ENVIRONMENTAL RADIOACTIVITY MEASUREMENTS OF SOME EGYPTIAN SAND SAMPLES

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A long list of raw materials are brought from the desert and invested in feeding of many industries. The investigation of radionuclides distribution and radiation levels of such materials is important to assess the potential hazard of radiation exposure for those working in such industries. In this work the high-resolution gamma-ray spectroscopic system was used to measure the radionuclides content and to calculate some related parameters in three sand samples collected by a local company working in glass industry. Based on the characteristic spectral peaks, the radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K concentrations were determined. The activity concentrations for $^{238}$U were $91.51 \pm 4.94$, $111.47 \pm 6.01$ and $81.83 \pm 4.41$; for $^{232}$Th were $73.37 \pm 3.96$, $91.30 \pm 4.92$, $75.98 \pm 4.10$; and for $^{40}$K were $572.24 \pm 30.89$, $723.00 \pm 39.03$, and $807.13 \pm 43.57$ BqKg$^{-1}$. The calculated radium-equivalent activities were $(Ra_{eq}) = 240.48 \pm 12.98$, $297.69 \pm 16.07$ and $252.62 \pm 13.63$ BqKg$^{-1}$, which were less than permitted value (370 BqKg$^{-1}$). Also, the hazard index ($H_{in}$) for the 3 samples were $0.89 \pm 0.04$, $1.10 \pm 0.05$, and $0.90 \pm 0.04$. The total absorbed dose rates in air were $111.69 \pm 6.03$, $138.32 \pm 7.47$, and $118.63 \pm 6.41$ (nGyh$^{-1}$), whereas the external annual effective dose rates were $0.136 \pm 0.007$, $0.169 \pm 0.009$, and $0.145 \pm 0.007$ (mSv$^{-1}$). The radon exhalation rates, measured by passive nuclear track detectors (CR-39), were $0.26 \pm 0.014$, $0.33 \pm 0.017$, and $0.23 \pm 0.012$ Bqm$^{-2}$h$^{-1}$. These data records the radioactivity background levels in sand samples used for glass industry and could be used as reference information to assess any changes in the radioactive background level due to geological processes.

Key words: $^{238}$U, $^{232}$Th and $^{40}$K Estimation in sand, track detectors, radon.

1. INTRODUCTION

The natural radioactiv chains from $^{238}$U, $^{235}$U and $^{232}$Th produce a group of radionuclides with a wide range of half-life times. Most of the radioisotopes are alpha-emitters, so when they are ingested or inhaled, they significantly contribute to the radiation dose that people receive [1]. On the other hand, taking into both uranium and thorium are always present in soil, their gamma radiation causes external exposures with the consequent absorbed doses. Radon gas accumulate in indoor air, where the
average dose that many populations receive from radon inhalation is half that from all
natural causes [2].

The earth contains numerous radioactive elements, the origin, for part of them,
dates back to the formation of our world, while others are continuously produced
through nuclear reactions in the universe [3]. Among the former elements, the most
abundant are potassium-40 and the radioisotopes of the natural series of uranium,
actinium and thorium including the parent nuclei $^{235}\text{U}$, $^{238}\text{U}$ and $^{232}\text{Th}$ (Table 1) and
the decay products from the successive alpha or beta decays [4].

Table 1

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Half-life ($10^9$ years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}\text{U}$</td>
<td>4.47</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>0.70</td>
</tr>
<tr>
<td>$^{232}\text{Th}$</td>
<td>14.1</td>
</tr>
<tr>
<td>$^{40}\text{K}$</td>
<td>1.28</td>
</tr>
</tbody>
</table>

Building and industrial materials, which are brought from the deserts like
sands, also contribute to environmental radioactivity in two ways. First, by gamma
radiation mainly $^{226}\text{Ra}$, $^{232}\text{Th}$, $^{40}\text{K}$ and their progenies to a whole body dose and in
some cases by beta radiation to a skin dose, and secondly by releasing the noble gas
radon, its radioactive daughters, which are deposited in the human respiratory tract
[5]. Enhanced or elevated levels of natural radionuclides in such materials may cause
doses in the order of several mSv y$^{-1}$ [6]. The measurement of natural radioactivity
due to gamma rays from the dose rate is needed to implement precautionary
measures whenever the dose is found to be above the recommended limits [7].

