Radioactivity Concentration in Sediment and Water Samples of Hot Springs in Mahallat and Soil Samples of Their Neighboring Environs

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ARTICLE INFO	ABSTRACT
	The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in water and sediment samples, taken from hot springs of Mahallat and soil samples from their neighboring environs, were measured using two high resolution HPGe detectors with relative efficiencies of 38.5% and 55%. In sediment samples, the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs varied from 1405.90±34.29 to 1630.56±20.84, 26.29±2.26 to 39.99±2.39, 125.40±6.23 to 137.33±7.99 and 3.53±0.53 Bqkg^{-1}, respectively. The measured specific activities of soil samples varied from 12.98±0.46 to 38.03±1.32, 18.15±0.77 to 27.63±1.58, 257.48±13.20 to 359.98±19.49 and 0.11±0.08 to 1.59±0.55 Bqkg^{-1}, respectively. In water samples taken from orifice of hot spring Donbe, Shafa and Soleymani, the specific activities varied from 0.90±0.19 to 1.62±0.24, 0.64±0.20 and 5.72±1.20 to 13.57±1.07Bqkg^{-1}, respectively. The average absorbed dose rates in sediment and soil samples varied from 623.52nGyh^{-1}to 728.14 nGyh^{-1} and 28.68nGyh^{-1}to 50.08 nGyh^{-1}, respectively.

1 INTRODUCTION

Humans are usually exposed to nuclear radiations. One of the external main sources of irradiation to human body is represented by gamma (γ) radiation (terrestrial environmental back ground radiation), emitted by natural radioisotopes. Natural environmental radioactivity mainly arises from primordial radionuclides, such as ^{232}Th, ^{235}U, ^{238}U, ^{87}Rb and ^{40}K and their decay products. These radionuclides were present in the environment since the formation time of the Earth. Significant amounts of artificial radionuclides such as ^{137}Cs and ^{90}Sr are also present in the soil and sediment resulted by the nuclear weapon tests or nuclear accidents (Kannana V. et al., 2002). Nuclear weapons tests release high radioactivity fission fragments into atmosphere and their deposition over the earth's surface by atmospheric process and their distribution depends on distance from the location of pollution origin. Water of the hot springs passes from the deep layer of the earth's crust near volcanic magma layers and returns to the surface. The dissolution of the salts of uranium and thorium enters the rocks

and sediment layers in the soil and pollutes the ground water (Mokhtari et al., 2004).The average ^{238}U content in the Earth's crust has been estimated to be 2.7 mgkg^{-1} and its concentration may be as high as 120 mg kg^{-1}in phosphate rock (Padam et al., 1996). The average ^{232}Th content of the Earth's crust is about 9.6 mg kg^{-1} (Firestoneet al., 1996). The presence of these elements in the ground water and sediment in the orifice of springs depends on the three main factors; geological, hydraulic and chemical properties of water (Azarvand, 2011). The isotopes of uranium (^{238}U, ^{235}U, ^{234}U) have non-negligible radio toxicity (WHO, 1978; Malcome, 1979). The most radio toxic of their chain are Radium isotopes and Radon gas. Among the radionuclides, ^{226}Ra, ^{222}Rn from ^{238}U decay series and ^{228}Ra from ^{232}Th decay series have the greatest impact on the human health (Piroozfar et al., 2007). Radium concentration in surface water normally ranges from 0.01 to 0.1 Bql^{-1} (Iyenger, 1990) and the highest values have been observed in the water close to uranium mining and nuclear sites (Paschoa et al., 1979; Benes, 1990). In ground water, the radium concentration can reach values up to 38 Bql^{-1} depending on geological factors (Gascoyne, 1989). Hot springs are used by visitors as spas from inside and outside of this central province of Iran. The present study attempts to determine the distribution of ^{226}R, ^{232}Th, ^{40}K and ^{137}Cs in water and sediment of hot springs of Mahallat in Iran and soils from their neighboring environs using two HPGe gamma ray spectrometer system. The degree of harmful and hazards for visitors depends on the amount of radioisotopes in hot springs water and the residence time in and around hot springs water (Nakano-Ohta T. et al., 2007). In addition, the total dose rate in air due the presence of ^{238}U, ^{232}Th and ^{40}K in the sediment and soil samples was computed.

