Iran. J. Radiat. Res., 2011; 8(4): 237-242

Measuring γ–ray dose of terrestrial samples using β-γ Spectrometry

S. Ashrafi and Sh. Alaei*

Department of Nuclear Physics, Faculty of Physics, University of Tabriz, Iran

Background: The existence of some radionuclides in soil and some building materials produce a β-γ radiation field, which in some regions the exposure of these radionuclides to human is high. ***Materials and Methods:*** The air-absorbed dose, indoor and outdoor annual effective dose of soil and some building material samples (ceramic, granite, gypsum, etc.). The samples were collected from 35 different regions in the northwest of Iran were calculated. Specific activity of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K were measured by using a β-γ spectrometer consisted of NaI(Tl) and organic scintillators. The analysis of measured spectra was based on maximum likelihood estimation. The calculated data were compared with world's mean values. ***Results:*** It is found that the specific activity of ^{40}K ranges from 573.8 to 1392 Bqkg^{-1}, for ^{232}Th ranges from 6 to 54.6 Bqkg^{-1} and for ^{226}Ra ranges from and 5.1 to 36.2 Bqkg^{-1}. In some regions, the concentrations of natural radionuclides (40K, 232Th, 226Ra) were higher than standard level. ***Conclusion:*** Experimental results related to specific activity, indoor and outdoor annual effective doses of ^{226}Ra, ^{232}Th and ^{40}K revealed that radionuclide concentration in soil and some building material samples of some regions of northwest of Iran are of some radiological importance. **Iran. J. Radiat. Res., 2011; 8 (4): 237-242**

Keywords: *Environmental radioactivity, β-γ spectrometry, annual effective dose rate.*

INTRODUCTION

Many radionuclides exist naturally in terrestrial soils, rocks and in building materials developed from them. In term of dose, the most important primordial (half-life comparable to the age of the earth) radionuclides are ^{40}K, ^{232}Th and ^{238}U. Their absolute and relative concentrations in soil and construction materials can vary dramatically depending on their source [1]. The decay of these radionuclides in soil produces a β-γ radiation field that crosses the soil-air interfaces and produces exposure to humans. Many surveys in order to estimate their concentration and assessment of the associated dose rate in air have been made in most inhabited regions of the world [2-11] using gamma spectrometry.

The products of uranium and thorium chains are always present in soil and rock. Their gamma radiation causes external exposure with the consequent absorbed doses. The avialable geological data suggests that soils and rocks from the zone of Zonoz County, Caulan mines and Mishu Mountain in the northwest of Iran may have high level of naturally occuring radioactivity. The purpose of our research has been to clarify the amount of natural radiation exposure of these regions' population. Gamma spectrometry combined with beta spectrometry, and analyzing the measured spectra applying an algorithm based on maximum likelihood estimation allowed the identification of the potential source of contamination in order to estimate the possible radiological hazards to human health. Considerable attention should be paid to decrease exposure level arising from members of uranium and thorium decay chains and by potassium-40 soils.

MATERIALS AND METHODS

Sample collection and preparation

Samples of soils, rocks and building materials were collected from 35 different regions of Eastern Azerbaijan, Iran. The studied areas, which lie at 38°30′N latitude and 45°45′E longitudes, in figure 1 are shown. Surface soil samples (0–5 cm deep)

Corresponding author:
Mrs. Sholeh Alaei,
Department of Nuclear Physics, Faculty of Physics, University of Tabriz, Iran.
Fax: *+98 441 2356742*
E-mail: *sholeh.alaei@gmail.com*

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were carefully collected from areas, which were not covered by any type of vegetation and thought to be undisturbed. We grounded thoroughly homogenized and changed the rock samples into the powder form before the measurements. All soil samples and building materials converted into very small particles and screened with a sieve of about 1mm mesh. The prepared and homogenized samples were weighted and put into Marinelli beakers with the volume of 1 liter and flat vessels 0.1, 0.2 and 0.5 liter volume. Then they were packed into radon-impermeable plastic containers. Each beaker was kept for 4 weeks (>seven half-lives of ^{222}Rn and ^{224}Ra) prior to counting in order to ensure that the daughter products of ^{226}Ra up to ^{210}Pb and of ^{228}Th up to ^{208}Pb achieve equilibrium with their respective parent radionuclides [12].

