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In Egypt, nineteen water springs with temperatures exceeding 35 °C, which can be considered thermal waters, were reported. The radiological aspects of thermal water in three northern regions of Egypt (Gulf of Suez, Cairo, and Bahariya Oasis) were investigated. The activity concentrations of natural radionuclides of  $^{226}\text{Ra}$ ,  $^{40}\text{K}$ , total uranium and  $^{210}\text{Pb}$  ( $^{210}\text{Po}$ ) in water samples were determined using gamma spectrometry, laser fluorimetry and alpha spectrometry after chemical separation, respectively. The highest concentrations are detected in two locations, namely Sudr and Hammam Faroun springs, which are the hottest waters in Egypt (89 and 72 °C respectively). It seems that the presence of  $^{226}\text{Ra}$  is correlated to the water mineralization and the depth and the temperature of the reservoir's base. Also, these results seem evidence of uranium accumulation at depth in the mother bedrock and/or uranium immobilization due to uranium reduction to the immobile 4+-oxidation state in the reservoir base. The average activity concentrations of  $^{40}\text{K}$  are dependent on the water's origin, *i.e.* meteoric water mixed with sea water (Gulf of Suez springs) and meteoric water (Bahariya Oasis wells). There is no prescribed dose limit for short term and temporary exposure to thermal water. Workers at the thermal springs should be regularly monitored because of their continuous exposure to enhanced radiation levels over a long period of time. The hydrochemical characteristics, environmental isotopes aspects and water origin are briefly discussed based on published data.

## Introduction

Thermal and mineral water springs are considered to be important natural sources. Owing to their therapeutic and health-improving effects, some of them are widely popular for medical therapy, tourism, recreation, rehabilitation, and drinking.<sup>1</sup>

Chemical studies of thermal waters provide information regarding the type of geothermal reservoir (liquid or vapor dominated), reservoir base temperature, subsurface flow patterns and chemical quality. Liquid dominated geothermal systems are generally characterized by chloride rich waters and are associated with thermal springs discharging large volumes of water. Such systems occur in permeable rocks. Vapor dominated geothermal systems are characterized by acid, sulfate rich waters containing little chloride and are associated with thermal springs discharging low volumes of water. Such systems occur in impermeable rock.<sup>2</sup>

Swanberg reported nineteen water points with temperatures exceeding 35 °C which can thus be considered thermal water. The hottest waters are located along the east shore of the Gulf of Suez: Uyun Musa (48 °C) and Hammam Faroun (75 °C). Several artesian wells from the principle oases of the Western Desert (Dakhla, Kharga and Bahariya) have temperatures in the range 35–43 °C. In the Eastern Desert, there is only one thermal water well (Umm Khariga, 35.8 °C).<sup>2</sup>

Some of these waters have been used since the Pharaohs era for medical purposes.<sup>3</sup> The national authorities have renewed attention on developing tourist facilities, which are based on using the thermal waters for medical therapy, recreation and rehabilitation.

The main objective of this study is to shed more light on the radio-ecological characterization of thermal water in the northern regions of Egypt (namely the Gulf of Suez, Cairo and the Bahariya Oasis), Fig. 1, based on investigation of the natural radionuclide content in the studied water. Such research is essential to obtain more information for

physico-chemical characterization of thermo-mineral waters, planning and improving the development of the tourist facilities according to its planning applications (medical, therapy, recreation and/or drinking) and assessment of environmental impacts of this water on the surrounding region.

## Geological and hydro-geological setting

The surface exposures within the studied provinces are composed of sedimentary rocks, Fig. 2, the oldest of which

