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Technical note

Radionuclides in hot mineral spring waters in Jordan

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Abstract

Hot mineral springs in Jordan are very attractive to people who seek physical healing but they are unaware of natural radioactive elements that may be contained in the hot mineral water. The activities of the natural radioactive isotopes were measured and the concentrations of the parents of their natural radioactive series were calculated. The measured radionuclides were ^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{228}Th, ^{212}Pb, ^{212}Bi and ^{208}Tl. In addition the activities of ^{235}U and ^{40}K were measured. The activities ranged from 0.14 to 34.8 Bq/l, while the concentrations of parent uranium and thorium isotopes ranged from 3.0×10^{-3} to 0.59 mg/l. The results were compared with those for drinking water.

Keywords: Gamma-ray; HPGe; Radionuclides; Activity; Concentration; Hot mineral water

1. Introduction

Springs of hot mineral water in Jordan are very attractive to the external and internal tourism industries. They have been exploited for their alleged healing powers. Visitors to the bath houses of these springs are unaware that they can breath ^{222}Rn emanated from the surrounding hot spring water or be exposed to hazardous radiation emitted from radionuclides found in the hot mineral water (Eisenbud, 1977). Naturally occurring radioactivity in drinking water and hot mineral water has

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increasingly been considered to constitute a major source of radiation exposure (Cothern, Lappenbusch & Michel, 1986); in general the most active γ-emitters produced by the decay series are ^{214}Bi and ^{214}Pb.

The average ^{238}U content in the earth's crust has been estimated to be 2.7 mg/kg and concentrations may be as high as 120 mg/kg in phosphate rocks (Padam, Rana, Azaam, Naqvi & Srivastava, 1996). Meanwhile, the average ^{232}Th content of the earth's crust is about 9.6 mg/kg (Table of Isotopes, 1996). Enhanced levels of uranium, thorium and their daughter products might be present in ground water in areas that are rich in natural radioactivity. The earth's crust in Jordan is covered by phosphate rocks that make the water sources rich in ^{238}U and its daughters which may be found dissolved or suspended in the water. The geology of the area indicates that it is covered by a sedimentary sequence with some interruption of sandstone that contains iron–uranium. The upper part of the sequence consists of phosphorite beds and limestone that contain ^{238}U minerals up to 100–200 mg/kg (Smith, Powell, Bradley, Gedeon & Armo, 1995).

Earlier studies conducted by the Ministry of Water and Irrigation and the Ministry of Energy and Mineral Resources in Jordan concentrated on measuring the natural radioactivity in drinking water at specified regions around the capital Amman (Gedeon, Armo, Jawawdeh & Kilani, 1995). Sheraiteh (1998) measured the natural radioactivity in water and sediments from hot springs. He used the degassing technique to measure the concentration of ^{222}Rn in the water samples. He found a poor correlation between ^{222}Rn concentration and the natural radioactivity in sediments.

In spring water the following radionuclides could be identified by γ-spectrometric measurements: ^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{228}Th, ^{212}Pb, ^{212}Bi, ^{208}Tl, ^{235}U and ^{40}K. Meanwhile, ^{238}U and ^{232}Th levels have been estimated by assuming secular equilibrium. Depending on the different solubilities, secular equilibrium might not be present in the mineral water. Nevertheless, it seems to be a useful approach to gain an idea on the level at which the parent nuclides are present in the spring water.

In this study, we are again interested in natural radioactivity. The concentrations of ^{238}U and ^{232}Th are derived from those of their daughter products, assuming equilibrium. The results are also compared with the concentrations of these isotopes in drinking water at Yarmouk University.

