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Assessment of Radium Isotopes in Some Hot Springs in the Kingdom of Saudi Arabia

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Abstract

Radioactivity measurements have been conducted to determine the activity concentrations of radium isotopes (^{226}Ra and ^{228}Ra) in water Samples acquired from some hot springs at the south and south-western area of the Kingdom of Saudi Arabia. The radium isotopes were measured by gamma spectrometry using high purity germanium detector, after radiochemical separation of the isotopes with ion-exchange chromatography using a strong cation resin. The ^{226}Ra showed an average activity concentration of 82.4mBq/L, while the average activity concentration of ^{228}Ra was 236.4 mBq/L. These values were comparable to the values of radium activities in thermal waters reported in literature. The chemical analyses showed that good correlations have been observed between the radium activities and the concentrations of sulphates and total dissolved soilds in the investigated thermal waters. Further studies regarding The radon concentrations, the immersion time for bathers and the resulting annual committed effective dose are considered.

Keywords: Hot springs, radium, cation exchange resin, TDS, Sulphates, Saudi Arabia.

1. INTRODUCTION

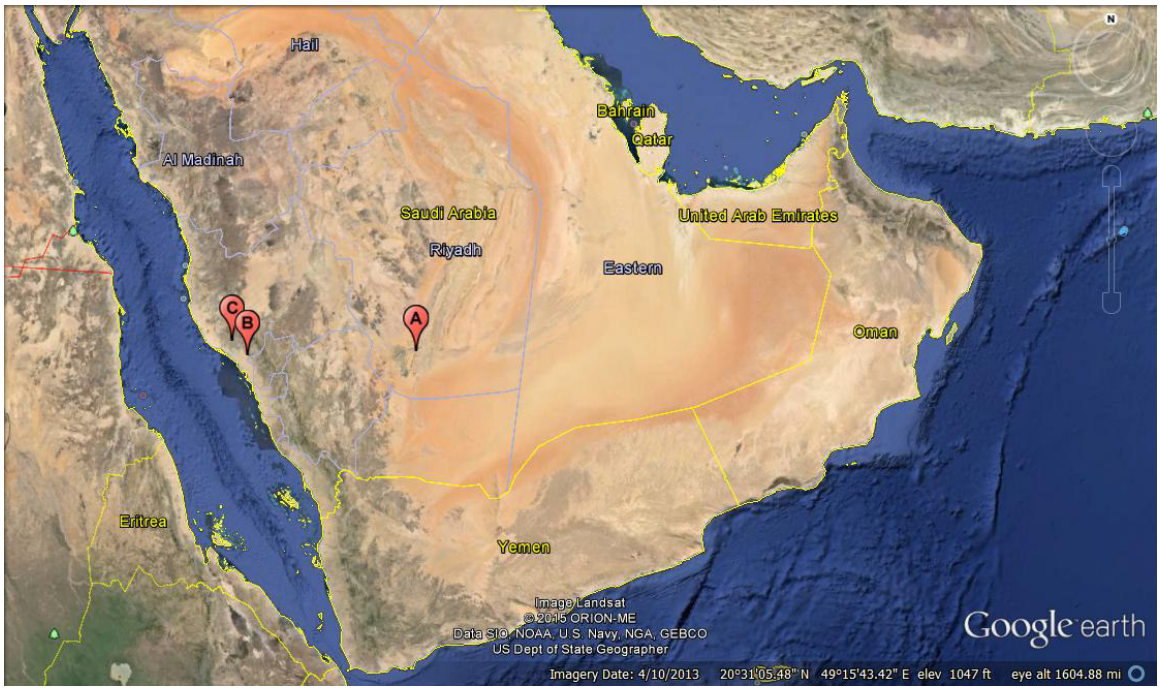
One of the most important aspects of radioecological studies is to provide the scientific basis for prediction of the impact to man and his environment, due to different radionuclides. To assess these impacts quantitatively, it is necessary to estimate the activity concentrations of these isotopes in our environment. In Saudi Arabia, there are ten geothermal springs, many of them are located in the south (Wadi Al-Dawaser) and south-western area (Wadi Al-Lith area) [1]. The four hot springs selected for the present study are the most popular in Saudi Arabia, as they have been exploited for their therapeutic and curative effects. 226Ra has been measured worldwide in many water supplies, especially in mineral and thermal groundwater. Radium-226 in groundwater is derived mainly by the interaction between the groundwater aquifer and radium-bearing materials, such as rocks, soil and ore deposits which are often responsible for enhanced ^{226}Ra concentrations [2]. It is well known that some thermal waters may contain high concentrations of ^{226}Ra [3], which could be of a considerable radiation hazard, as it might lead to the buildup of ^{222}Rn and related daughters, causing potential health implications due to the long exposure time. Considering the popularity of these spas and the habits of the population to spend long hours to use the beneficial effects of these springs, it was a useful approach to estimate the radioactivity from these thermal waters. Therefore, the aim of this study is to determine the activity concentrations of radium isotopes (^{226}Ra and ^{228}Ra) in the selected hot springs, which may enable us to assess if these springs could be of considerable radiation hazard to the public visiting these hot springs for both curative and recreational purposes.