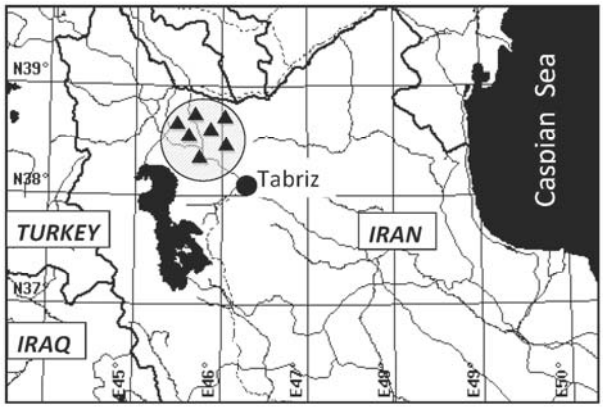

Figure 1. Simplified geology of Northwestern Iran. Locations of soil sampling is shown by solid triangles.

Experimental setup

The β-γ spectrometer which was used [13] consisted of two cylindrical scintillators: a Φ128 × 8 mm paraterphenyl-activated polystyrene plastic for beta detection and a Φ63 × 63 mm NaI(Tl) for gamma detection. The scintillators were made by ATOMTEX, Belarus. The NaI(Tl) detector had relative efficiencies of < 8.5% for the 662 keV γ-ray line of ^{137}Cs. To derive maximum detection efficiency, the beta and gamma detectors were both surrounded by 5 cm thick low-activity lead shield. The source-detector geometry has been shown in figure 2. In case of voluminous environmental samples, the best limit of detection per mass or volume unit was achieved when the sample were filled in Marinelli beaker. The minimum detectable activity (MDA) for the beta-gamma spectrometer system used in the present study was ~2 Bq/kg for ^{137}Cs for a counting time of 3 hours and for one liter Marinelli beaker sample [14].

The detectors were connected to personal computer (which has built in detector HV bias supply, ADC and MCA) for simultaneously beta and gamma ray energy spectrum acquisition. The measured spectra were analyzed by SPTR–ATM program based on maximum likelihood algorithm for activity estimation. One of the measured energy spectrum and assigned gamma line is shown in figure 3. The detector efficiency calibration was performed by using standard point and volume sources.

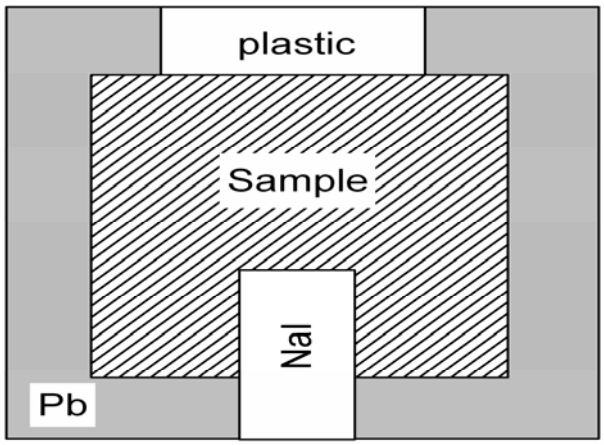

Figure 2. Source-detector geometry. β and γ radiations are emitted from voluminous sample and are detected by plastic and NaI scintillators, respectively.

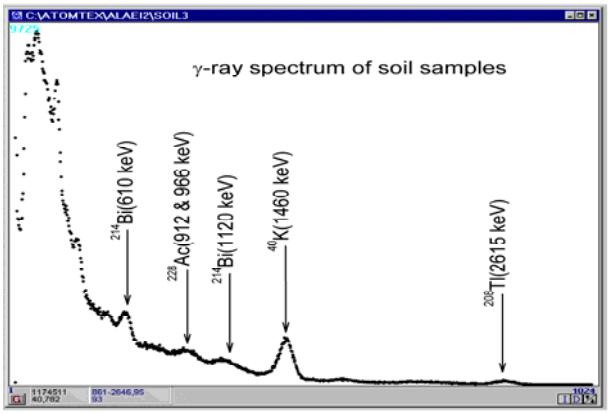

Figure 3. A γ-ray spectrum of a soil sample measured by using NaI(Tl) scintillator.