2 MATERIALS AND METHODS
2.1 Sample collection and preparation

Abegarm-e-Mahallat is a region in north east of Mahallat, a town in the central part of Iran. Five hot springs, which exist in Abegarm-e-Mahallat, are Shafa, Soleymani, Donbe, Soda, and Romatism with a mean temperature of 46.1°C. In this work, water samples were taken from Shafa, Donbe, and Soleymani. Polyethylene bottles were used for sampling water. Nitric acid was added to decrease the sample's PH down to 2 that is necessary to prevent the wall absorption (Peres-Moreno et al., 2001). For comparison, drinking water sample was taken from the west of Arak city originated from Gharekahriz zone. Sediment samples were taken from the orifice of hot springs of Donbe and Solaymani, and two soil samples from their neighboring environs in order to identify the original radioactivity source in the Abegarm- Mahallat region. Two soil samples were taken 50 and 10 m away from the orifice of Donbe and Soleymani at 5 cm soil depth, using the template method. They were named A and B, respectively. The sediment and soil samples were dried at a 200° C oven for 12 hours. In order to obtain homogeneous samples, the dried soil and sediment samples were pulverized by a grinder into fine powder and passed through 10-mesh and 50-mesh screen (IAEA-TECDOC-360, 2003). All samples were prepared in Marinelli Beaker and Negin containers for gamma spectrometry analysis. The collection of samples requires particular care because radon is a short lived gaseous nuclide and tends to escape from samples. In this study, Marinelli Beaker and Negin with volume 800 cc and 300 cc were used and sealed, respectively. Weight of dried sediments and soils were 950 g for Marinelli Beaker and 293 g for Negin containers. After the minimum 60 days of preparing sealed samples, gamma ray was registered because this time it is necessary to

take radioactive chain equilibrium (International Atomic Energy Agency 2003).

All water samples, sediment samples and soil sample A were prepared in Marinelli container with 950 g net weight and then were sealed. The soil sample B with a mass of 293g was packed in Negin container and then was sealed.

2.2 Experimental setup

Gamma ray spectra were done using two HPGe spectrometers coaxial p type with relative efficiency of 38.5% and 55% setup and multichannel analyzer of 4096 channel. The energy resolutions of these detectors were 1.95 keV and 1.80 keV for gamma energy line at 1332.520 keV due ^{60}Co, respectively. Operating voltage was 3000 V and the detectors were shielded in a chamber of three layers 100mm thick lead and two layers of 2 mm thick of each by cadmium and copper. This shield serves to reduce the background radiation. The soft components of cosmic ray are reduced to a very low level by 100 mm of lead shielding. The X-ray (73.9 keV) emitted from lead by its interaction with external radiation is suppressed by copper layer and cadmium layer successively absorbs thermal neutrons produced by cosmic ray. To minimize the effect of scattering radiation from the shield, the detectors were located in the center of their own chamber. The samples were placed in a face-to-face geometry over detectors for 86400 s. The γ-ray spectrum measured from sediment sample of Soleymani by 38.5% HPGe detector is showed in Fig. 1. The respective efficiency calibrations were carried out for Marinelli Beaker and Negin standard source including ^{241}Am, ^{109}Cd, ^{57}Co, ^{60}Co, ^{65}Zn, ^{152}Eu and ^{137}Cs. According to registered gamma ray spectra, absolute efficiency of detector (ε_i) can be calculated by:
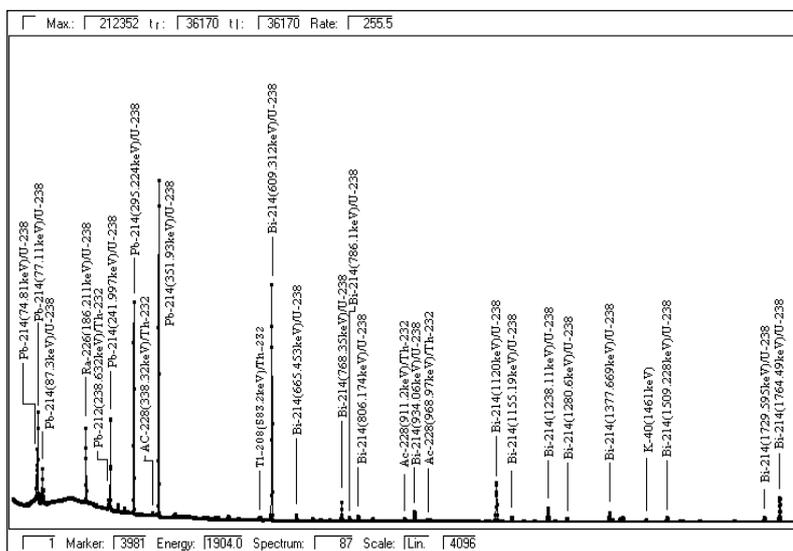