In the work presented here, the high-purity germanium (HPGe) detector was
employed to determine the natural radioactivity of $^{40}\text{K}$, $^{208}\text{Pb}$, $^{214}\text{Bi}$, and $^{228}\text{Ac}$
in different sand samples (collected from Gabal El Khashab, east Cairo and from
Sinai, Egypt) and invested in glass and crystals industry and used as a building
material as well. Also, the solid state nuclear track detector (SSNTD) was
employed to measure the radon gas in these samples.

2. EXPERIMENTAL

SAND SAMPLES

Sand samples were collected from Gabal El Khashab near Maadi, Cairo and
from Sinai, Egypt. About 2 kg from each sample were sieved to about 100 mesh,
then dried at 100°C for 48h. Weighted samples were placed in polyethylene bottles
of 350 cm$^3$ volume and the bottles were completely sealed for 4 weeks to allow
GAMMA-RAY MEASUREMENTS

The spectrometer used for detection consisted of n-type HPGe coaxial detector of 17% efficiency having a resolution of 1.75 keV at 1.33 Mev and coupled to 4096 channel multi-channel analyzer (MCA). The detector was surrounded with a lead shielding to reduce the background radiation entering it. The schematic diagram of the gamma spectrometry used is shown in the Fig. 1.

![Diagram of gamma spectrometry system](image)

Fig. 1. – The typical gamma spectroscopy system used.

Gamma transitions of 1.460 Mev for $^{40}$K, 0.295 Mev & 0.352 Mev for $^{214}$Pb, 0.609 Mev, 1.12 Mev & 1.76 Mev for $^{214}$Bi, (were used to determine the $^{238}$U concentration), 0.911 Mev for $^{228}$Ac and 2.61 Mev for $^{208}$Tl were used for the laboratory measurement of activity concentrations. The radioactivity concentration in the investigated samples was obtained as follows:

$$A = \frac{(\text{cps})_{\text{net}}}{I \times E_{\gamma} \times m}$$

where $A$ is the activity concentration in Bq/kg, $(\text{cps})_{\text{net}}$ is the (count per second) and equal $(\text{cps})_{\text{sample}} - (\text{cps})_{\text{background}}$, $I$ is the intensity of the $\gamma$-line in a radionuclide, $E_{\gamma}$ is the measured efficiency for each $\gamma$-line observed and $m$ is the mass of the sample in kilograms.

The chemical composition of samples investigated were obtained from the Sinai Manganese company and shown in table (2), where SiO$_2$ represents the main constituent of the samples (ranges from 99 to 99.9%). The concentrations (in percent of weight) of other oxides Fe$_2$O$_3$, Cr$_2$O$_3$, TiO$_2$, L.O.1, Al$_2$O$_3$, CaO and MgO are listed.
Table 2

Chemical composition (and concentrations) of investigated samples

<table>
<thead>
<tr>
<th>Chemical Composition</th>
<th>Type (1) %</th>
<th>Type (2) %</th>
<th>Type (3) %</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>99.6–99.8</td>
<td>99.5–99.7</td>
<td>99–99.5</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>0.013–0.018</td>
<td>0.02–0.03</td>
<td>0.08–0.1</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>Max</td>
<td>0.0004 Max</td>
<td>0.001 Max</td>
</tr>
<tr>
<td>TiO₂</td>
<td>Traces</td>
<td>Traces</td>
<td>Traces</td>
</tr>
<tr>
<td>L.O.1</td>
<td>0.1–0.2</td>
<td>0.1–0.2</td>
<td>0.3–0.5</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>0.1–0.28</td>
<td>0.2–0.4</td>
<td>0.3–0.6</td>
</tr>
<tr>
<td>CaO</td>
<td>0.02–0.05</td>
<td>0.05–0.15</td>
<td>0.1–0.2</td>
</tr>
<tr>
<td>MgO</td>
<td>0.002–0.005</td>
<td>0.004–0.006</td>
<td>1.05–0.1</td>
</tr>
</tbody>
</table>