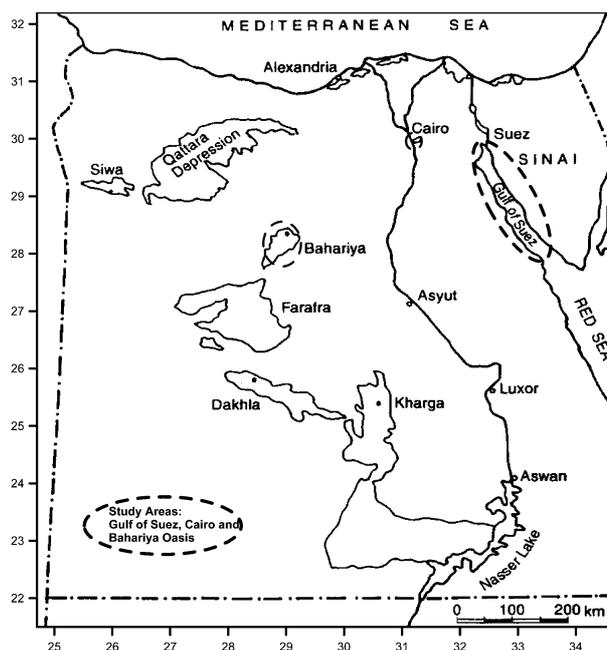


Fig. 1 Map of Egypt and sample location.<sup>22</sup>

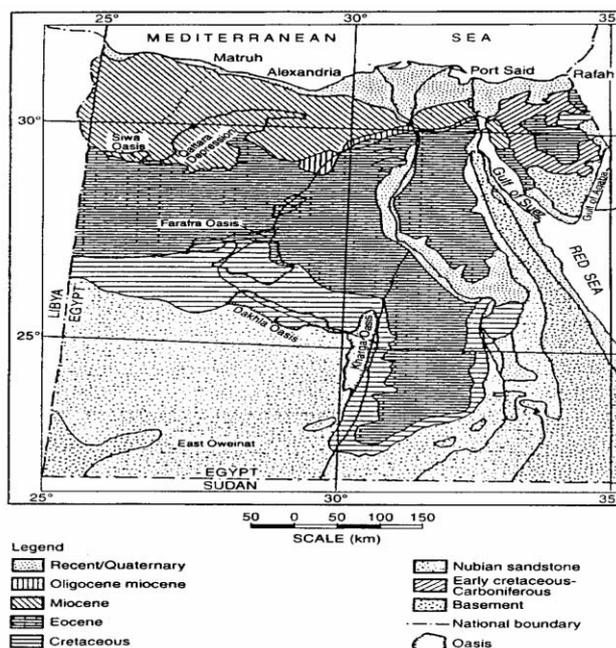


Fig. 2 Geological map of Egypt.<sup>22</sup>

belongs to the Cambrian period in the eastern side of the Gulf of Suez and Carboniferous in its western side,<sup>4</sup> Cretaceous in Cairo and vicinities<sup>9</sup> and Jurassic–Lower Cretaceous in the Western Desert Oases.<sup>5</sup> These are overlain by rock units covering all the ages till Quaternary with some zones of unconformity in the different regions. The rock units that belong to the ages from Oligocene to Pleistocene are dominated by clastic facies composed of sandstone, shales, conglomerates, evaporites and foraminiferal marls as well as alluvial deposits. On the other hand, the ones that extend from Cenomanian to Eocene in age are dominated by non clastic carbonate facies formed mainly of marls, chalks, limestones, dolomites and shales. The lowermost part of the succession (Carboniferous to Lower Cretaceous) is formed of a sandstones and shale complex known, particularly in its upper part, as the Nubian sandstone system.

The Paleozoic and Mesozoic section of the Nubian sandstone consists of alternating beds of sandstones and clay. The clay beds are laterally discontinuous and separate the sequence into a multilayered system that rests unconformably on the basement. The Nubian sandstone complex has groundwater aquifer properties and is capped by a series of marine deposits (limestones and shales) that belong to Upper Cretaceous through

Quaternary with zones of unconformity in between. These act as aquicludes to the underlying water bearing strata. The Upper Cretaceous, Eocene and Miocene rock units at some localities are strongly fissured and are developed into aquifers of importance. These are thought to be recharged through upward leakage from the underlying aquifer system which have higher static head than the overlying ones. The recharge of these aquifers is mostly through direct infiltration of precipitation that falls on the outcropped sections of the different aquifers as well as seepage from basin underflow from bordering fractured rocks. These aquifers also got recharge from meteoric water during Pleistocene–Early Holocene pluvial times where a very cool and humid climate was prevailing. The studied springs are surface manifestations of the mentioned groundwater aquifers where the level of the ascending fluids from deeper aquifers through conduit lineaments (faults, joints and fractures) meet the ground surface. The groundwater issues in El-Ain El-Sukhna, Ayun musa and Hammam Sidna Musa springs from faulted and jointed Miocene strata.<sup>4,6,7</sup> The Hammam Faroun spring issues from faulted dolomitic Eocene limestone at the shore cliff of Gabal Hammam Faroun.<sup>8</sup> The hydrothermal emanations near Cairo (e.g. Helwan spring) are usually associated with faulting of Cyclic trend NW–SE and Mediterranean E–W trends.<sup>9</sup> In the Western Desert Oases, the occurrence of springs is synonymous with the existence of the Oases themselves since the Oases are located in depressions where the ground surface is near to the regional water table. The studied thermal water in the El-Bahariya Oasis emerges from pre-Cenomanian water bearing sediments at more than 700 m depth.

