoor effective dose rate is also higher than the world's mean value of 0.07 mSv/yr [31]. The highest outdoor annual effective dose of 0.25 was observed for Datta Khel. External & internal hazards indices were computed using equations 3 & 4 (31-33) to evaluate the radiological health hazards linked with the clay and soil of this area. These varying values of internal and external hazards indices are presented in the 4th & 5th column of table 2. It is recommended that H_{ext} ≤1 and H_{int} ≤ 1 but some of area studied in this investigation show both the hazard indices to be greater than unity. It can be seen from table 2 that the external hazard index has a minimum value 0.31, maximum value 1.13 and, mean value of 0.75. Similarly, the internal hazard index (H_{int}) for the collected samples from the area under investigation the minimum value was 0.42, the maximum value 1.42 and the average value 0.94. As shown in table 1, Cs-137 has been detected in Khaisur forest and Bannu Miran Sha Road area. In order to confirm the activity level of Cs-137 in these areas, a further 6 samples of soil were obtained from different lactions and, the measured values of Cs-137 for these sites are shown in table 3. Some studied areas showed Cs -137 activity below the limit of detection (0.2 Bq/kg), while other regions' activity lies in the range 2.6 to 8.33 Bq/kg, which is probably due to fallout from worldwide reactor accidents and nuclear weapon testing in the past. The correlation amongst the calculated radioactivity and the corresponding radiological variables were analyzed using multivariate Pearson's correlation and shown in table 4.

Table 3. The calculated activity of Cs-137 (Bq/kg) in clay samples of North Waziristan.

Sample Location	Sample Code	Cs-137
Khaisur Forest	SKF01	4.13
Khaisur Forest	SKF02	6.5
Khaisur Forest	SKF03	2.7
Khaisur Forest	SKF04	4.8
Khaisur Forest	SKF05	2.9
Khaisur Forest	SKF06	2.6
Khaisur Forest	SKF07	Not detected
Khaisur Forest	SKF08	Not detected
Bannu Miran Sha Road	BMR01	3.8
Bannu Miran ShaRoad	BMR02	8.33
Bannu Miran Sha Road	BMR03	6.83
Bannu Miran Sha Road	BMR04	Not detected
Bannu Miran Sha Road	BMR05	Not detected
Bannu Miran Sha Road	BMR06	Not detected
Bannu Miran Sha Road	BMR07	Not detected
Bannu Miran Sha Road	BMR08	Not detected

Table 4. Multivariate (Pearson's correlation) matrix of measured radionuclides and corresponding radiological variables.

	Ra-226	Th-232	K-40	Ra_{eq}	D_r	H_{ext}	H_{int}	E_{out}	E_{in}
Ra-226	1								
Th-232	0.35	1							
K-40	0.15	0.12	1						
Ra_{eq}	0.54	0.97	0.03	1					
D_r	0.64	0.92	0.14	0.99	1				
H_{ext}	0.54	0.97	0.03	1	0.99	1			
H_{int}	0.67	0.92	0.05	0.99	0.99	0.99	1		
E_{out}	0.64	0.92	0.14	0.99	1	0.99	0.99	1	
E_{in}	0.64	0.92	0.14	0.99	1	0.99	0.99	1	1

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DISCUSSION

According to UNSCEAR-2000, the internationally reported medians values of radioactivity concentrations of radionuclides Ra-226 is 35 Bq/kg, Th - 232 is 30 Bq/kg, and k - 40 is 400 Bq/kg [31]. A comparison of the mean radioactivity concentration of the aforementioned three radionuclides in the present work was made with the world's average values as well as some other countries of the world and the results are shown in figure 2.