RADON MEASUREMENTS

The concentrations of radon and its progeny were measured in different sand samples using solid state nuclear track detector (SSNTD) (Inter cast, Italy). This detector is thin sheets of dielectric materials such as cellulose nitrate (CN) and polycarbonates. They are sensitive to alpha but not beta and gamma radiations. Also, it is not affected by moderate humidity, heat and light. The CR-39 detector used in this work is 500 µm thick. Its sensitivity for ionizing radiation is higher than other track-recording solid and can record all alpha energies without threshold [8]. Each sample was placed in a glass cylindrical chamber with 3.5 cm radius and 10 cm length. Dosimeters were prepared by putting two CR-39 detectors in the bottom of the chamber cover. The cylindrical container was sealed and the samples were stored for at least 30 days to establish the equilibrium between radium and radon before the start of each experiment. The CR-39 films were then exposed to radon and its progeny in the chamber for 8 weeks after which they were etched and the tracks were counted and the concentrations of both radon and its progeny were determined according to Hafez et al. (2000), [9]. The optimum etching conditions were obtained using 6.25 N NaOH etching solution at 70°C for 6 hours. The resulting alpha tracks were counted using an optical microscope at magnification of 400X and the track density (in Tcm⁻²) was recorded for each sample.

The measurement of the exhalation rate of radon is of great interest. The most important isotope of radon is ²²²Rn, which belongs to the ²³⁸U series, since its short-lived progenies are the major contributor to lung dose. The exhalation rate of ²²²Rn EX from different sand samples was calculated from the relation:

\[ E_X = CV\lambda/ A [T + 1/\lambda (e^{-\lambda T} – 1)] \]

Where \( C \) is the radon concentration (in Bqm⁻³), \( V \) is the effective volume of the can in m³, \( \lambda \) is the decay constant (in h⁻¹), \( A \) is the area of the can in m² and \( T \) is the exposure time in hours [10].
3. RESULTS AND DISCUSSION

Fig. 2 shows a typical gamma-ray spectrum obtained by the HPGe detector for different sand samples. The accumulation time for each sample was 24h to obtain the gamma-spectrum with good statistics.

The natural radioactivity in different sand samples is presented in Table 3. The maximum $^{238}\text{U}$ and $^{232}\text{Th}$ series concentrations appeared in type 2 and the minimum $^{238}\text{U}$ and $^{232}\text{Th}$ appeared in type 3 and type 1, respectively. The $^{40}\text{K}$ had the highest concentrations compared to other elements investigated. The concentrations of $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$ ranged from 81.83 to 111.47; 73.37 to 91.30; and 572.24 to 807.13 Bq kg$^{-1}$, respectively.

The activity concentration of detected radioisotopes in different sand samples is illustrated in Figure 3. The content of $^{40}\text{K}$ in type 3 are higher than other but the concentrations of radioisotopes $^{208}\text{Tl}$, $^{214}\text{Bi}$, $^{214}\text{Pb}$ and $^{228}\text{Ac}$ were higher in type 2 than other (74.83, 106.11, 116.84 and 107.78 Bq kg$^{-1}$), respectively. So the concentrations of $^{232}\text{Th}$ and $^{238}\text{U}$ are higher in type 2 than other samples.

The distribution of $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in different sand samples was not uniform (the inter variability of sample contents is shown in Table 4). With respect to exposure to radiation, the radioactivity has been defined in terms of radium equivalent activity $\text{Ra}_{\text{eq}}$ in Bq kg$^{-1}$ to compare the specific activity of materials containing different amounts of $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$. It was calculated through the following relation [11].

$$\text{Ra}_{\text{eq}} = C_{\text{Ra}} + 1.43 C_{\text{Th}} + 0.077 C_{\text{K}}$$

Where $C_{\text{Ra}}$, $C_{\text{Th}}$, and $C_{\text{K}}$ are the activity concentrations in Bq kg$^{-1}$ of $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$, respectively. It may be noted that $^{238}\text{U}$ has been replaced with decay product $^{226}\text{Ra}$ because there may be a dis-equilibrium between $^{238}\text{U}$ and $^{226}\text{Ra}$. While defining $\text{Ra}_{\text{eq}}$ activity according to the above equation, it has been assumed that 10 Bq of $^{226}\text{Ra}$, 7 Bq of $^{232}\text{Th}$, and 130 Bq of $^{40}\text{K}$ produce the same gamma doses [12]. The $\text{Ra}_{\text{eq}}$ is related to the external gamma dose and the internal dose due to radon and its daughters [13]. In the present work, $\text{Ra}_{\text{eq}}$ was estimated for the investigated samples and are given in Table (3).