## Experimental

### Sample collection

Water samples were collected from 11 locations in the three northern regions of Egypt (Gulf of Suez, Cairo and El Bahariya Oasis), Fig. 1. The water samples were collected directly from the spring's outlet in polyethylene containers. The pH and temperature of water samples were measured *in situ* using portable pH- and thermo-meters. Then the samples were transferred to the central laboratories in Cairo for analyses. The physico-chemical characteristics of the thermal waters are given in Table 1. The samples were acidified after being transported to the laboratory with concentrated nitric acid to pH value < 2.

### Gamma spectrometry

One liter volumes of the collected samples were transferred to Marinelli beakers and sealed for four weeks to ensure a secular

Table 1 Physico-chemical characteristics of some Egyptian thermo-mineral waters<sup>23</sup>

|   | Gulf of Suez                |               |          |                   |                 | Cairo             |                 | Bahariya Oasis |           |               |               |
|---|-----------------------------|---------------|----------|-------------------|-----------------|-------------------|-----------------|----------------|-----------|---------------|---------------|
|   | Ain Sukhna (1) <sup>a</sup> | Ayun Musa (2) | Sudr (1) | Hammam Faroun (3) | Hammam Musa (1) | Halwan Sulfer (1) | Ain El Sira (1) | Sigam (1)      | Halfa (1) | El Bishmu (1) | El Masara (1) |
| Source <sup>b</sup>                               | S                           | AW            | S        | S                 | S               | S                 | SL              | AW             | AW        | S             | AW            |
| pH  | 7.7                         | 7.9           | 6.5      | 7.4               | 8.2             | 7.3               | —               | 8.2            | 7.1       | 7.7           | 7.7           |
| Temp./°C  | 31.6                        | 37.1          | 89.0     | 72.0              | 48.0            | 28.9              | —               | 31.5           | 41.7      | 38.3          | 42.0          |
| °Base Temp./°C                                    | 138                         | 119           | 101      | 140               | 134             | 101               | —               | 80             | 67        | 88            | 79            |
| TDS/mg l <sup>-1</sup>                            | 7534                        | 2670          | 7296     | 14561             | 8730            | 6910              | —               | 217            | 191       | 172           | 198           |
| Na <sup>+</sup> /mg l <sup>-1</sup>               | 1897                        | 568           | 975      | 3840              | 1720            | 1219              | —               | 39             | 38        | 22            | 32            |
| K <sup>+</sup> /mg l <sup>-1</sup>                | 76                          | 41            | 30       | 102               | 71              | 23                | —               | 10             | 10        | 8             | 10            |
| Mg <sup>2+</sup> /mg l <sup>-1</sup>              | 241                         | 45            | 42       | 293               | 365             | 143               | —               | 15             | 13        | 13            | 10            |
| Ca <sup>2+</sup> /mg l <sup>-1</sup>              | 435                         | 251           | 390      | 1151              | 629             | 279               | —               | 12             | 9         | 10            | 13            |
| SO <sub>4</sub> <sup>2-</sup> /mg l <sup>-1</sup> | 136                         | 450           | 240      | 981               | 1183            | 797               | —               | 18             | 24        | 39            | 19            |
| Cl <sup>-</sup> /mg l <sup>-1</sup>               | 3671                        | 971           | 1751     | 8013              | 4130            | 1991              | —               | 47             | 67        | 23            | 21            |
| HCO <sub>3</sub> <sup>-</sup> /mg l <sup>-1</sup> | 214                         | 210           | 665      | 122               | 196             | 251               | —               | 71             | 37        | 44            | 98            |

<sup>a</sup>Number of samples. <sup>b</sup>S = spring, SL = Salt lake, AW = Artesian well. <sup>c</sup>The deep subsurface base-temperature of the reservoir to the last temperature of water–rock equilibrium, according to empirical cation geothermometers (Na/K/Ca).