2. METHODOLOGY

Unless otherwise indicated, all references to water refer to deionized water (DDW).

2.1 Sampling

The studied thermal springs are situated at two different areas. Two samples were collected from Wadi Al-Dawaser, Al Gubah center, which is about 650 km from Riyadh city, at this location (A), two springs were selected for sampling. The third sample (B) was collected from Bani Helal spring at a distance of 70 km east of Al-Lith governorate, while the last sample was collected from Ain Al Hara, which is located at a distance of 30 km from Al-Lith governorate, as shown in map.



Five liters of each water sample were collected directly from the spring. The sampling containers were rinsed thoroughly with the water to be sampled prior to sample collection. For radioactivity measurements, the water samples were filtered through 0.45µ membrane filter, acidified with 11 M HCl at the rate of 10 mL per liter of sample immediately after filtration to avoid the adsorption of radionuclides on the walls of the container and growth of micro-organisms, and transferred to polyethylene bottles. For chemical analysis, the water samples were collected in suitable bottles without the acidification step, and the pH and total dissolved solids (TDS) were monitored with a calibrated multi-parameter TDS and pH meter (Hach HQ40).

2.2 Materials and Apparatus

Radium extractions from water samples were carried out using a strong cation exchange resin, Purolite C-100, Na form, supplied by Veolia Water Co. (Riyadh, Saudi Arabia). Standard reference solutions of 226Ra and ^{228}Ra were supplied by the National Institute of Standards and Technology (NIST), (SRM 4967A, SRM 4339B). The 133Ba standard solution was supplied by North American Technical Services (NATS) (EZ-83879-767). The cation exchange resin was used in a column mode with BioRad Glass Econo columns of 0.9 cm diameter, together with polypropylene funnels and Teflon end fittings connected with plastic taps. All gamma radioactivity measurements were carried out using a Canberra HPGe coaxial detector (Model GC4020) with relative photo-peak efficiencies of 40% for the 1332 keV line of 60Co. The germanium detector was connected to a Digital Spectrum Analysis model DSA-1000. The alpha spectrometric analysis were carried out using a Canberra Alpha Analyst, with a chamber containing a passivated implanted planar silicon (PIPS) detector with an active area of 450 mm2. The efficiency of the detector was calibrated against a standard alpha multi-source (67970-121, Analytics Co.) using the certified activity of the measured radionuclides. Diphonix Resin (50-100 and 100-200 mesh) was supplied from Triskem International, 35170 Bruz, France. All other chemicals used in this study, including KMnO4, isopropanol, ammonium sulphate and different mineral acids were of analytical grade.

2.3 Radioanalyses and Measurements

The radium isotopes (^{226}Ra and ^{228}Ra) were determined in the water samples using the procedure described by A.El-Sharkawy et al, 2013 [4]. Four liters of each sample were allowed to pass through the purolite resin packed columns, the resin was transferred to standard counting containers and the containers were tightly sealed for four weeks to allow secular equilibrium between ^{226}Ra, ^{228}Ra and their decay products. The efficiency calibration of the germanium detector for the radium isotopes (Ra-226 and Ra-228) was carried out using standard samples.

