A Study of the Distribution of Natural Radionuclides and their Environmental Impacts in Halayeb Triangle Area, along the South Eastern Coast of the Red Sea, Egypt

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ABSTRACT

Thirty samples (14 surficial soil, 3 Sea water – 3 desalinated water, and 10 plant) were collected from halayeb triangle area along the south eastern Coast of the Red Sea, Egypt. Distributions of natural radionuclides of $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs in these samples were determined using γ-ray spectrometry. The average activities of natural radionuclides in all samples in the study area are 19.38 (3.35–61.09) Bq kg$^{-1}$ for $^{226}$Ra, 10.87 (1.39–24.28) Bq kg$^{-1}$ for $^{232}$Th, 245.25 (132.46–531.19) Bq kg$^{-1}$ for $^{40}$K, and 1.48 (0.67-2.52) Bq kg$^{-1}$ $^{137}$Cs for surficial soil samples. These values were 8.59 (1.48 – 21.57) Bq kg$^{-1}$ for $^{226}$Ra, 11.34 (2.64–25.44) Bq kg$^{-1}$ for $^{232}$Th, and 254.66 (69.73–450.58) Bq kg$^{-1}$ for $^{40}$K, for plant samples. For sea water samples, the average activity for $^{226}$Ra is 3.20 (1.99 – 3.91) Bq kg$^{-1}$, for $^{232}$Th is 1.73 (1.24 – 2.45) Bq kg$^{-1}$, and 15.10 (8.67–23.25) Bq kg$^{-1}$ for $^{40}$K. For desalinized water the average activity is under detection limits for $^{226}$Ra, $^{232}$Th, and 13.49 (8.67–17.85) Bq kg$^{-1}$ for $^{40}$K. A comparison of radionuclide activities in the study area and in other coastal and aquatic environments is given. The radiation hazard parameters (absorbed dose rate, radium equivalent activity, internal hazard index, external hazard index, annual gonadal dose equivalent); and transfer factor of elements from soil to plant were calculated. The results of measurements will serve as base line data and background reference level for south Egyptian coastlines.

Keywords: Surficial soil/ water/ plant/ Natural radionuclides/ Red Sea coast Egypt

INTRODUCTION

Natural background radiation is the combined radiation field produced by the primordial and cosmogenic radioactive materials that are around us plus cosmic radiation from space. Everyone is exposed to this background radiation all through his life, at levels that depend on the ambient concentration of those radioactive materials and the altitude at which we live. This background radiation is the major source of radiation exposure to humans. Natural background dose rates are frequently used as a standard of comparison for doses from various man-made sources of radiation and also from the production, testing and use of nuclear weapons.

Radioactivity monitoring in the marine environment is normally based on analyses of specific nuclides in seawater, suspended particulate matter and sediments. The knowledge of the concentrations and distributions of natural radionuclides is of interest since it provides useful information in the monitoring of environmental contamination by natural radioactivity. Yi et al., 2009.

All uses of natural or man-made radionuclides require an understanding of their environmental behavior. Such knowledge is needed for their effective application for estimation of human health risks.

Site Description

The investigated area (Halayeb triangle) belongs to the Red Sea governorate located between the Nile River and the Red Sea in the southeast of Egypt. It is situated between latitude 23° 10’ N in the
north at Shalateen city, latitude 22º00'N in the south at the border with Sudan, the Red sea in the east, and the Red Sea Hills in the west as shown in Fig (1). The area runs parallel to the coast for about 250 km. It is also occupied from the western side by basement rocks, Nubian sandstone outcrops close to water divide. Tertiary volcanic rocks are extruded at the foot slope of the Red Sea mountainous shield. Isolated patches of Miocene sediments outcrops due to its location at the west of Abu-Ramad and Halayeb area. Alternating limestone and marl formation, Ismail, Y. L., 2005.

The study area is a promising district for tourism, herding (camel and sheep), fishing, mining works and agricultural development. Also, it has natural preserves, rare kinds of animals and birds. Its shores have huge fishing investment potentials. Also its farms which is depending on deep and rain water supplies, the northern limits at Gebel-Elba, made it a unique region among Egypt and North African ecosystems. There is also a dense cover of acacias, mangroves and other shrubs, in addition to endemic species of plants, Peter & Stephen Goodman 1996.