Fig. 1. Gamma ray spectrum of sediment sample collected from Soleymani hot spring registered by HPGe detector with 38.5% relative efficiency.
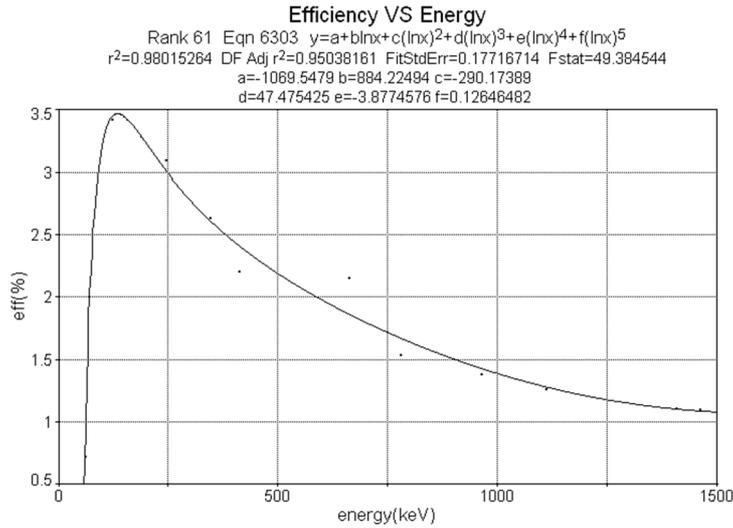
Efficiency VS Energy
Rank 61 Eqn 6303 y=a+blnx+c(lnx)²+d(lnx)³+e(lnx)⁴+f(lnx)⁵
r²=0.98015264 DF Adj r²=0.95038161 FitStdErr=0.17716714 Fstat=49.384544
a=-1069.5479 b=884.22494 c=-290.17389
d=47.475425 e=-3.8774576 f=0.12646482

Fig. 2. Efficiency curve for standard water sample Marineli Beaker arrangement using HPGe with 55% relative efficiency.

Efficiency VS Energy
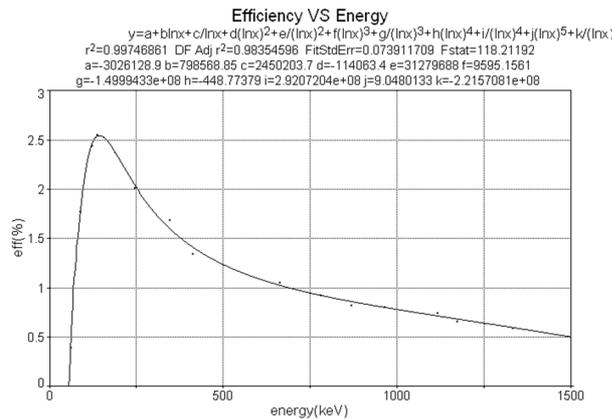y=a+blnx+c/lnx+d(lnx)²+e/(lnx)²+f(lnx)³+g/(lnx)³+h(lnx)⁴+i/(lnx)⁴+j(lnx)⁵+k/(lnx)
r²=0.99746861 DF Adj r²=0.98354596 FitStdErr=0.073911709 Fstat=118.21192
a=-3026128.9 b=798568.85 c=2450203.7 d=-114063.4 e=31279688 f=9595.1561
g=-1.4999433e+08 h=-448.77379 i=2.9207204e+08 j=9.0480133 k=-2.2157081e+08

Fig. 3. Efficiency curve for standard soil sample Negin container using HPGe with 38.5% relative efficiency.

$$\varepsilon_i = \frac{N_i}{P_n(E_i).\ Act.T} \qquad (1)$$

Where N_i is the net count under the full energy peak corresponding to E_i, Act is radionuclide activity, $P_n(E_i)$ is the photon emission probability with energy E_i, and T is the counting time. In Fig. 2 and 3 the efficiency curves for Marinelli standard source are shown, respectively. In Table 1, the functions fitted to experimental data have been given in Eqs. (2)-(6).

The knowledge about the detector absolute peak efficiencies is required to obtain reliable activities as the counting rates depend on the characteristics of matrix, source geometry, and source-detector configuration.