equilibrium between  $^{226}\text{Ra}$  and its progenies. Radium-226 ( $^{238}\text{U}$ ) series,  $^{232}\text{Th}$  series and  $^{40}\text{K}$  activity concentrations were measured using gamma spectrometry based on hyper-pure germanium detectors with efficiencies of 30–40% and full width at half maximum (FWHM) of 1.95 keV for the  $^{60}\text{Co}$  gamma line at 1332 keV. The gamma spectrometers were calibrated using both a  $^{226}\text{Ra}$  point source and potassium chloride standard solutions in the same geometry as the sample. The relative efficiency curve was obtained using the most intensive gamma line of  $^{226}\text{Ra}$  in the energy range from 186 to 2450 keV. Then the relative efficiency curve was normalized to an absolute efficiency of the 1460.7 keV gamma line of  $^{40}\text{K}$  from a solution of chemically pure KCl. Finally the normalization coefficients ( $\text{Bq kg}^{-1}/\text{counts per second}$ ) of different gamma energy lines were obtained to determine the activity concentration in  $\text{Bq kg}^{-1}$ .<sup>12</sup> The gamma transmission used for activity calculations are 352.9 keV ( $^{214}\text{Pb}$ ), 609.3, 1120.3 and 1764.5 keV ( $^{214}\text{Bi}$ ) for  $^{226}\text{Ra}$  ( $^{238}\text{U}$ ) series, 338.4, 911.1 and 968.9 keV ( $^{228}\text{Ac}$ ) for  $^{232}\text{Th}$  series and 1460.7 keV for  $^{40}\text{K}$ . The lower limit of detection, with 95% confidence, for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are 0.28, 0.16 and 1.0  $\text{Bq kg}^{-1}$ , respectively for 20 h counting time and 1 l sample volume.<sup>10,11</sup>

### Alpha spectrometry

Lead-210 and  $^{210}\text{Po}$  are in equilibrium because the samples were collected at least two years ago. A one liter sample was spiked, for chemical recovery and activity calculation, with about 80  $\text{mBq } ^{208}\text{Po}$ , and evaporated to near dryness. A few ml of  $\text{HNO}_3$  were added and evaporated to near dryness. A few ml of  $\text{HCl}$  were added and evaporated to near dryness. Finally, the samples were dissolved in 30 ml 0.5 M  $\text{HCl}$ . Then, polonium was self-plated from the solution at temperatures between 80–90 °C onto a rotating stainless steel disk fixed in a Teflon disk holder.<sup>13</sup> The plated disks were counted with alpha spectrometers (CANBERRA, Mod. 7401 VR) based on passivated implanted planar silicon (PIPS) detectors of 450  $\text{mm}^2$  surface areas, about 20% efficiency and 17 keV full width at half maximum (FWHM) at the  $^{241}\text{Am}$  5.49 MeV energy line. The alpha spectrometers are coupled to low noise preamplifiers, amplifiers, and a multichannel analyser operating with Genie 2000 Software (CANBERRA). The samples were measured for

1000 min, applying a lower limit of detection of 1  $\text{mBq}$ .<sup>10</sup> The average chemical recovery was 75%, and the individual values ranged from 50 to 100%. The measured  $^{210}\text{Po}$  specific activity is equivalent to  $^{210}\text{Pb}$  specific activity.

### Laser fluorimetry

Total uranium analysis; total uranium determination was carried out on a solution containing five ml of the water sample and 2 ml FLURAN reagent (sodium pyro-phosphate, sodium di-hydrogen phosphate) using uranium analyzer model Sintrex UA-3. This technique is based on the fluorescence of the uranium complex formed with the FLURAN reagent, which converts the uranyl species into a single form that has a high luminescent yield. In the UA-3, a nitrogen laser tube provides UV pulse excitation fluorescence. The uranium concentration in ( $\text{ng ml}^{-1}$ ) was calculated from the formula:

$$U (\text{ng ml}^{-1}) = D_1 V_1 C / (D_2 - D_1) V_2 \quad (1)$$

Where:  $D_1$  is the fluorescence due to the uranium in the sample,  $D_2$  is the fluorescence due to the uranium in the standard (spiked),  $V_1$  is the volume of the sample (ml),  $V_2$  is the volume of the standard (ml), and  $C$  is the concentration of uranium standard solution ( $\text{ng ml}^{-1}$ ). The minimum detection limit of the system is 0.05  $\text{ng ml}^{-1}$  (0.63  $\text{mBq l}^{-1}$ ,  $^{238}\text{U}$ ) and the precision is about 15% at the 0.05  $\text{ng ml}^{-1}$  level. By using an internal standard spike, the quenching effect caused by interfering elements such as Fe, Mn, Cu, Ni, and organic matter was eliminated.<sup>14</sup>

### Results and discussion

It is well known that many thermal and mineral springs waters contain relatively high concentrations of some natural radio-nuclides such as  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$  and  $^{210}\text{Pb}$ .<sup>1,15,16,17</sup>