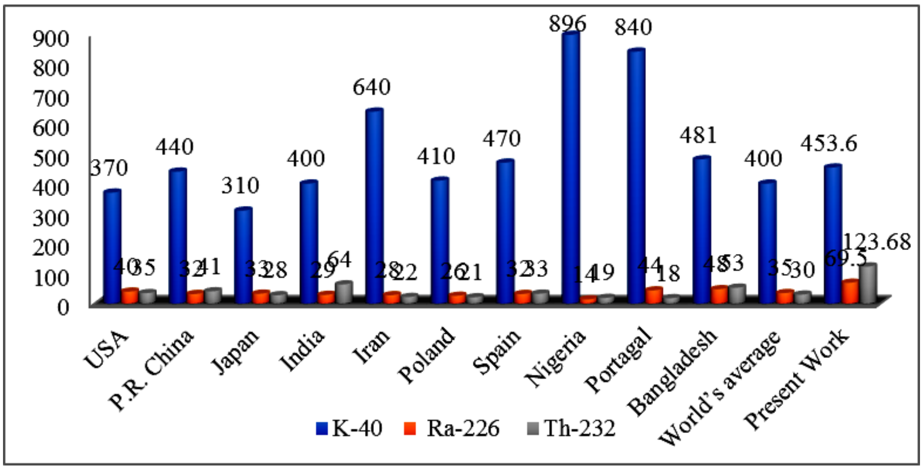

Figure 2. Comparison of mean activity concentration of present work with world's average and other countries.

It is evident from figure 2 that the average radioactivity concentrations of Ra-226, and Th-232 in the collected samples were found to be higher than the world's average values by a factor of 2.10 for radium, and 2.75 for Thorium. Results also revealed that the radioactivity concentration of radium and thorium in this study were higher than other published data in Pakistan [36–41] as well as some other countries of the world such as USA, P. R. China, Japan, India, Iran, Poland, Spain, Thailand, Bangladesh, Nigeria and, Portugal [31, 42, 43]. These higher values of Ra-226 and Th-232 measured in the present study may be accredited to the geography and geology of this area.

However, the average value for K-40 of this study lies in the world's median range of (140-850 Bq/kg) [31] but higher than the reported values in literature for some countries including USA, China, Japan, India, and Poland [31].

The concentrations of activity of Radium, Thorium, and Potassium in the Earth's crust fluctuate with the half-lives of the nuclides (i.e. the longer the half-life, the higher the activity concentration of that particular radionuclide). The radium equivalent (Ra_{eq}) values of all sites were found to be generally less than the accepted world's value 370 Bq/kg [31], while exceeded the values for Murree, District Chakwal, Southern Punjab, Rawalpindi, Mardan, Noshera and Peshawar [37–40, 44]. However some sites such as Datta Khel (422.08 Bq/kg), Jani Khel (379.938 Bq/kg) and Khaisur Forest (407.938) in the present study showed higher values of Ra_{eq} than world's average. This indicates that the soil of these three locations may pose health risks if used as construction materials.

The mean absorbed dose rate (D_r) in this study is almost twice of the world's average value 59 nGy/h, Which is also higher than some countries of the world [31] as well as other parts of Pakistan such as Sindh, Baluchistan, Peshawar, Punjab etc. [36–39, 45].

The Internal hazards index (H_{int}) is helpful for controlling the internal exposure from ^{222}Rn and its radioactive progenies, which badly effect respiratory organs [46]. H_{int} > 1 was observed in

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samples S1, S2, S3, S5, S13, S18, S20, S21, S22, S225 and S26, with an average value less than 1 being recommended by UNSCEAR-2000. Similarly external hazard index (H_{int}) in samples S3, S18 and S20 was found greater than 1 with the mean value being less than world's permissible limit of 1[31]. It is therefore advised that the use of soil of those areas with H_{int} & H_{ext} greater than one is avoided for construction of houses to live in.

A strong positive correlation amongst the radioactive nuclides and the corresponding radiological variables at $P \leq 0.01$ confirmed that Ra-226 and Th-232 were the significant contributors to the γ- radiation emission all over the study area.

CONCLUSION

The strong positive correlation amongst the radioactive nuclides and the corresponding radiological variables at $P \leq 0.01$ confirmed that Ra-226 and Th-232 were the significant contributors to the γ- emission over the entire study area.