2. Experimental work

Samples of hot spring waters from five different sites in Jordan were collected directly from the springs in plastic bottles and closed tightly. Then, spectral analysis of the radionuclides was carried out using a γ-ray spectrometer with a high-purity germanium detector (HPGe) of high resolution. This advanced spectrometer consists of an HPGe detector of resolution 1.73 keV at 1.33 MeV connected via a Spectrum Master from EG&G to the multi-channel analyzer card (MCA) installed in a PC computer. The Spectrum Master provides a high-voltage power supply, a

computer-controlled amplifier and an analogue-to-digital converter (ADC). The detector is shielded from the background radiation by cylindrical lead walls. A software program called GammaVision was used to accumulate and analyze the data, with the help of the chosen gamma libraries that include over 300 nuclides and about 1900 gamma lines.

The system was calibrated for energy and efficiency. The energy calibration was carried out by acquiring a spectrum from radioactive standards of known energies like ^{137}Cs and ^{60}Co. For the efficiency calibration a multi-element Marinelli beaker standard of known activities was used. Finally, each sample was placed in a Marinelli beaker of the same size as that of the multi-element standard. Then each sample spectrum was acquired for 24 h. The decay-corrected activity of each nuclide found in the sample was calculated. Also, the measurement uncertainty and the minimum detectable activity (MDA) were reported.

3. Data analysis

The sites studied are at Ma'in, Himma, Al-Shona, Afra and Barbeita (see Fig. 1). They are all located on the eastern slopes of the Jordan rift valley. The heat flows from the interior of the earth to heat the rocks and the water in contact with them. The sites have different temperatures; this is believed to be due to the depths of origin of the

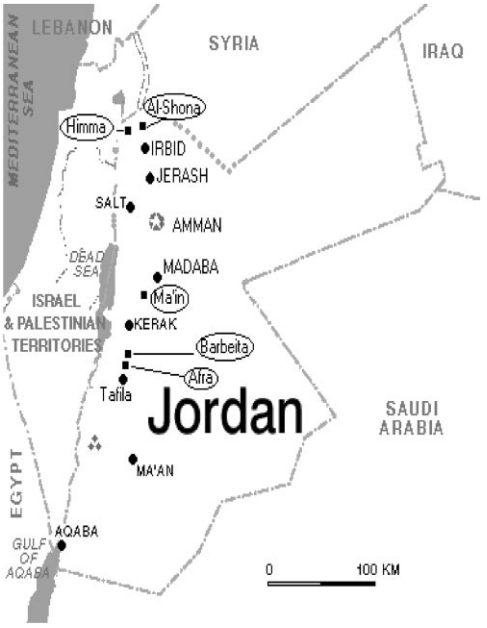

Fig. 1. Map of Jordan showing the locations of the studied sites.

drawn water at these sites. A typical spectrum of the observed radioisotopes is shown in Fig. 2.

The parents of the natural radioactive series (^{238}U, ^{235}U and ^{232}Th) have very long physical half-lives compared to their daughter products. This led us to an acceptable assumption of approximate secular equilibrium. In this case, the concentration of the parent nuclide could be calculated from the relation

$$N_p \lambda_p = N_d \lambda_d, \tag{1}$$

where N_p and λ_p are the number of atoms in a given sample and the decay constant of the parent, while N_d and λ_d are the number of atoms in the same sample and the decay constant of the daughter. From this relation the concentration of the parent can be calculated as

$$m = \frac{N_p}{A_{av}} A_p \tag{2}$$

where A_{av} is Avogadro's number (6.022×10^{23}) and A_p is the mass number of the parent nucleus.

The concentration of ^{238}U ($T_{1/2} = 1.41 \times 10^{17}$s) was calculated from the activity of ^{234}Th ($T_{1/2} = 24.1$ d). The activities of the two lines of ^{234}Th (92.8 and 92.3 keV) were summed and emission percentages of 2.72 and 2.69%, respectively, were used. This was done because the separation of the two lines was difficult and the full-width resolution at half-maximum (FWHM) of the pure germanium detector at this region was only 0.82 keV. Other daughter products such as ^{214}Bi and ^{214}Pb were not considered because of the break-up of the series due to the escape of ^{222}Rn gas. For comparison purposes, the concentration of ^{238}U calculated from ^{214}Pb or ^{214}Bi was less than that calculated from ^{234}Th at most sites studied.

