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^{222}Rn in Some Underground Water-Samples and Examination of Correlation to ^{238}U Concentration

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The concentration of ^{238}U and ^{222}Rn has been assessed in underground water samples collected from the Makkah Al-Mukarramah area west of Saudi Arabia. Observed radon activities lie in the range 0.6-3.9 Bq/L. This range is within the natural limits of concentration of radon. Radon concentrations observed in this study are compared to those observed in a study of radon concentrations in the central region of Saudi Arabia. In the experimental setup, CR-39 detectors are exposed to radon emanating from water samples for 30 d. Uranium concentrations lie within the range 1.02-31.97 μg/L. No direct correlation is found between concentrations of the two isotopes; however radon content in water is much higher than that of uranium.

Key Words: Radon, Uranium, Environmental radioactivity, Nuclear track detectors, Underground water.

INTRODUCTION

Radioactive elements in the Earth's crust and extraterrestrial sources are the main sources of natural radiation. Two categories of naturally occurring isotopes can be identified, uranium, thorium and actinium series and singly occurring isotopes. The uranium series originates with ^{238}U which decays in successive steps to the stable ^{206}Pb isotope. High levels of uranium in food and drinking water may pose a health risk due to internal exposure to α-radiation and chemical toxicity. Uranium measurement in water samples is important to assess risks and compare to accepted levels.

^{222}Rn is the only gaseous member of the uranium series. It is an α-emitter with a half-life of 3.82 d. Because of its gaseous nature, radon escapes from rocks, soil and water that contain its immediate parent, ^{226}Ra. Dissolved radon escapes water and if present in high concentrations it may result in elevated levels of radon and its progeny in the dwellings. Correlation between exposure to radon and its daughters and lung cancer has been established in the late 1960's. From the decay chain, it can be seen that the half-lives of the short-lived products of radon vary from 164 microseconds for ^{214}Po to a maximum of 26.8 min for ^{214}Pb and that all these daughter products of radon are heavy metals. When formed the decay products of radon are

electrically charged and tend to attach themselves to the inert aerosols that are normally present in the atmosphere. The ICRP standard man inhales about 2.3×10^4 liters per day and this volume contains many of pollutants such as aerosols, dust and microorganisms[1].

Radon levels in eight water supply municipalities of the Central Region of Saudi Arabia were measured and 77 wells and 6 treatment plants were investigated[2]. No data on uranium or radon levels are available for underground water in the western region. The aim of this study is to investigate uranium and radon levels in underground water collected from selected locations in and around Makkah area and to examine if there is any correlation between ^{222}Rn and its grand-parent, ^{238}U.

Radon and α-activity determination is an active research area. Investigation of radon and its parent nuclides in soil, industrial materials, waters, consumable products and air has attracted many investigators. Measurement of radon in different types of water samples was carried out by many investigators. Marques *et al.*[3] measured radon activities in several types of natural waters from the Satos region in Brazil using makrofol E polycarbonate plastic detectors. The measured radon activities ranged from 0.95 to 36.00 Bq/L for ground waters, from 0.43 to 0.54 Bq/L for sea waters, from 0.39 to 0.47 Bq/L for tap waters and from 0.43 to 2.40 Bq/L for river waters. Ghosh *et al.*[4] studied the presence of α-emitting nuclides in drinking water in different places of South and North of West Bengal, where arsenic contamination is severe. 2.5-36.5 Bq/L concentration range was found in waters in Mexican state of Chihuahua[5].

Solid state nuclear track detectors are widely used for α-activity determination in general and for radon determination. Measurement of radiation using solid state nuclear track detectors is based on the fact that when a heavily charged particle traverses a material it produces tracks. By enlarging these tracks, an indication of the concentration of the charged particles can be obtained. Commercially known CR-39 plastic track detectors have been intensively used in radon dosimetry and other radiation related measurements[4,6,7].

EXPERIMENTAL

Uranium determination: ICP-MS technique was used for uranium determination. ICP-MS is a powerful technique for elemental determination and can be used to determine most elements of the periodic table with a detection limit in the range of few parts per billion (ppb). Water samples were directly measured for uranium content. Calibration and quality control was performed using standard solutions provided by Perkin-Elmer.

Radon measurement: Solid state nuclear track detectors were used for direct determination of radon content in water samples. The detection chamber consists of a CR-39 detector made to hang in an empty 2 L glass container. The container was mounted on the water holding beaker. The system was sealed carefully to avoid any gas leakage. Two round holes were made in the lid of the water-holding container,

fiberglass filters (Ahlstrom 121) were placed to allow diffusion of radon into the detection chamber and to discriminate against ^{220}Rn as it has a very short half-life. The filters will also limit the humidity in the detection chamber. Fig. 1 represents a schematic drawing of the ^{222}Rn detection chamber set up. Detectors were exposed to samples for 30 days. This period ensures that all radon present in the water will decay.