The average values of the radioactivity concentrations in the investigated samples were found to be generally higher than the world's mean concentration as well as some other parts of Pakistan.

Samples from Datta Khel, Khaisur forest and Jani Khel were found with Ra_{eq} higher than the accepted permissible limit (370 Bq/kg), which corresponds to H_{ext} & H_{int} > 1, and therefore soil from these areas must be avoided for use as a building material.

The estimated E_{out} to the resident of this area was found to be 0.15 mSv/yr which is also higher than the world's outdoor annual effective dose of 0.07 mSv/yr. The higher values of radiometric variables for the specific studied areas confirmed the existence of high radiological risks for the residents of those areas. So the radioactivity concentration of these areas needs to be monitored regularly to avoid any possible health consequences.

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REFERENCES

1. Bangotra P, Mehra R, Jakhu R, Kaur K, Pandit P, Kanse S (2018) Estimation of 222 Rn exhalation rate and assessment of radiological risk from activity concentration of 226 Ra, 232 Th and 40 K. *Journal of Geochemical Exploration*, ***184**: 304–310*.
2. El-Bahi SM, Sroor A, Mohamed GY, El-Gendy NS (2017) Radiological impact of natural radioactivity in Egyptian phosphate rocks, phosphogypsum and phosphate fertilizers. *Applied Radiation and Isotopes*, ***123***: *121–127*.
3. Kant K, Gupta R, Kumari R, Gupta N, Garg M (2015) Natural radioactivity in Indian vegetation samples. *Int J Radiat Res*, ***13(2)**: 143–150*.
4. Usikalu MR, Rabiu AB, Oyeyemi KD, Achuka JA, Maaza M (2017) Radiation hazard in soil from Ajaokuta North-central Nigeria. *Int J Radiat Res*, ***15*(2)***: 219–224*. doi:10.18869/acadpub.ijrr.15.2.219
5. Amanjeet Kumar A, Kumar S, Singh J, Singh P, Bajwa BS (2017) Assessment of natural radioactivity levels and associated dose rates in soil samples from historical city Panipat, India. *Journal of Radiation Research and Applied Sciences*, ***10*(3)***: 283–288*.
6. Selçuk Zorer Ö (2019) Evaluations of environmental hazard parameters of natural and some artificial radionuclides in river water and sediments. *Microchemical Journal*, ***145**: 762-766*.
7. Jibiri NN and Biere PE (2011) Activity concentrations of 232 Th , 226 Ra and 40 K and gamma radiation absorbed dose rate levels in farm soil for the production of different brands of cigarette tobacco smoked in Nigeria. *Iranian Journal of Radiation Research*, ***8*(4)***: 201–206*.
8. Alharbi WR (2013) Natural radioactivity and dose assessment for brands of chemical and organic fertilizers used in Saudi Arabia. *Journal of Modern Physics*, **04(03)**: 344–348.

[DOI: 10.18869/acadpub.ijrr.18.2.243]