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Maximum likelihood activity estimation

For those radionuclides with half life longer than the time period of measurement β-particles or γ-rays came from a very large number of independent nuclei within the sample, Poisson statistical model was appropriate. For this reason, in the gamma and beta spectrometry of the environmental samples, the Poisson distribution is usually used to analyze the measured spectra. The maximum likelihood (ML) method of activity estimation was derived from the condition of the maximum of the logarithm of likelihood function [15]. The logarithm of likelihood function for the Poisson distribution of counts in the beta or gamma spectrum channels is given as:

$$L^* = \ln L = \sum_{i=1}^{N}\left[-t\sum_{k=1}^{M} A_k f_k(i) + S(i)\ln\left(t\sum_{k=1}^{M} A_k f_k(i)\right) - \ln(S(i)!)\right] \quad (1)$$

Where S(i) indicates the beta or gamma experimental spectrum of a mixture of radioactive nuclides, I, is the channel number; N, is the number of channels (=1024); t, is the spectrum accumulation time, M is the number of spectral components; f_k (i) for k =1,2, …, M -1, is the spectrum of the kth nuclide normalized to unit activity and unit time; f_M (i), is the spectrum of a background normalized to unit time; A_k is the unknown activity of the kth nuclide.

The estimates of A_k are determined by solving the following system of the M equations deriving from the condition of the maximum of equation 1:

$$\frac{\partial L^*}{\partial A_k} = 0$$

$$\sum_{i=1}^{N}\left[\frac{s(i)\, f_j(i)}{\sum_{k=1}^{M} A_k f_k(i)} - t f_j(i)\right] = 0, \quad j = 1, 2, \ldots, M \quad (2)$$

The system of equation 2 is solved by iteration method. In every q^{th} iteration (q=1, 2, …) one solves the set of the linear equations:

$$\sum_{k=1}^{M} t\, A_k \sum_{i=1}^{N} \frac{f_k(i)\, f_j(i)}{D_q(i)} = \sum_{i=1}^{N} \frac{S(i)\, f_j(i)}{D_q(i)} \quad (3)$$

$$D_q(i) = \begin{cases} \sum_{k=1}^{M} A_k^{(q-1)} f_k(i) & if \quad q > 0 \\ S(i)+1 & if \quad q = 0 \end{cases} \quad (4)$$

Where:
$A_k^{(q-1)}$ are the estimates of the activities obtained in (q-1) iteration, D_q(i) are the variances and $1/D_q$(i) are statistical weights of the number of counts in each channel. Such proposed iteration scheme for calculation of ML estimates seems to be very effective, as a rule, 2-3 iterations provide the error of approximation of <1%.

Absorbed dose rates

The gamma dose rates in air were calculated using the results of ^{40}K, ^{232}Th and ^{226}Ra activities and dose coefficients (nGy h^{-1} per Bq kg^{-1}) 0.462, 0.604 and 0.0417 given in UNSCEAR (2000) for ^{226}Ra sub-series, ^{232}Th series and ^{40}K respectively and by assuming secular equilibrium between ^{238}U and ^{226}Ra [16]. The absorbed dose is given by:

$$D(nGyh^{-1}) = [(0.462\times A_{Ra}) + (0.604\times A_{Th}) + (0.0417\times A_k)] \quad (5)$$

Where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in $Bqkg^{-1}$ in the samples under study. We converted the absorbed dose in air into annual effective indoor and outdoor doses received by individuals. Annual estimated average effective dose equivalent received by a member is calculating using a conversion factors of 0.7, 0.8 and 0.9 $SvGy^{-1}$ for adults, children and infants, respectively [16] which is used to convert the absorbed rate to annual effective dose with an outdoor occupancy of %20 and%80 for indoors [17]. The annual effective doses are determined as follow:

$$eff\ .ann\ .Dose = \begin{cases} D(nGyh^{-1}) \times 8760\ (h) \times 0.7(SvGy^{-1}), & Indor \\ D(nGyh^{-1}) \times 8760\ (h) \times 0.2 \times 0.7(SvGy^{-1}), & Outdoor \end{cases} \quad (6)$$

where 0.2 and 8760 h are outdoor occupancy and time factors, respectively as recommended by [18].