Activity concentrations of  $^{226}\text{Ra}$  and  $^{40}\text{K}$  ( $\text{Bq l}^{-1}$ ), total uranium (ppb),  $^{238}\text{U}$  ( $\text{mBq l}^{-1}$ ),  $^{210}\text{Pb}$  ( $\text{mBq l}^{-1}$ ), and  $^{226}\text{Ra}/^{238}\text{U}$ ,  $^{226}\text{Ra}/^{210}\text{Pb}$  and  $^{210}\text{Pb}/^{238}\text{U}$  activity ratios in some Egyptian thermal waters are given in Table 2. Activity concentrations ( $\text{Bq l}^{-1}$ ) of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{238}\text{U}$ ,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  in thermal and mineral waters in different countries are given in

**Table 2** Activity concentrations  $\pm$  error ( $1\sigma$ ) of  $^{226}\text{Ra}$  and  $^{40}\text{K}$  ( $\text{Bq l}^{-1}$ ), and total uranium (ppb),  $^{238}\text{U}^b$  ( $\text{mBq l}^{-1}$ ),  $^{210}\text{Pb}$  ( $\text{mBq l}^{-1}$ ),  $^{226}\text{Ra}/^{238}\text{U}$ ,  $^{226}\text{Ra}/^{210}\text{Pb}$  and  $^{210}\text{Pb}/^{238}\text{U}$  activity ratios in some Egyptian thermal waters

| Ser.                 | Code | Locations                      | Ra-226 $\pm$ E       | K-40 $\pm$ E        | $U_{\text{total}}$  | $^{238}\text{U}^b$  | $^{210}\text{Pb}$    | $^{226}\text{Ra}/^{238}\text{U}$ | $^{226}\text{Ra}/^{210}\text{Pb}$ | $^{210}\text{Pb}/^{238}\text{U}$ |
|----------------------|------|--------------------------------|----------------------|---------------------|---------------------|---------------------|----------------------|----------------------------------|-----------------------------------|----------------------------------|
| (I) Gulf of Suez     |      |                                |                      |                     |                     |                     |                      |                                  |                                   |                                  |
| 1                    | AS   | Ain Sukhna                     | <0.28                | $2.55 \pm 0.42$     | $4.02 \pm 0.48$     | $49.9 \pm 6.0$      | $2.97 \pm 0.52$      | —                                | —                                 | 0.06                             |
| 2                    | AM   | Ayun Musa <sup>a</sup>         | <0.28                | $2.78 \pm 0.27$     | $6.00 \pm 0.10$     | $74.4 \pm 1.2$      | $3.07 \pm 0.33$      | —                                | —                                 | 0.04                             |
| 3                    | SA   | Suder Spring                   | $3.94 \pm 0.91$      | $3.00 \pm 0.52$     | $7.26 \pm 0.56$     | $90.0 \pm 7.0$      | $26.1 \pm 2.06$      | 43.8                             | 151                               | 0.29                             |
| 4                    | HF   | Hammam Faroun <sup>a</sup>     | $3.33 \pm 1.30$      | $5.78 \pm 1.26$     | $6.00 \pm 0.10$     | $74.4 \pm 1.2$      | $20.15 \pm 1.23$     | 44.8                             | 165                               | 0.27                             |
| 5                    | HM   | Hammam Sidna Musa <sup>a</sup> | <0.28                | $3.15 \pm 0.31$     | $2.82 \pm 0.32$     | $35.0 \pm 4.0$      | —                    | —                                | —                                 | —                                |
|                      |      | Average (range)                | 3.64<br>(<0.28–3.94) | 3.45<br>(2.55–5.78) | 5.22<br>(2.82–7.26) | 64.7<br>(35.0–90.0) | 13.07<br>(2.97–26.1) | 44.3                             | 158                               | 0.17<br>(0.04–0.29)              |
| (II) Cairo           |      |                                |                      |                     |                     |                     |                      |                                  |                                   |                                  |
| 6                    | C 1  | Halwan Sulfer Spring           | <0.28                | $1.97 \pm 0.22$     | —                   | —                   | —                    | —                                | —                                 | —                                |
| 7                    | C 2  | Ain El Sira Spring             | <0.28                | $13.65 \pm 0.60$    | —                   | —                   | —                    | —                                | —                                 | —                                |
|                      |      | Average (range)                | <0.28                | $7.81 (1.97–13.65)$ | —                   | —                   | —                    | —                                | —                                 | —                                |
| (III) Bahariya Oasis |      |                                |                      |                     |                     |                     |                      |                                  |                                   |                                  |
| 8                    | B 1  | Sigam                          | <0.28                | <1                  | —                   | —                   | —                    | —                                | —                                 | —                                |
| 9                    | B 2  | Halfa                          | $0.77 \pm 0.29$      | <1                  | —                   | —                   | —                    | —                                | —                                 | —                                |
| 10                   | B 3  | El Bishmu                      | $0.63 \pm 0.24$      | <1                  | —                   | —                   | —                    | —                                | —                                 | —                                |
| 11                   | B 4  | El Masara                      | <0.28                | <1                  | —                   | —                   | —                    | —                                | —                                 | —                                |
|                      |      | Average (range)                | $0.70 (<0.28–0.77)$  | <1                  | —                   | —                   | —                    | —                                | —                                 | —                                |