To calculate the concentration of ^{234}Th ($T_{1/2} = 4.434 \times 10^{17}$s), the activity of the ^{228}Ac ($T_{1/2} = 6.15$ h) line at 911.21 keV with an emission percentage of 26.6%, was considered. In addition, the 968.97 keV line with an emission percentage of 17.4% was also used and the results averaged. The activity of ^{228}Ra was not measured because of the very long physical half-life ($T_{1/2} = 5.75$ yr) compared with that of ^{228}Ac.

In order to calculate the concentration of ^{235}U, the 185.71 keV line of emission percentage 53% was used. This line interferes with the ^{226}Ra line at 186.1 keV. The program used to analyze the data usually separated the two peaks by successive deconvolution. At two different sites it was found that the two lines from ^{235}U and ^{226}Ra were present with a large difference in their activities.

Corrections due to coincidence summing were not considered because a standard source of the same size and geometry to that of the sample under study was used to carry out the efficiency calibration. Random summing was also ignored because of the very low count rate and there was no pile-up or losses from the full energy peak. It was noted that the concentration of ^{232}Th calculated from the ^{208}Tl line at 510.77 keV was much higher than that calculated from the 583.19 keV ^{208}Tl line. This is attributed to

Table 1
Activities of the natural radionuclides in Bq/l

Nuclide	Drinking water	Ma'in			Himma		Afra	Barbeita	Al-Shona
		Family fall	Assail	Al-Amir	Spring	Makla			
^{234}Th	1.65 ± 0.54	2.72 ± 0.94	2.18 ± 0.75	2.42 ± 0.83	2.10 ± 0.73	3.34 ± 1.16	4.27 ± 1.49	2.85 ± 0.99	5.95 ± 2.07
^{226}Ra	—	—	—	3.80 ± 2.43	—	—	6.81 ± 2.97	4.54 ± 2.11	—
^{214}Pb	0.95 ± 0.42	1.12 ± 0.36	1.40 ± 0.40	1.36 ± 0.35	1.34 ± 0.40	1.11 ± 0.36	1.24 ± 0.38	0.83 ± 0.28	1.03 ± 0.60
^{214}Bi	1.57 ± 0.17	2.37 ± 0.23	2.39 ± 0.21	2.30 ± 0.20	2.38 ± 0.21	2.12 ± 0.21	2.45 ± 1.14	1.57 ± 0.19	2.92 ± 0.65
^{228}Ac	1.16 ± 0.28	2.37 ± 0.38	—	1.77 ± 0.33	1.81 ± 0.46	1.79 ± 0.47	2.06 ± 0.55	1.42 ± 0.30	1.67 ± 1.03
^{228}Th	12.18 ± 3.24	13.79 ± 3.52	—	—	—	—	—	—	—
^{212}Pb	0.74 ± 0.28	0.92 ± 0.28	1.07 ± 0.31	0.80 ± 0.30	0.91 ± 0.30	0.65 ± 0.29	11.1 ± 0.37	0.76 ± 0.28	—
^{208}Tl	0.22 ± 0.09	0.21 ± 0.06	0.26 ± 0.06	0.25 ± 0.06	0.25 ± 0.06	0.20 ± 0.06	0.21 ± 0.07	0.14 ± 0.05	0.30 ± 0.15
^{40}K	24.45 ± 2.21	—	—	—	—	—	34.80 ± 2.42	23.23 ± 1.73	—
^{235}U	0.26 ± 0.13	0.50 ± 0.13	0.35 ± 0.14	0.24 ± 0.13	0.41 ± 0.15	0.33 ± 0.13	—	—	—
^{212}Bi	1.45 ± 1.00	1.88 ± 1.29	—	1.27 ± 0.80	—	—	—	—	—
Total activity	44.64	25.88	7.64	14.2	9.21	9.54	52.97	35.34	11.87