RESULTS AND DISCUSSION

A result summary of natural radionuclide activity concentrations in samples taken from 35 locations is shown in table 1. The specific activities for ^{226}Ra, ^{232}Th and ^{40}K are reported in $Bqkg^{-1}$. The ± values

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shown are because of the *s* variation due to counting errors. The specific activity of ^{226}Ra, ^{232}Th and ^{40}K in various materials which were studied in the present work, vary from 5 to 36.2, from 6 to 54.6 and from 573.8 to 1392 Bqkg^{-1}, respectively. The world's mean values of ^{226}Ra, ^{232}Th and ^{40}K activity concentrations are 32, 45 and 420 Bqkg^{-1} [13] . A strong positive correlation of r =0.705 was observed between specific activity of ^{226}Ra and ^{232}Th in the collected soil samples. This clearly indicated the presence of significant amounts of monazite and zircon sands in the samples. A weak correlation of r =0.44 was observed between specific activity of ^{226}Ra and ^{40}K in the

Table 1. Specific activities of ^{226}Ra, ^{232}Th, ^{40}K of all samples.S2, S3 and S4 are second, third and forth samples of a given site.

Sampling sites (kind of samples)	**Specific activity (Bqkg^{-1})**		
	^{226}Ra	**^{232}Th**	**^{40}K**
Zonoz (soil)	17.7±0.1	37.2±1.1	784.4±7.4
S2	18.8±0.5	39.1±1.7	778.0±5.1
S3	18.5±0.4	38.0±0.9	772.0±6.0
S4	19.0±0.7	37.0±0.8	780.0±6.1
Zonoz (cement)	11.1±0.1	32.9±0.1	610.0±1.8
S2	12.0±0.2	32.0±0.1	614.0±1.9
S3	12.5±0.2	34.0±0.2	600.0±0.4
Zonoz (granite)	13.0±0.3	30.0±0.1	605.0±0.5
Zonoz (brick)	14.5±0.3	35.0±0.3	620.0±1.9
S2	13.8±0.2	36.0±0.2	622.0±2.0
Caulan mine1(soil)	29.8±0.3	49.5±1.4	1206.2±11.1
S2	32.1±0.9	52.8±1.8	1220.0±15.2
Caulan mine1 (gypsum)	34.6±1.0	50.1±0.9	1199.2±10.3
S2	35.5±1.1	54.6±0.9	1200.0±9.8
S3	36.2±1.8	53.8±0.8	1209.1±11.3
Caulan mine2 (soil)	10.7±0.5	11.4±0.1	1317.2±15.5
S2	10.3±0.4	13.5±0.3	1298.0±10.1
S3	10.1±0.5	12.8±0.2	1320.0±14.3
Caulan mine2 (gypsum)	14.2±0.3	18.0±0.5	1392.0±11.6
S2	14.1±0.2	16.5±0.4	1371.0±18.2
S3	16.3±0.4	20.0±0.8	1345.0±13.1
Caulan mine3 (soil)	5.1±0.1	8.2±0.1	573.8±13.6
S2	6.2±0.1	7.2±0.1	592.1±14.8
S3	7.0±0.1	8.0±0.1	602.0±18.1
Caulan mine3 (gypsum)	6.0±0.1	7.5±0.1	608.0±19.8
S2	5.5±0.1	6.0±0.1	599.0±15.0
Mahbobabad village (cement)	9.6±0.5	25.9±0.9	910.2±7.4
S2	8.1±0.3	27.1±1.1	911.1±7.6
Mahbobabad village (brick)	8.7±0.1	30.2±1.3	921.5±8.1
S2	9.9±0.3	33.5±1.8	919.0±1.8
Eyshabad village (cement)	9.0±0.4	41.8±2.1	98.0±6.5
S2	8.4±0.3	44.6±2.5	915.0±8.0
Eyshabad village (brick)	9.8±0.5	40.5±1.9	935.1±8.8
Eyshabad village (granite)	10.0±0.6	39.0±1.8	928.7±6.1
Mishu Mountain (soil)	33.6±2.1	35.1±2.0	960.0±15.0

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collected soil samples which indicates that ^{40}K concentrations may not be related to the presence of ^{226}Ra-bearing mineral sands.

The absorbed and annual effective dose rates for different samples were calculated using equations 5 and 6 and are listed in table 2. It was assumed that ^{137}Cs, ^{90}Sr and the ^{235}U decay series can be neglected as they contribute very little to the total dose from environmental background [19-21]. The minimum and maximum values of absorbed dose and indoor and outdoor annual effective doses which were found, vaied from 33.1 to 99.6 $nGyh^{-1}$, from 0.191 to 0.611 mSv

Table 2. Radiation absorbed dose and dose equivalents for all samples.