<sup>a</sup>Average value. <sup>b</sup>1 ppm = 12.4  $\text{Bq l}^{-1}$ . — The uncertainties of the reported results are evaluated considering counting statistics and calibration error only.

**Table 3** Activity concentrations (Bq l<sup>-1</sup>) of <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>238</sup>U, <sup>210</sup>Pb and <sup>40</sup>K in thermal and mineral waters in different countries

| Country             | <sup>226</sup> Ra | <sup>228</sup> Ra | <sup>238</sup> U | <sup>210</sup> Pb | <sup>40</sup> K | Ref.       |
|---------------------|-------------------|-------------------|------------------|-------------------|-----------------|------------|
| Jordan              | 3.80–6.81         | 1.67–2.37         | 2.10–5.95        | —                 | 23.23–34.80     | 24         |
| Spain <sup>a</sup>  | <0.06–1.10        | —                 | <0.03–0.50       | —                 | —               | 25         |
| Croatia             | 0.07–4.40         | —                 | —                | —                 | —               | 1          |
| Brazil <sup>a</sup> | <0.01–0.22        | <0.01–0.75        | —                | <0.01–0.51        | —               | 26         |
| Brazil <sup>a</sup> | —                 | —                 | —                | <0.004–0.78       | —               | 27         |
| Tunisia             | 0.034–3.90        | —                 | 0.002–0.043      | —                 | <2.40–3.90      | 18         |
| Egypt               | <0.28–3.64        | <0.16             | 0.035–0.09       | 0.003–0.026       | <1.00–13.65     | This study |

<sup>a</sup>Mineral waters.

Table 3. Average concentrations of <sup>226</sup>Ra in thermal water samples in the Gulf of Suez, Cairo and the Bahariya Oasis regions are 3.64, <0.28 and 0.70 Bq l<sup>-1</sup> respectively, which generally match the published values.<sup>16, 18, 23</sup> The highest <sup>226</sup>Ra activity concentrations are found in the Gulf of Suez region springs and associated with the hottest spring water, Sudr (89 °C) and Hammam Faroun (72 °C) springs, 3.94 and 3.33 Bq l<sup>-1</sup> respectively. Published values of <sup>226</sup>Ra in mineral waters ranged to more than 3.7 Bq l<sup>-1</sup>, which is a thousand times greater than the values normally reported for public water supplies.<sup>16</sup> In Tunisian thermo-mineral springs, the activity concentrations of <sup>226</sup>Ra ranged between 0.034 and 3.9 Bq l<sup>-1</sup>.<sup>18</sup>

Total uranium, <sup>238</sup>U and <sup>210</sup>Pb average (range) concentrations in water samples from the Gulf of Suez region were 5.2 (2.8–7.3) ppb, 64.7 (35.0–90.0) mBq l<sup>-1</sup> and 13.1 (2.97–26.1) mBq l<sup>-1</sup> respectively. In conclusion, the uranium activity concentrations of the whole set of waters investigated are low. In Finland, for example, some waters have shown activities above 1.5 Bq l<sup>-1</sup> in uranium, which is due to high uranium concentration in the subterranean rocks. Among French springs, those of the Massif Central often contain higher levels of uranium. The most remarkable one is the Badoit water with 900 mBq l<sup>-1</sup> of <sup>238</sup>U and 2500 mBq l<sup>-1</sup> of <sup>234</sup>U. Thermal waters in the Gulf of Suez region are generally characterized by low uranium activities and relatively high <sup>226</sup>Ra activities. This is due to the elevated temperatures and the elevated concentration of chlorine. In springs, both radium and uranium distribution are affected by temperature, salinity and redox conditions.<sup>18</sup> Water from the hot springs of the east Pacific rise is depleted in uranium. Such depletion is probably caused by reduction of U<sup>6+</sup> to insoluble U<sup>4+</sup> during sea water circulation through the vent system. The total removal of uranium in association with high temperature hydrothermal circulation has been estimated. During sea water circulation, sea water loses virtually all its uranium, ~3 ppb. The global flux of water through the ridge hydrothermal system is 1.35 × 10<sup>17</sup> g y<sup>-1</sup> and the total uranium loss is then ~0.04 10<sup>10</sup> g y<sup>-1</sup>.<sup>17</sup> The depletion of <sup>210</sup>Pb in the investigated waters could be explained due to its particle reactive properties. Also, enhanced removal of particle reactive radionuclides such as <sup>210</sup>Pb, <sup>210</sup>Po, <sup>230</sup>Th and <sup>231</sup>Pa takes place during the precipitation of metalliferous hydrothermal deposits (formed primarily by the precipitation of iron and manganese oxyhydroxides).<sup>19</sup>