Table 2
The concentrations of the natural radioactive parents in mg/l

Nuclide	Drinking water	Ma'in			Himma		Afra	Barbeita	Al-Shona
		Family fall	Assail	Al-Amir	Spring	Makla			
^{238}U	0.13 ± 0.04	0.22 ± 0.08	0.19 ± 0.06	0.19 ± 0.07	0.17 ± 0.06	0.27 ± 0.09	0.34 ± 0.17	0.23 ± 0.08	0.48 ± 0.17
^{235}U	0.0033 ± 0.0016	0.0062 ± 0.0017	0.0043 ± 0.0019	0.003 ± 0.0019	0.0052 ± 0.0019	0.0041 ± 0.0018	—	—	—
^{232}Th	0.29 ± 0.07	0.59 ± 0.10	0.26 ± 0.08	0.44 ± 0.08	0.45 ± 0.11	0.44 ± 0.12	0.51 ± 0.14	0.35 ± 0.08	0.41 ± 0.25
^{40}K	0.94 ± 0.01	—	—	—	—	—	1.34 ± 0.01	0.90 ± 0.01	—

coincidence summing with the background annihilation peak at 511 keV. Therefore, in this work, the activity of the 510.77 keV line was not considered (Debertin & Schotzig, 1979).

4. Results and discussion

Table 1 shows the radionuclide activities along with the counting uncertainties. It is noticed that the activity of ^{232}Th was dominant at all sites, except in Afra where it was not detected. It is also noticed that the activity of ^{214}Bi is higher than that of ^{214}Pb and that their ratio is almost constant at most sites. The activity of ^{214}Pb agrees well with the activity of ^{212}Bi within the uncertainty, considering the difference in their half-lives.

Table 2 shows the concentrations of the parent nuclides of the natural radioactive series in mg/l of untreated water. The concentration of ^{238}U was calculated from the activity of the ^{234}Th at all sites. The concentration varies from one site to another and from one position to another in the same spring. The highest concentration of ^{238}U was 0.48 mg/l in Al-Shona and the lowest was 0.17 mg/l in Himma–Spring. In Ma'in, it varies between 0.22 mg/l in Family Fall and 0.19 mg/l in Assail.

^{235}U was not detected at most sites. It was only detected in Ma'in and Himma. Its concentration, as given in Table 1, varies between 3.0×10^{-3} mg/l in Al-Amir Fall and 6.2×10^{-3} mg/l in Family Fall, both at Ma'in. At Himma, it varies between 4.1×10^{-3}