Sampling sites (kind of samples)	Annual effective dose (mSv)		
	Absorbed dose ($nGyh^{-1}$)	Indoor	Outdoor
Zonoz (soil)	63.4±1.0	0.388±0.006	0.077±0.001
S2	64.7±1.8	0.397±0.011	0.079±0.002
S3	63.7±0.9	0.391±0.005	0.078±0.001
S4	63.7±1.1	0.391±0.006	0.078±0.001
Zonoz (cement)	50.4±0.2	0.309±0.001	0.062±0.0002
S2	50.5±0.2	0.310±0.001	0.062±0.0002
S3	51.3±0.2	0.315±0.001	0.063±0.0002
Zonoz (granite)	49.4±0.2	0.303±0.001	0.061±0.0002
Zonoz (brick)	53.7±0.4	0.329±0.002	0.065±0.0004
S2	54.1±0.3	0.332±0.002	0.066±0.0003
Caulan mine1(soil)	93.9±1.5	0.576±0.009	0.115±0.001
S2	97.6±2.1	0.598±0.013	0.119±0.002
Caulan mine1 (gypsum)	96.3±1.4	0.591±0.009	0.118±0.001
S2	99.4±1.5	0.609±0.009	0.122±0.001
S3	99.6±1.8	0.611±0.011	0.122±0.002
Caulan mine2 (soil)	66.8±0.9	0.410±0.006	0.082±0.001
S2	67.1±0.8	0.411±0.004	0.082±0.0009
S3	67.4±0.9	0.413±0.006	0.083±0.001
Caulan mine2 (gypsum)	75.5±0.9	0.463±0.006	0.093±0.001
S2	73.6±1.1	0.451±0.006	0.090±0.001
S3	75.7±1.2	0.464±0.007	0.092±0.001
Caulan mine3 (soil)	31.2±0.7	0.191±0.004	0.038±0.0008
S2	31.9±0.7	0.196±0.004	0.039±0.0008
S3	33.2±0.8	0.203±0.005	0.041±0.0009
Caulan mine3 (gypsum)	32.7±0.9	0.201±0.005	0.040±0.001
S2	31.1±0.7	0.191±0.004	0.038±0.0008
Mahbobabad village (cement)	58.0±1.1	0.356±0.006	0.071±0.001
S2	58.1±1.1	0.356±0.006	0.071±0.001
Mahbobabad village (brick)	60.7±1.2	0.372±0.007	0.074±0.001
S2	63.1±1.3	0.386±0.007	0.077±0.002
Eyshabad village (cement)	33.5±1.7	0.205±0.010	0.041±0.002
S2	60.9±2.0	0.373±0.012	0.075±0.002
Eyshabad village (brick)	68.0±1.8	0.416±0.011	0.083±0.002
Eyshabad village (granite)	66.9±1.6	0.410±0.009	0.082±0.002
Mishu Mountain (soil)	76.8±2.8	0.471± 0.017	0.094± 0.003

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and 0.038 to 0.122 mSv, respectively.

It should be noted that the measured specific activity values for ^{226}Ra and ^{232}Th were analogous to the world values, which were determined for soil and building materials. However, specific activity of ^{40}K in all of samples specially Caulan mine2 with 1392 Bqkg-1 was much higher than the world values. Hence, these samples pose some health hazard for the occupants residing in these regions and for those using these building materials. The external world wide average annual effective dose for adults is 0.07 mSv [19]. According to these results, it seems that the radionuclides concentrations in the examined samples are of some radiological importance. The natural radioactivity levels in the building construction materials and soils are to a small degree higher than the acceptable limit. Consequently, permanent settlers of these regions are exposed to an increased level of natural ionizing radiation from these samples.

CONCLUSION

Measurement results presented in this work confirm that radiation exposure and attributed risk could be reduced by careful choice of building materials during construction. The experiments also showed that the spectrometer system had a better discrimination performance for low energy beta and gamma sources.

Since radionuclides are also present in water, milk, fruits, meat and so forth, therefore the tracing of these radionuclides should be carried out for agriculture products from the northwest of Iran.

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