Known activity resins were prepared by spiking water (DDW) samples with known amounts of ^{226}Ra and ^{228}Ra. The spiked resin samples containing a known amount of the radionuclide of interest were used to provide an identical matrix with a known activity, and all other conditions were followed typically (flow rate, resin volume, counting time, geometry). The 226Ra activities were determined via its daughters 214Pb and 214Bi through the gamma energy lines 295.22, 351.93 and 609.31 keV. The 228Ra activities were determined through the gamma energy lines of 338.32 and 911.2 keV. The calculated specific activities were basically performed using a comparison method:

$$A_{unk} = \frac{A_{std}}{CR_{std}} . CR_{unk}$$

where;

Aunk is the calculated activity of the sample;

Astd is the activity of the standard resin;

CRstd is the counting rate for the standard resin; and

CRunk is the counting rate of the unknown sample.

2.4 Quality Assurance

For quality assurance and validation purposes, blank samples were prepared in the same manner as the corresponding samples, and measured for background estimation. Reference water samples were determined using the same analysis and measurement protocol, and were compared against their certified values to test the closeness of the measured samples to its reference values. Also, intercomparison tests were carried out with the French Institute for radioprotection and nuclear safety (IRSN, 96 SL-300 reference sample), and with the International Atomic Energy Agency, IAEA-CU-2010. Errors were propagated due to nuclear counting statistics, tracers and volume. To evaluate the accuracy of the method, a low activity selected water sample (TS4) was analyzed for 226Ra following an alpha spectrometric method described by S. Nour et al, 2004 [5]. In this approach, 226Ra and 133Ba tracer are co-precipitated with MnO2, dissolved in 2M HCl, loaded into a Diphonix resin column to eliminate other interfering radionuclides and the collected Ra/Ba fraction is then precipitated using BaSO4 micro-precipitation by adding (NH4)2SO4, barium carrier and isopropanol in a tube, mixed well and allowed to stand in an ice bath for 30 min before vacuum filtration [6]. The filter is mounted on a disk, counted by gamma for the 133Ba recovery and the 226Ra is assessed by alpha spectrometry.

3. RESULTS AND DISCUSSION

In the following section, the results of the radioactivity levels of radium isotopes in the collected water samples will be represented.

3.1 Chemical Analyses of Water Samples

The water samples were analyzed for total dissolved solids (TDS) and the sulphates concentration, as shown in table (3-1). It can be seen that there is a relatively large variations in the TDS and sulphate content between TS1 and TS2 samples, which were collected from AlGubah site, about two kilometers apart.

3.2 Radium Isotopes in Water Samples

The gamma result of ^{226}Ra in sample TS4 was within the low limit of detection (less than 1 pCi). The activity concentration of ^{226}Ra in TS4, when analyzed by alpha, was 17.6 mBq/L. The activity concentrations of ^{226}Ra and ^{228}Ra in the water samples are represented in fig.3-1. The activities of radium in the hot springs water samples ranged from 17.6 to 179.4, with an average of 82.4 mBq/L for ^{226}Ra, and for ^{228}Ra, it ranged from 51.8 to 431.2, with an average of 236.4 mBq/L.

These values are comparable with those reported from Morocco for ^{226}Ra and ^{228}Ra (0.65 and 0.09 Bq/L respectively) [7], and ^{226}Ra in hot springs in Taiwan (37.8 mBq/L) [8]. Radium-226 has been estimated in 115 slovenian springs, and found to be in the range of 7.8-43.1 Bq/l [9], and the activity concentration of 226Ra in hot springs water in Yemen was found to be 3.48 Bq/L [10]. In thermal waters from the French Massif Central, the activity of ^{226}Ra ranged from 0.95 to 2.3 Bq/L [11].

3.3 Correlations Between Radium Activities and Water Chemistry

It has been observed that samples TS2 and TS3, with the lowest SO4-2 content, showed relatively higher radium activities, as represented in fig.3-2, there is a strong correlation between radium activities and the concentrations of SO4-2 in the investigated hot springs samples, which may be explained by the co-precipitation of radium with sulphates [8], and several authors reported that the disappearance of sulphates allow the presence of higher concentrations of radium in water [12].