The average <sup>226</sup>R/<sup>238</sup>U, <sup>226</sup>R/<sup>210</sup>Pb and <sup>210</sup>Pb/<sup>238</sup>U activity ratios were 44.3, 158 and 0.17 respectively. The great difference in the chemical characteristics of uranium and radium, especially the important difference of solubility, imply that isotopic equilibrium is not often achieved in these waters.<sup>18</sup> Also, at 2 km depths, groundwaters are usually reducing and slightly acidic. They can dissolve barium, sulfur (as H<sub>2</sub>S), and radium, which are transported back to the surface where oxidation of S to SO<sub>4</sub> coprecipitates barium and radium.<sup>20</sup> Swanberg *et al.*<sup>2</sup> suggested that the thermal waters along the Gulf of Suez were heated by deep convective circulation along a regional geothermal gradient and were mixed with Red Sea

water during migration to the surface.<sup>3</sup> The convection geothermal model may explain the elevated <sup>226</sup>Ra, U and <sup>210</sup>Pb concentrations that are associated with the hottest water springs (Sudr and Hammam Faroun springs), in the Gulf of Suez region, where the deep circulated water comes in contact with gases and magmatic fluids that escape through fractures at the base of the reservoir. This finding is supported by the presence of some basaltic dikes in the Hammam Faroun block.<sup>21</sup> Also, these results seem evidence of uranium accumulation at depth in the mother bedrock and/or uranium reduction to the immobile 4+-oxidation state in the reservoir base where the soluble compounds of <sup>226</sup>Ra are leached by water circulation.<sup>16</sup> Hydrothermal fluids discharging from an oceanic spreading centre have undergone profound chemical changes with respect to sea water. These changes, resulting from interaction of sea water and basalt at high temperature, include the addition, removal and fractionation of radionuclides in the fluids and ultimately in deposits derived from the precipitation of minerals from the fluids.<sup>19</sup>

The maximum specific activities of <sup>226</sup>Ra in soil and shore sediment samples near the Hammam Faroun spring are highly elevated; up to 2997 and 368 Bq kg<sup>-1</sup> respectively, which are about 100 times higher than the normal levels.<sup>23</sup> Whereas uranium is mobile in oxidizing water, radium appears to be mobile in many natural waters. Both radium and uranium can be adsorbed by sediments but under different conditions. Anomalies with excess radium are very common around springs, seepages and hot springs, whereas uranium excess anomalies are usually found in sediments under reducing conditions. Radioactive deposits around springs were found to contain mostly <sup>226</sup>Ra with minor amounts of <sup>228</sup>Ra, <sup>238</sup>U and <sup>232</sup>Th.<sup>20</sup>

Wollenberg (1975) suggests that in Nevada, and perhaps in thermal springs elsewhere, the presence of <sup>226</sup>Ra and <sup>222</sup>Rn in the water is evidence of an accumulation of uranium at depth. The basis for his conclusion rests on a convection geothermal model: normal regional groundwaters are heated by rocks near the base of the system, flow upwards, and are replaced by more inflow from the outer water of the regional aquifer.<sup>3,16</sup>

Thorium-232 is not detectable (less than 0.16 Bq l<sup>-1</sup>) in these Egyptian thermal waters. Thorium is a particularly insoluble element in natural waters and is usually found associated with solids.<sup>18</sup>

The average activity concentrations of <sup>40</sup>K in the Gulf of Suez and Bahariya Oasis thermal waters are 3.45 and <1 Bq l<sup>-1</sup> respectively and are dependent on the origin of the water, *i.e.* meteoric water mixed with sea water (Gulf of Suez springs) and meteoric water (Bahariya Oasis wells). The maximum activity concentration of <sup>40</sup>K in the Gulf of Suez hot springs was 5.78 Bq l<sup>-1</sup>, while its maximum in Tunisian springs is 3.9 Bq l<sup>-1</sup>. Both values are low compared to results of studies on French spring waters, the spring of Preusdorf containing 13.7 Bq l<sup>-1</sup> of <sup>40</sup>K which is similar to sea water. Ain El Sira spring water, Cairo, is a salt lake water containing 13.7 Bq l<sup>-1</sup> <sup>40</sup>K.<sup>18</sup>

Activity concentration (Bq l<sup>-1</sup>) of <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>238</sup>U, <sup>210</sup>Pb

and  $^{40}\text{K}$  in thermal and mineral waters in different countries are given in Table 3.