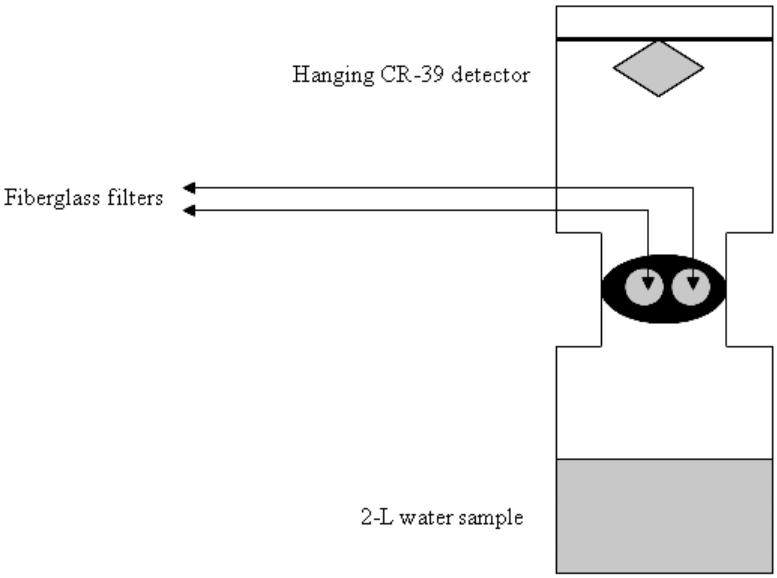

Fig. 1. Schematic drawing of ^{222}Rn detection chamber set up

After exposure detectors were etched using 20 % KOH solution at 70 ºC for 6 h using a temperature regulated water bath. Detectors were removed from the etching solution, washed in deionized water at 70 ºC twice, wiped clean using isopropyl alcohol wipes and stored in a sealed bag for counting. Tracks were manually counted using an optical microscope using 40x objectives.

A one mL standard solution of ^{226}Ra with absolute activity of 21.7288 Bq/mL was used to determine the response function of the detection system. In the passive determination of radon no standard reference material can be used. Every radon containing material will have its own unique characteristics. Quality assurance is achieved through the use of standard radon source (generated from radium), check of the linearity of the response of the detection system and blind tests. For calibration purposes, a detector was exposed to radon generated from the standard radium solution for 15 d. The reference detector was placed in an atmosphere of radon generated from the ^{226}Ra source. 21.7288 Bq ^{226}Ra in a container with certain volume is eventually equivalent to a saturation ^{222}Rn concentration of 21.7288 Bq/volume. However, the detector was exposed to radon from the radium standard for a shorter period (15 d) than is necessary for saturation in growth (*ca.* 30 days). Radon will

start with initial concentration of 0.0 Bq as the container was opened to place the detector, so any radon present will escape from the container. After sealing the container, radon concentration will start to rise according to secular equilibrium. The activity to which the reference detector was exposed during the exposure period is the area under the curve from 0 to the fifteenth day of exposure. This activity can be evaluated by integrating the secular equilibrium equation between the limits of 0 and 15.

$$\lambda N\, dt = l\, N^{o} (1 - e^{-\lambda t})\, dt$$

where, λ is the decay constant for ^{222}Rn; N^{o} is the initial concentration of ^{226}Ra; t is the time of exposure.

Poisson statistics are assumed to apply to track counting (in common with other nuclear events). In these statistics, the error is given by the square root of the events actually counted[8]. Other sources of error include radium solution volume error (< 0.3 %) and systematic errors such as detection device volume < 0.5 % and background correction.

RESULTS AND DISCUSSION

Uranium and radon concentrations obtained for underground water samples from different areas in the Makkah area using ICP-MS and CR-39, are presented in Table-1. It is clear from the table that radon activities in investigated samples lie in the range 0.4-2.9 Bq/L. These activities are well below the maximum level of contamination of 11.1 Bq/L proposed by the US environmental protection agency. Generalization of this conclusion cannot be made until an extensive survey of activities of radon in all underground waters in the region is conducted. A range of 0.89-35.55 Bq/L was reported by Alabdula'aly for the well water radon level in the central region of Saudi Arabia. In that study, 77 wells were investigated, however only seven samples has a maximum concentration of more than 18 Bq/L[2].

TABLE-1
RADON ACTIVITY AND URANIUM CONCENTRATIONS

#	Sample location	Rn Activity (Bq/L)	U concentration (μg/L)	RSD
1	Usfan	0.6 ± 0.07	0.690	0.4
2	Hada Alsham	2.9 ± 0.30	0.580	2.1
3	Wadi Alaf	1.0 ± 0.10	2.640	0.2
4	Wadi Noman	1.6 ± 0.20	31.970	0.5
5	Wadi Garran	1.5 ± 0.10	1.860	0.4
6	Wadi Um-aljarm	3.9 ± 0.30	1.750	0.9
7	Wadi Faidah	1.0 ± 0.10	0.450	0.3
8	Hada Alshaam	2.3 ± 0.20	1.020	1.7
9	Hada Aalshaam	0.9 ± 0.10	0.250	3.4
10	Wadi Khaif	2.1 ± 0.20	1.390	0.4
11	Wadi Aljumoom	0.9 ± 0.10	5.497	0.6
12	Wadi Aljumoom	1.9 ± 0.20	2.920	0.3

Uranium concentrations are within the range 1.02-31.97 μg/L. The recommended limit of uranium for safe water set by the world health organization is 15 μg/L whereas the limit set by USEPA is 30 μg/L. Concentration of uranium in samples investigated in this study fall well within the lower limit of the safe use of water. Only one sample show a concentration of 31.97 μg/L, while all others has a concentration of less than 5.5 μg/L.

On converting the concentration of uranium from μg/L to absolute activity in the Bq/L units and comparing those with radon concentrations observed in water samples, one can see that radon is present in higher concentrations than uranium. The results also show no direct correlation between the two isotopes in water. One can explain the higher concentration of radon due to its gaseous nature and high solubility in water. Similar conclusions were made by Singh *et al.*[9] in their study on the estimation of uranium and radon concentrations in some drinking water samples.

Conclusion

For the 12 samples studied, the activity of radon lies in the range 0.6-3.9 Bq/L which is in the lower end of natural limits. Uranium concentrations lie in the range 1.02-31.97 μg/L. This range is also within the allowed limits set by various safety bodies. It is necessary to investigate a wide sample population in order to make a general conclusion about radon and uranium levels in the Makkah Al-Mukarramah area.

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