### Table 3
Activities of U, Th, and K (in Bq kg$^{-1}$), as well as the calculated Ra equivalent (Ra<sub>eq</sub>) in different sand samples

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>$^{238}\text{U}$ (Bq kg$^{-1}$)</th>
<th>$^{232}\text{Th}$ (Bq kg$^{-1}$)</th>
<th>$^{40}\text{K}$ (Bq kg$^{-1}$)</th>
<th>$\text{Ra}_{\text{eq}}$ (Bq kg$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>91.51 ± 4.94</td>
<td>73.37 ± 3.96</td>
<td>572.24 ± 30.89</td>
<td>240.48 ± 12.98</td>
</tr>
<tr>
<td>2</td>
<td>111.47 ± 6.01</td>
<td>91.30 ± 4.92</td>
<td>723.00 ± 39.03</td>
<td>297.69 ± 16.07</td>
</tr>
<tr>
<td>3</td>
<td>81.83 ± 4.41</td>
<td>75.98 ± 4.10</td>
<td>807.13 ± 43.57</td>
<td>252.62 ± 13.63</td>
</tr>
</tbody>
</table>
Fig. 2. – Portion of gamma-ray spectrum from 600 to 1140 and 1700 to 2240 channel.
Table 4

Multiple comparison test (ANOVA) of 238U, 232Th and 40K contents in different samples

<table>
<thead>
<tr>
<th>Element</th>
<th>Samples</th>
<th>S1</th>
<th>S2</th>
<th>S3</th>
</tr>
</thead>
<tbody>
<tr>
<td>U238</td>
<td>S1</td>
<td>--</td>
<td>**</td>
<td>ns</td>
</tr>
<tr>
<td>Th232</td>
<td>S1</td>
<td>--</td>
<td>**</td>
<td>ns</td>
</tr>
<tr>
<td>K40</td>
<td>S1</td>
<td>--</td>
<td>**</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>S2</td>
<td>--</td>
<td>**</td>
<td>ns</td>
</tr>
</tbody>
</table>

(*,**,***, ns): indicate significant, highly, extremely and none significant differences between samples, respectively.

![Graph of radioisotopes](image)

**Fig. 3.** – Radioisotopes in different sand samples.

The average values of $R_{aq}$ were found to be 240.48, 297.69, 252.62 Bq kg$^{-1}$ for type 1, type 2 and type 3, respectively.

The external hazard index $H_{ex}$ was calculated for the investigated samples using the model proposed by [14] assuming infinitely thick walls without windows and doors, where the external hazard index is given by

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \leq 1$$

Where $C_{Ra}$, $C_{Th}$, and $C_{K}$ are the activity concentrations of uranium, thorium, and potassium in Bq kg$^{-1}$, respectively. The value of this index must be less than unity inorder to keep the radiation hazard be insignificant. The calculated external hazard is lower than unity, listed in table (5).
External hazard index (Hex), internal hazard index (Hin), exhalation rate (Ex), absorbed dose and effective dose in different sand samples

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Hex (Bqm⁻² h⁻¹)</th>
<th>Hin (Bqm⁻² h⁻¹)</th>
<th>Ex (Bqm⁻² h⁻¹)</th>
<th>Absorbed dose (ngy⁻¹)</th>
<th>Effective dose (mSvy⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.646 ± 0.034</td>
<td>0.895 ± 0.04</td>
<td>0.26 ± 0.014</td>
<td>111.69 ± 6.03</td>
<td>0.136 ± 0.007</td>
</tr>
<tr>
<td>2</td>
<td>0.803 ± 0.04</td>
<td>1.104 ± 0.05</td>
<td>0.33 ± 0.017</td>
<td>138.32 ± 7.47</td>
<td>0.169 ± 0.009</td>
</tr>
<tr>
<td>3</td>
<td>0.681 ± 0.056</td>
<td>0.90 ± 0.04</td>
<td>0.23 ± 0.012</td>
<td>118.63 ± 6.41</td>
<td>0.145 ± 0.007</td>
</tr>
</tbody>
</table>