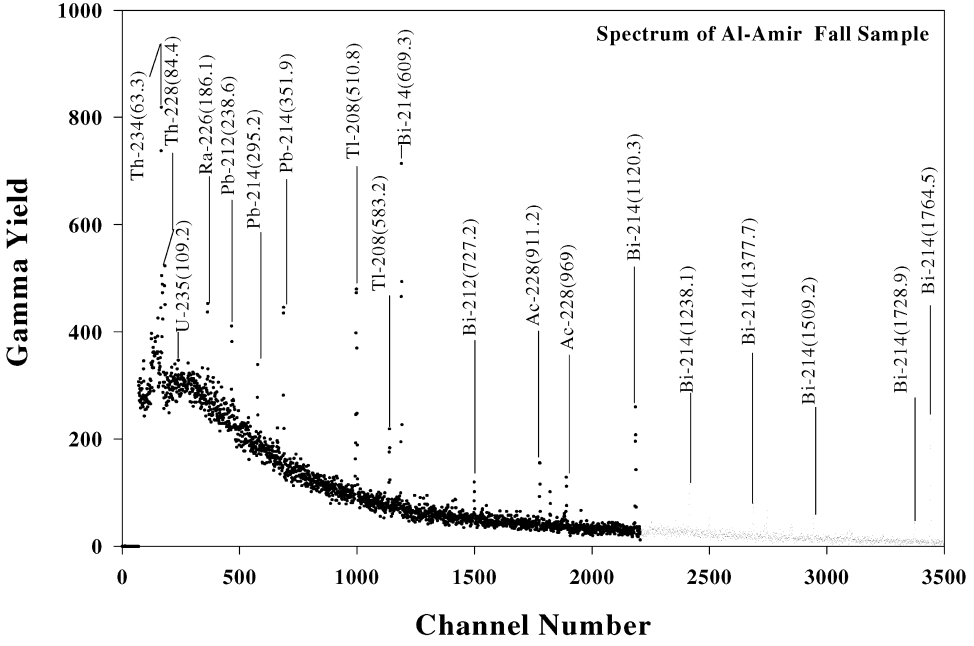

Fig. 2. A typical gamma spectrum.

and 5.2×10^{-3} mg/l (see Fig. 2). It can be noticed that the concentration ratio of $^{235}U/^{238}U$ is almost constant at 2–3%.

The concentration of the ^{232}Th calculated from the ^{228}Ac activity was found to vary between 0.26 mg/l in Assail and 0.59 mg/l in Family Fall. It was also noticed that the concentrations of ^{232}Th were higher than the concentrations of ^{238}U at all sites except Al-Shona. Other workers (Philip, Jaqueline & Whlard, 1982) have confirmed this.

Variations of the concentrations of ^{238}U, ^{235}U and ^{232}Th from one site to another or even from one position to another at the same site indicate that the origins of these hot waters are not the same and that they come from different depths and pass through different geological layers. As an example, the Barbeita and Afra sites are very close to each other (Fig. 1) and the surrounding earth's crust is similar, with granite rocks being the major component in this region. However, ^{226}Ra was found in Barbeita but not in Afra and the concentrations of other nuclides were not close.

^{40}K was detected at two sites only, Afra and Barbeita, and at low concentration, which is approximately the same as in drinking water from north Jordan (Table 2).

5. Conclusion

The concentrations of natural radioactive series nuclides differ widely at the studied sites. The differences are possibly due to different origins, depths and pathways of the outflowing water. In addition, the drinking water, which is mainly rainwater filtered through the phosphorite and granite geological layers of the catchment, shows approximately the same radionuclide content.

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References

Cothern, C., Lappenbusch, W., & Michel, J. (1986). Drinking-water contribution to natural background radiation. *Health Physics*, *50*(1), 33–47.

Debertin, K., & Schotzig, U. (1979). Coincidence summing corrections in Ge(Li) spectrometry at low source-to-detector distances. *Nuclear Instruments and Methods*, *158*, 471–477.

Eisenbud, M. (1977). Environmental radioactivity from natural industrial military sources (4th edition). New York: Academic Press.

Gedeon, R., Armo, H., Jawawdeh, J., & Kilani, S. (1995). Natural radioisotopes in ground water from Amman-Zarqa Basin, Jordan. Application of tracers in Arid Zone Hydrology. *Proceedings of the Vienna symposium*. 232 (pp. 81–97). Ontario: IAHS press.

Padam, S., Rana, N., Azaam, A., Naqvi, A., & Srivastava, D. (1996). Levels of uranium in waters from some Indian cities determined by fission track analysis. *Radiation Measurements*, *26*(5), 683–687.

Philip, T., Jaqueline, M., & Whlard, S. (1982). Ground Water Geochemistry of ^{228}Ra and ^{226}Ra and ^{222}Rn. *Geochimica et Cosmochimica Acta*, *46*, 1173–1182.

Sheraiteh, F. (1998). Studies of the environmental radioactivity in water. In *M.Sc. thesis*: Jordan University.

Smith, B., Powell, J., Bradley, A., Gedeon, R., & Armo, H. (1995). Naturally occurring uranium pollution in Jordanian mining. *Environmental Management*, *19*, 7–10.

Table of Isotopes. (1996) web page at: http://isotopes.lbl.gov/isotopes/toi.html