The guideline activity concentration for  $^{226}\text{Ra}$  as recommended by the World Health Organisation (WHO) and International Atomic Energy Agency (IAEA) is  $1 \text{ Bq l}^{-1}$  as the maximum permissible concentration (MPC) for  $^{226}\text{Ra}$  in drinking water. There is no regulation for  $^{226}\text{Ra}$  occurring in other water categories, such as thermal and mineral waters, and to possible contributions by pathways of exposure other than ingestion. Mineral and thermal waters are not subject to the prescribed radiological dose limits because of the short term and temporary exposure during medical therapy, recreation and rehabilitation. Workers at the springs should be regularly monitored because of their continuous exposure to enhanced radiation over a long period of time.<sup>1</sup>

The annual effective dose values from drinking spring water are not representative of any actual exposures. The thermo-mineral waters considered here are not suitable for drinking water purposes due to the significant degree of mineralization, especially in the Gulf of Suez springs. We assumed the following scenario, the annual effective dose values due to ingestion of the measured radionuclides ( $^{226}\text{Ra}$ ,  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{210}\text{Pb}$ ) in one liter of the water, for the age group  $> 17$  y are calculated using the following equation:

$$D = \sum A_i F_i$$

Where:

$D$  = the annual effective dose/ $\mu\text{Sv y}^{-1}$ .

$A_i$  = the activity concentration of nuclide I/ $\text{Bq l}^{-1}$

$F_i$  = the effective dose per unit intake via ingestion/ $\mu\text{Sv y}^{-1} \text{ Bq}^{-1} \text{ l}$

( $F_{\text{Ra-226}} = 3.3 \times 10^{-7}$ ,  $F_{\text{K-40}} = 6.2 \times 10^{-9}$ ,  $F_{\text{U-238}} = 4.4 \times 10^{-8}$ ,  $F_{\text{Pb-210}} = 8.3 \times 10^{-7}$ )<sup>28</sup>

We have not considered the radionuclide concentrations, which are less than the minimum detectable activity of the measuring techniques for the dose calculations. The average annual effective dose values from drinking one liter of water were 1.24, 0.05 and 0.23  $\mu\text{Sv y}^{-1}$  for the Gulf of Suez, Cairo and Bahariya Oasis thermo-mineral water springs, respectively.

The mineralization is highly developed in the thermal water of the Gulf of Suez and Cairo regions, under the effects of the dissolution of marine limes and evaporite deposits, and the possible existence of residual old marine water. The thermal waters in the Bahariya Oasis on the other hand reflect the early stage of mineralization under the effect of meteoric water invasion and complete displacement of residual sedimentation water. The chemical compositions of the studied water samples are different from the assumed equilibrium water chemical compositions at the reservoir base. This disequilibrium occurs while fluid rises up due to the thermogenic regime.<sup>23</sup>

The isotopic composition of the studied thermal water is significantly depleted in  $^{18}\text{O}$  and D (deuterium) concentration relative to the possible recharging sources. This can be due to a contribution in the recharge from the latest pluvial times meteoric water. The isotopic depletion is lower in the Gulf of Suez and Cairo compared to Bahariya thermal water. This could be related to a recharge from recent water or due to deep thermogenic effects or remnants from old marine water.<sup>23</sup>

## Conclusion

Thermal water in Egypt owes its existence to natural upward flowing of deep groundwater through conduit lineaments in the rock strata. This generally takes place under normal geothermal gradient. The activity concentration of natural radionuclides in thermal waters varies in the studied regions. It depends on the water origin and chemistry, water-rock interaction and their chemical behavior. The highest  $^{226}\text{Ra}$ , U and  $^{210}\text{Pb}$  concentrations are associated with the highest water temperatures, in the Gulf of Suez region. It is highly

recommended to measure the radon gas concentration in thermal water and the radioactivity levels in the surrounding environment of the thermal water resources. Finally we shall be able to control the public and occupational radiation exposure due to utilities of the thermal water resources.

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