The internal exposure to ²²²Rn and its radioactive progeny is controlled by the internal hazard index (H_in) which is given by [5].

\[ H_{\text{in}} = \frac{C_{\text{Ra}}}{185} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810} \]

For the safe use of a sand in the construction of dwellings, index (H_in) should be less than unity [15]. But the values of H_in in type 2 and type 3 are higher than unity. The average calculated values of (H_in) for different sand samples are 0.89, 1.10 and 0.90 for type 1, type 2 and type 3 respectively as shown in table 5. The radon concentration and exhalation rate are shown in table 6.

![Fig. 4. – Radon exhalation rate versus ²³⁸U concentration for the different sand samples.](image)

**Table 6**

Radon concentration (Bqm⁻³) and exhalation rate (Bqm⁻²h⁻¹)

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Rn con.</th>
<th>Ex</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4383</td>
<td>0.23</td>
</tr>
<tr>
<td>2</td>
<td>4961</td>
<td>0.26</td>
</tr>
<tr>
<td>3</td>
<td>6297</td>
<td>0.33</td>
</tr>
</tbody>
</table>
Environmental radioactivity measurements

The plot of the exhalation rate measurements (Fig. 5) showed a linear relationship with $^{238}\text{U}$ concentration for different sand samples. Fig. 6 shows the average calculated exhalation rate and radium equivalent for the investigated samples. This figure shows that the maximum exhalation rate is observed in type 2, which is characterized by high concentration of uranium. However, $\text{Raeq}$ is high in type 2 which are characterized by high concentration of $^{232}\text{Th}$.

![Fig. 5. – Variation of the average exhalation rate and average Ra equivalent activity in different sand samples.](image)

**MEASUREMENT OF ABSORBED GAMMA DOSE RATE**

The absorbed gamma dose rate in air 1m above the ground surface for the uniform distribution of radionuclides ($^{232}\text{Th}$, $^{238}\text{U}$, and $^{40}\text{K}$) was computed on the basis of guidelines provided by UNSCEAR [16, 17]. The conversion factors used to compute absorbed gamma dose rate ($D$) in air per unit activity concentration in (1Bqkg$^{-1}$) sand correspond to 0.621 nGyh$^{-1}$ for $^{232}\text{Th}$, 0.462nGyh$^{-1}$ for $^{238}\text{U}$, and 0.0417nGyh$^{-1}$ for $^{40}\text{K}$ [18].

$$D = (0.621C_{\text{Th}} + 0.462C_{\text{U}} + 0.0417C_{\text{K}})\text{nGyh}^{-1}$$

Where $C_{\text{Th}}$, $C_{\text{U}}$ and $C_{\text{K}}$ represent the average activity concentrations of $^{232}\text{Th}$, $^{238}\text{U}$ and $^{40}\text{K}$ in Bqkg$^{-1}$, respectively. The absorbed dose rates for the samples under investigation are listed in table 4. The dose rate at sand samples were found to be 111.69 ± 6.03, 138.32 ± 7.47, and 118.63 ± 6.41 nGyh$^{-1}$ for type 1, type 2, and type 3 respectively. Which is higher than international recommended value 55 nGyh$^{-1}$ [19].

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7 svGy$^{-1}$) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) [17] were used. The effective dose rate in
units of msv per year was calculated according to Arafa, (2004) [20] using the following formula.

\[
\text{Effective dose rate (msv}^{-1}) = \text{Dose rate (nGy h}^{-1}) \times 8760h \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6}
\]

The obtained values of effective dose effective dose rate are listed in table 4. The values of effective dose were found to be 0.136 ± 0.007, 0.169 ± 0.009 and 0.145 ± 0.007 mSv y\(^{-1}\) for type 1, type 2, and type3, respectively.

4. CONCLUSION

The levels of natural radioactivity in different sand samples collected from Gabal El khashab near Maadi, Sina, Egypt were determined using HPGe coaxial detector. Radon exhalation rate measured using solid state nuclear track detectors. The measurements of radon exhalation rate is good indicator for the uranium present in the sand samples. The data obtained showed that the radon exhalation rate varies linearly with uranium concentration.

REFERENCES