The variation of radium activities in the springs with the TDS content showed a positive correlation as observed in fig. 3-3. Several studies have revealed a positive correlation between radium and chloride content [13]. Although the lowest radium activity was found in sample TS4, which showed the maximum value of temperature (100ºC), we couldn't conclude an inverse correlation between radium activities and the temperature of water samples. In literature, Gordana et al, 1996, has reported that the ^{226}Ra content of thermal waters had an average of 3.2 Bq/L at a temperature of 96.0 ºC [14], and it was reported that the ^{226}Ra activities are generally correlated to spring temperatures [15, 16].

4. CONCLUSION

The activity concentrations of ^{226}Ra and ^{228}Ra have been determined in water samples collected from some hot springs in Kingdom of Saudi Arabia. The ^{226}Ra showed an average activity of 82.4 mBq/L, while the average activity concentration of 228Ra was 236.4 mBq/L. These values were comparable to the values of radium activities in thermal waters reported by many authors in literature. The radium activity concentrations in the investigated springs have shown a good inverse correlation with the concentrations of SO4-2, and a direct correlation with the TDS content in these hot springs. In Morocco, M. A. Misdaq et al, 2012 [17] estimated a committed effective dose to skin due to ^{238}U, ^{232}Th and ^{222}Rn average activities of 59.2, 14.6 mBq/L and 57.0 Bq/L respectively. The annual committed effective dose to skin from immersion of bathers in thermal water had an average value of 1.33 mSv/y, based on six hours/week contact time. Therefore, additional measurements of radon concentrations, and the annual committed effective dose should be estimated to assess any radiological hazards to the public visiting these hot springs for curative and recreational purposes.

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Table (3-1) : results of chemical analysis

Code	Location	TDS mg/L	Sulphate mg/L	Temperature ºC
TS1 (A)	AlGubah 1	3200	750	74
TS2 (A)	AlGubah 2	3660	125	85
TS3 (B)	Bani Helal	3890	250	92
TS4 (C)	AlHara	2300	625	100

Presents the concentrations of total dissolved solids and the sulphates in mg/L in the thermal springs water samples.

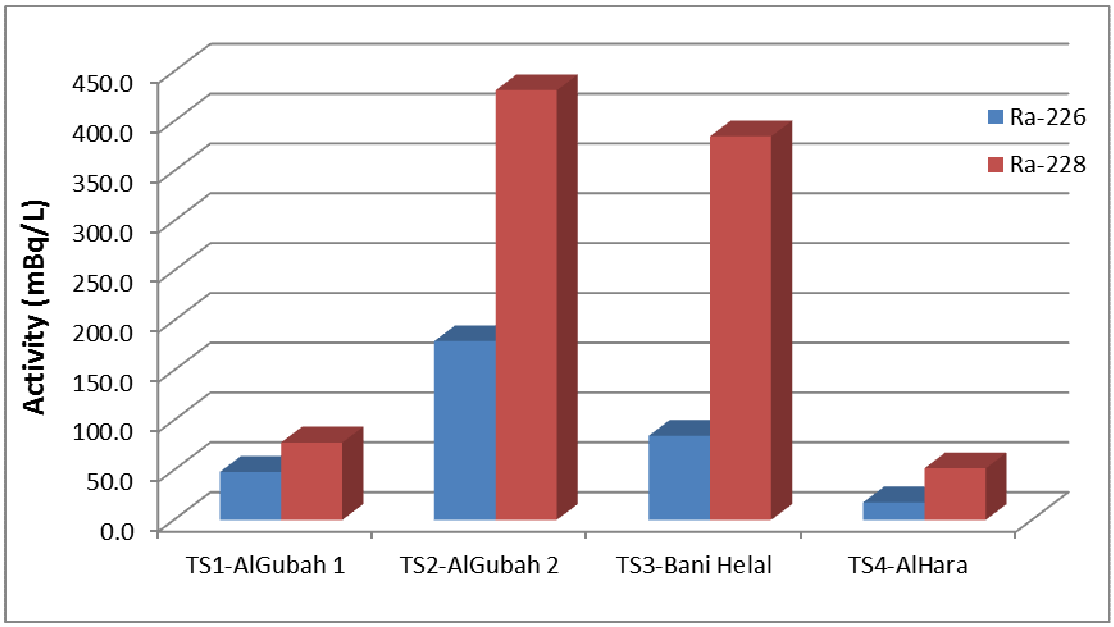

Fig. 3-1 : The activity concentrations of 226Ra and 228Ra in hot springs water samples.