Table 1
Efficiency equations for different sample configurations.

Equation	configuration	No
Y= a+ blnx+c(lnx)² +d(lnx)³ e(lnx)⁴ +f(lnx)5	Standard water sample, Marinelli, HPGe 55%)	(2)
Y= a+blnx+c(lnx)²+d(lnx)³+ e(lnx)⁴+f(lnx)⁵	standard water sample, Marinelli, HPGe 55%)	(3)
Y= a+blnx+c/lnx+d(lnx)²+e/(lnx)²+f(lnx)³+ g/(lnx)³+h(lnx)⁴+i/ (lnx)⁴+j(lnx)	standard water sample, Marinelli, HPGe 38.5%	(4)
Y=a+blnx+c(lnx)²+d(lnx)²+e/(lnx)³+e(lnx)⁴ +f(lnx)⁵+ g(lnx)⁶ + h(lnx)⁷	standard soil sample, Marinelli, HPGe38.5%	(5)
Y=a+blnx+(c/lnx)+d(lnx)²+e/(lnx)²+f(lnx)³+g/(lnx)³+ h(lnx)⁴+i/(lnx)⁴+ j(lnx)⁵+k/ (lnx)⁵	standard soil sample Negin, HPGe 38.5%	(6)

y denotes the efficiency (%). x(keV) is the gamma energy. a, b, c, d, e, f, g, h, i and j are constants.

It is essential to determine the efficiency for each setup separately.

In Figs. 2 and 3, the efficiency curves versus γ-photon energy have been shown. The registered gamma ray spectra were analyzed and specific activities calculated using OMNIGAM Ortec software. In all the analyzed spectra, the correction was done for background gamma ray measured for empty Marinelli and Negin containers in the same condition.

In order to calculate the specific activity of radionuclides, we used the following equation.

$$Act = \frac{NetArea}{\varepsilon\,(B.R\,\%).t.v \quad or \quad (m)} \tag{7}$$

Where Net Area is the net count under peak, Act (Bql^{-1} or $Bqkg^{-1}$) is the activity concentration, ε is the efficiency for gamma ray, B.R. (%) is the branching ratio of gamma intensity, t is the counting time of spectrum, v (litr) is the volume, and m (kg) is mass of sample.

The γ-transition energies 241.99keV (^{214}Pb), 295.22keV (^{214}Pb), 351.93 keV (^{214}Pb), 609.31keV (^{214}Bi), 1120.28keV (^{214}Bi), 1764.47keV (^{214}Bi) were used to determine the mean activity concentrations of the ^{226}Ra series. The γ-transition energies 238.63 keV (^{212}Pb), 338.32 keV (^{228}Ac), 911.2 keV (^{228}Ac) 968.97 keV (^{228}Ac) were used to determine the mean activity concentrations of the ^{232}Th series.

The γ-transition energy 1460.80keV was used to determine ^{40}K activity concentration in different samples. The γ-transition energy 661.66 keV was used to determine ^{137}Cs activity concentration.

2. 3 Radiological hazard assessment
2.3.1 Radium equivalent activities

Radium equivalent activities can be calculated according to specific activities of ^{226}R, ^{232}Th and ^{40}K by a single quantity which takes into account the radiation hazards associated with them. This can be determined by (Amekudzie et al., 2011):

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_K \tag{8}$$

Where C_{Ra}, C_{Th} and C_K are radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively. The maximum amount of Ra$_{eq}$ in building materials must be lower than 370 Bq.kg^{-1} for safe use (Amekudzie et al., 2011).

2. 3. 2 Representative level index values

Another radiation hazard index called the representative level index, used to estimate the level of the gamma radiation associated with different radionuclides with specific activities can be defined using equation 9 (NEA Group, 1979).

$$I\gamma = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \tag{9}$$

This index can be used to estimate the level of gamma radiation hazard associated with the radionuclides in materials.

2. 3.3 Dose rate calculation

According to the formula given by UNSARE (1998) report, the absorbed gamma radiation dose rate is estimated as (Kannana et al., 2002):

$$D = 0.427\ C_{Ra} + 0.662\ C_{Th} + 0.0432 C_K \tag{10}$$

Table 2

Radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in water samples (Bq.l^{-1}), collected from different hot springs of Mahallat.