doi:10.4236/jmp.2013.43047.
9. Dizman S, Görür FK, Keser R (2016) Determination of radioactivity levels of soil samples and the excess of lifetime cancer risk in Rize province, Turkey. *Int J Radiat Res*, ***14*(3)***: 237–244*.
10. El-Taher A and Al-Zahrani JH (2014) Radioactivity measurements and radiation dose assessments in soil of Al-Qassim region, Saudi Arabia. *Indian Journal of Pure and Applied Physics*, ***52*(3)***: 147–154*.
11. Omar Nazir L, Shi X, Moller A, Mousseau T, Byun S, Hancock S, Mothersill C (2018) Long-term effects of ionizing radiation after the Chernobyl accident: Possible contribution of historic dose. *Environmental Research*, ***165****: 55–62*.
12. Yoschenko V, Ohkubo T, Kashparov V (2018) Radioactive contaminated forests in Fukushima and Chernobyl. *Journal of Forest Research*, ***23(1)****: 3–14*.
13. Asghar M, Tufail M, Sabiha J, Abid A, Waqas M (2008) Radiological implications of granite of northern Pakistan. *Journal of Radiological Protection*, ***28*(3)***: 387–399*.
14. Jabbar A, Arshed W, Bhatti AS, Ahmad SS, Akhter P, Rehman SU, Anjum MI (2010) Measurement of soil radioactivity levels and radiation hazard assessment in southern Rechna interfluvial region, Pakistan. *Environmental Monitoring and Assessment*, ***169*(1–4)***: 429–438*.
15. Rafique M, Rehman H, Matiullah MF, Rajput MU, Rahman SU, Rathore MH (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*.
16. Isinkaye M, Jibiri N, Bamidele S, Najma L (2018) Evaluation of radiological hazards due to natural radioactivity in bituminous soils from tar-sand belt of southwest Nigeria using HpGe-Detector. *Int J Radiat Res*, ***16(3)****: 351–362*.
17. Korkmaz ME, Agar O, Uzun E (2017) Assessment of natural radioactivity levels for Karadağ Mountain, Turkey. *Int J Radiat Res, **15(4)**: 399-406.*
18. Manigandan PK (2009) Activity concentration of radionuclides in plants in the environment of Western Ghats. *Iran J Radiat Res*, ***7(2)****: 85–90*.
19. Usikalu MR, Maleka PP, Malik M, Oyeyemi KD, Adewoyin OO (2015) Assessment of geogenic natural radionuclide contents of soil samples collected from Ogun state, south western, Nigeria. *Int J Radiat Res*, ***13*(4)***: 355–361*.
20. El Samad O, Baydoun R, Nsouli B, Darwish T (2013) Determination of natural and artificial radioactivity in soil at North Lebanon province. *Journal of Environmental Radioactivity*, ***125****: 36–39*.
21. Karunakara N, Somashekarappa HM, Narayana Y, Avadhani DN (2001) 137Cs concentration in the environment of Kaiga of south west coast of India. *Health Physics Society*, ***81*(2)***: 148–155*.
22. Miller KM, Kuiper JL, Helfer IK (1990) 137Cs fallout depth distributions in forest versus field sites: Implications for external gamma dose rates. *Journal of Environmental Radioactivity*, **12(1)**: 23–47.
23. Wazir MA, and Goujon A (2019) Assessing the 2017 Census of Pakistan Using Demographic Analysis: A Sub-National Perspective. *Vienna Institute of Demography Austrian Academy of Science*, (April), 0–43.
24. Spain JW (1954) Pakistan's North West Frontier Middle East Journal. (n.d.).
25. A Guidebook (1989) *Measurement of Radionuclides in Food and the Environment*. Vienna: International Atomic Energy Agency. Retrieved from https://www.iaea.org/publications/1398/measurement-of-radionuclides-in-food-and-the-environment.
26. Ramasamy V, Dheenathayalu M, Ravisankar R, Ponnusamy V (2004) Natural radioactivity measurements in beachrock samples of south-east coast of Tamilnadu, India. *Radiation Protection Dosimetry*, ***111*(2)**: 229–235.
27. Faheem M, Mujahid SA, Matiullah U (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(8)****: 1443–1447*.
28. Fathivand AA, Amidi J, Najafi A (2007) The natural radioactivity in the bricks used for the construction of the dwelling in Tehran areas of Iran. *Radiation Protection Dosimetry, **123(3)**: 391–393*.
29. Farai IP and Ademola JA (2005) Radium equivalent activity concentrations in concrete building blocks in eight cities in Southwestern Nigeria. *Journal of Environmental Radioactivity*, ***79*(2)***: 119–125*.
30. Hamid BN, Alam MN, Chowdhury MI, Islam MN (2002) Study of natural radionuclide concentrations in an area of elevated radiation background in the Northern districts of Bangladesh. *Radiation Protection Dosimetry*, ***98(2)****: 227–230*.
31. UNSCEAR (2000) Effects of Ionizing radiation, United Nations scientific committee on the effects of Atomic radiation. *Exposures from Natural Radiation Sources, Annex B*. United Nations, New York.
32. El-Arabi AM (2007) 226Ra, 232Th and 40K concentrations in igneous rocks from eastern desert, Egypt and its radiological implications. *Radiation Measurements*, ***42(1)****: 94–100*. doi:10.1016/j.radmeas.2006.06.008
33. Merdanoğlu B and Altınsoy N (2006) Radioactivity concentrations and dose assessment for soil samples from Kestanbol granite area, Turkey. *Radiation Protection Dosimetry*, ***121(4)****: 399–405*.
34. Rahman S, Matiullah U, Mujahid SA, Hussain S (2007) Assessment of the radiological hazards due to naturally occurring radionuclides in soil samples collected from the north western areas of Pakistan. *Radiation Protection Dosimetry*, ***128*(2)***: 191–197*.
35. Mouandza SYL, Moubissi AB, Abiama PE, Ekogo TB (2018) Study of natural radioactivity to Assess of radiation hazards from soil samples collected from Mounana in south-east of Gabon. *Int J Radiat Rese*, ***16(4)****: 443–453*.
36. Mujahid SA and Hussain S (2010) Natural radioactivity in soil in the Baluchistan province of Pakistan. *Radiation Protection Dosimetry*, ***140(4)****: 333–339*.
37. Rahman SU, Matiullah U, Malik F, Rafique M, Anwa J, Ziafat M, Jabbar A (2011) 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 Radioan-*