The figure shows the variation of radium isotopes (^{226}Ra and ^{228}Ra) activities in the investigated spring water samples.

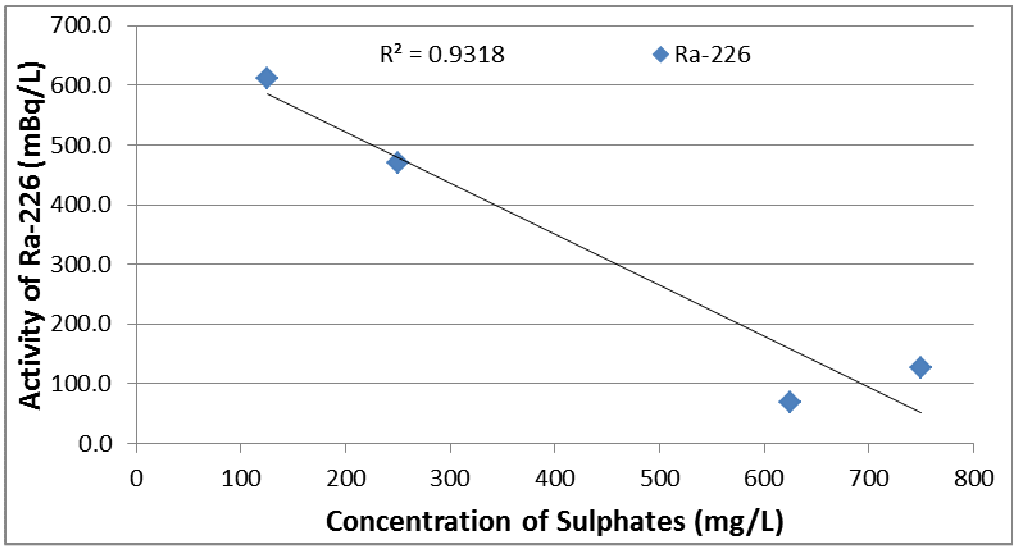

Fig. 3-2 : Correlation between 226Ra activities and sulphates content in hot springs water.

This figure presents the strong correlation between the ^{226}Ra activity concentrations and the sulphates content in the springs water samples.

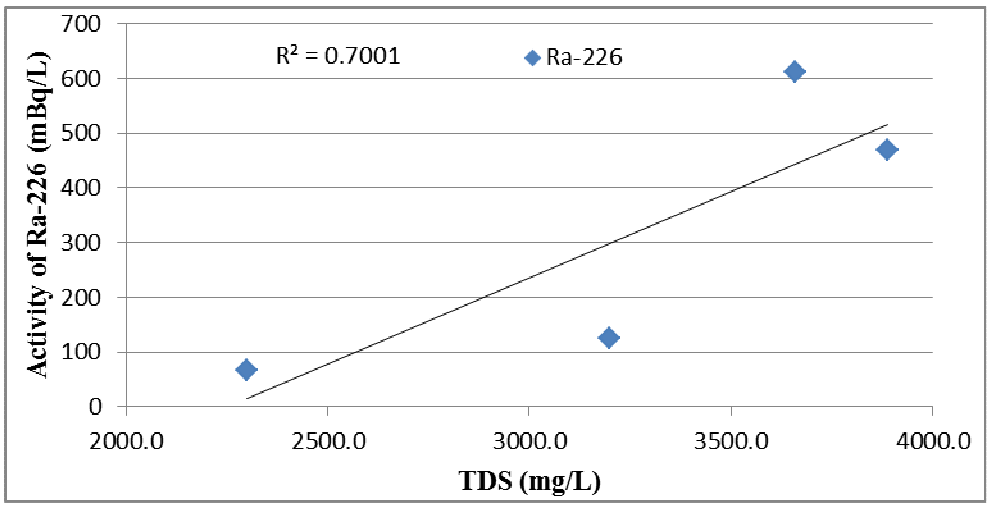

Fig. 3-3 : Correlation between 226Ra activities and TDS content in hot springs water.

This figure illustrates the correlation between the ^{226}Ra activity concentrations and the total dissolved solids concentration in the spring water samples.

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