Location	^{226}Ra	^{232}Th	^{40}K
Donbe	0.90±0.19	<MDA	5.72±1.20
Shafa	1.62±0.24	<MDA	12.39±1.66
Solaymani	1.09±0.15	0.64±0.20	13.57±1.07
Drinking water of Arak	<MDA	<MDA	5.32±0.99

Table 3

Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in sediment and soil samples, collected from different locations of Abegarm-e-Mahallat.

Location	Specific Activity (Bq.kg^{-1})			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Sediment of Donbe	1405.9±34.30	26.29±2.26	137.33±7.99	3.53±0.53
Sediment of Solaymani	1630.56±20.84	39.99±2.39	125.4±6.23	<MDA
Soil sample of A	12.98±0.46	18.15±0.77	257.48±13.20	0.11±0.08
Soil sample of B	38.03±1.32	27.63±1.58	359.98±19.49	1.59±0.55

3 RESULTS AND DISCUSSION

The radioactivity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were determined in sediments and waters of hot springs of Mahallat, and also soil of its neighboring environs. The ^{226}Ra reactivity concentration in water samples varied from 0.90±0.19 Bq.l^{-1} in Donbe spring up to 1.62±0.24Bq.l^{-1} in Shafa spring. The ^{232}Th was observed only in Soleymani spring water with amount of 0.64±0.20Bq.l^{-1}. In another hot spring, containment of this radionuclide was lower than MDA. The ^{40}K activity concentration in water samples varied from 5.72±1.20 Bq.l^{-1} in Donbe Spring to 13.57±1.07 Bq.l^{-1} in Soleymani Spring. The artificial radionuclide ^{137}Cs was lower than MDA (see Table 2). In Arak consumer water sample, only ^{40}K was determined and its activity concentration was 5.32±0.99 Bq.l^{-1}.

In sediment samples, high levels of ^{226}Ra radioactivity concentrations were observed that varied from 1405.9±34.30 Bqkg^{-1} in Donbe hot spring to 1630.56 ± 20.84 Bqkg^{-1} in Soleymani hot spring. The activity concentrations of ^{40}K,

^{232}Th and ^{137}Cs in sediment and soil samples are given in Table 3.

The highest radioactivity concentration of ^{137}Cs (3.53±0.53 Bq.kg^{-1}) was measured in sediment sample of Donbe. Radium equivalent activities (Ra$_{eq}$) varied from 1454.60 to 1697.42 and from 58.76 to 105.26 in Bq.kg^{-1} for sediment and soil samples, respectively. Further, for sediment samples they were higher and for soil samples were lower than the limit set in OECD report (370 Bq.kg^{-1}).

Also, the average absorbed dose rate in sediment and soil varied from 28.68 up to 728.14 nGyh^{-1}. Te representative level index values for investigated samples are given in Table 4.

4 CONCLUSION

In this study, the radioactivity concentrations in soil, sediment, and water samples taken from Abegarm-e-Mahallat, a region in northeast of Mahallat of Iran, were determined. This region contains five hot springs: Shafa, Solaymani, Donbe, Soda, and Romatism. We studied the specific activities of

Table 4
Radium equivalent activities (Bqkg^{-1}), Representative level index value (I$_\gamma$) in Bqkg^{-1}, Dose rate (D) in nGyh^{-1} in sediment and soil samples in hot springs in Mahallat in Iran

Sample name	Ra$_{eq}$ (Bqkg^{-1})	I$_\gamma$ (Bq.kg^{-1})	Dose rate (nGyh^{-1})
Sediment of Donbe	1454.60	9.72	623.53
Sediment of Soleymani	1697.42	11.35	728.14
Soil sample A	58.76	0.44	28.68
Soil sample B	105.26	0.77	50.08

the ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs radionuclides using high resolution gamma ray spectrometry system. In sediment samples, the present mean specific activity of ^{40}K is lower and the ^{226}Ra specific activity is much higher than the world's average. The ^{232}Th activity concentrations in the soil samples of Soleymani are approximately about the world's average, and sometimes are more. The average radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the world are 35, 30 and 400 Bq.kg^{-1}, respectively (UNSCEAR 2000). The ^{137}Cs radionuclide was observed in sediment and soil samples, which indicates radioactivity released by the nuclear power plant accidents in the world has been transferred to Iran by atmospheric processes. Abegarm-e-Mahallat is a region with high levels of natural radiation. It is highly recommended to investigate its effects on the people and tourists who are using these hot springs. We are planning to trace some of radionuclides in rivers that flow into the Persian Gulf.

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