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alytical and Nuclear Chemistry, ***287*(2)***: 647–655*.

38. Khizar Hayat S, Jabbar T, Dilband M, Khalid K, Rashid A, Jabbar A (2017) Assessment of background radiation levels and associated doses in soils of the most popular tourist place Muree, Pakistan. *Universal Journal of Engineering Science*, *5*(4)*: 64–69*.
39. Tufail M, Asghar M, Akram M, Javied S, Khan K, Mujahid SA (2013) Measurement of natural radioactivity in soil from Peshawar basin of Pakistan. *Journal of Radioanalytical and Nuclear Chemistry*, ***298(2)****: 1085–1096*.
40. Fatima I, Zaidi JH, Arif M, Daud M, Ahmad SA, Tahir SNA (2007) Measurement of natural radioactivity and dose rate assessment of terrestrial gamma radiation in the soil of southern Punjab, Pakistan. *Radiation Protection Dosimetry*, *128*(2)*: 206–212*.
41. Mujahid SA and Hussain S (2011) Measurement of natural radioactivity from soil samples of Sind, Pakistan. *Radiation Protection Dosimetry*, ***145(4)****: 351–355*.
42. Kabir K, Islam SM, Rahman M (2009) Distribution of radionuclides in surface soil and bottom sediment in the district of Jessore, Bangladesh and evaluation of radiation hazard. *Journal of Bangladesh Academy of Sciences*, ***33*(1)***: 117–130*.
43. Okeyode I and Oluseye A (2010) Studies of the terrestrial outdoor gamma dose rate levels in Ogun-Osun river basins development authority headquarters, Abeokuta, Nigeria. *Physics International*, ***1*(1)***: 1–8*.
44. Mehdi SA, Rahman SU, Khan K, Jabbar A, Rafique M (2016). Assessment of annual effective dose from measured soil radioactivity levels using HPGe detector. *Universal Journal of Engineering Science*, *4*(4)*: 79–83*.
45. Jabbar A, Khan K, Jabbar T, Rafique M, Rehman SU, Arshed W, Dilband M (2016) Radioactive contents and background doses from northern alluvial sediment plains between rivers Ravi and Chenab, Pakistan. *Nuclear Science and Techniques*, ***27*(4)***: 94*.
46. Al-Trabulsy HA, Khater AEM, Habbani FI (2011) Radioactivity levels and radiological hazard indices at the Saudi coastline of the Gulf of Aqaba. *Radiation Physics and Chemistry*, **80(3)**: 343–348.