The current work may come as a complementary scientific activity adding radiological data to the study area. The scope of the work included the collection of surficial soil, sea water, desalinized water and plant samples and radiological calculations including radium equivalent activity (Ra$_{eq}$), absorbed dose rate (D), internal hazard index (H$_{in}$), the external hazard index (H$_{ex}$) and the annual gonadal dose equivalent (AGDE) also, the transfer factor of the radioactive elements from soil to plant were calculated based on the obtained specific activities results. These findings are used to evaluate the distribution of radioactive elements and its environmental impacts, these data could be used as a reference data for future investigations.

**Fig. (1): Location of the Study area (Halayeb triangle)**

**Instrumentation**

Thirty samples (14 surficial soil, 3 Sea water, 3 desalinated water and 10 plant samples) were collected from different locations throughout Halayeb triangle. The surficial soil samples were collected using a template of 25x25 cm$^2$ area and 5 cm depth, the gravels size greater than 2 cm were discarded USDOE, 1992. Plant samples were collected in the same site of surficial soil samples and sea water samples of 5 liters of sea water and desalinated water samples were collected in polyethylene containers. The surficial soil and plant were crushed, sieved by a 0.8 mesh sieve, homogenized, mixed and weighed. The samples were transferred to Marinelli beakers (100 ml) for gamma activity analysis. Each sample was carefully sealed for 4 weeks to reach secular equilibrium between $^{220}$Rn and $^{232}$Th and their respective progeny Mollah et al., 1987.
Spectra for different samples were measured with a high-resolution gamma-ray spectrometer. To reduce gamma-ray background, the detector was shielded by a lead cylinder with a fixed bottom and a moveable cover, and a concentric copper cylinder. Surficial soil and plant samples were measured at the Karlsruher Institut für Technologie (KIT), Eggenstein Leopoldshafer, Germany, while the water samples were measured at the central lab., ENRA. Concentrations were measured in Bq/kg for $^{238}$U, $^{226}$Ra, $^{214}$Pb and $^{214}$Bi in the $^{238}$U series, $^{228}$Ac, $^{212}$Pb and $^{208}$Tl in the $^{232}$Th series, and also $^{137}$Cs and $^{40}$K elements.

**Radiation Exposure Hazard Indices**

**Radium Equivalent Activity**

Radium equivalent activity is an index that has been introduced to represent the specific activities of $^{226}$Ra, $^{232}$Th and $^{40}$K by a single quantity, which takes into account the radiation hazards associated with them. This index can be calculated according to Beretka and Mathew 1985 as:

$$\text{Raeq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K},$$

Where: $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the specific activities of $^{226}$Ra, $^{232}$Th and $^{40}$K in Bq kg$^{-1}$, respectively. The lowest limit of 370 Bq kg$^{-1}$ adopted by the Organization for Economic Cooperation and Development NEA-OECD, 1979.

**Absorbed Dose Rate**

Absorbed dose rate conversion factors to transform specific activities $A_{Ra}$, $A_{Th}$ and $A_{K}$ of $^{226}$Ra, $^{232}$Th and $^{40}$K in Bq kg$^{-1}$, respectively, in absorbed dose rate at 1.0 meter above the ground (in nGy h$^{-1}$ by Bq kg$^{-1}$) are calculated by the Monte Carlo method and the values in UNSCEAR, 1988.

$$D \ (\text{nSv/h}) = 0.49C_{Ra} + 0.67C_{Th} + 0.048C_{K}$$

Where $D$ is the absorbed dose rate in nGy h$^{-1}$ and $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the specific activities of $^{226}$Ra, $^{232}$Th and $^{40}$K in Bq kg$^{-1}$, respectively. The average and range of absorbed dose rate in air due to natural radionuclides in the Egyptian soil is $51$ nGy h$^{-1}$ and 20–400 nGy h$^{-1}$ in areas of high natural radiation background (Monazite sands) UNSCEAR, 2000.

**Internal Hazard Index**

The internal hazard index ($H_{in}$) can be examined according the following criterion Beretka and Mathew, 1985

$$H_{in} = C_{Ra}/185 \cdot C_{Th}/259 \cdot C_{K}/4810 \leq 1$$

Where, $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the specific activities of $^{226}$Ra, $^{232}$Th and $^{40}$K in Bq kg$^{-1}$, respectively.

**External Hazard Index**

The external hazard index is obtained from $R_{eq}$ expression through the assumption that its maximum allowed value (equal to unity) corresponds to the upper limit of $R_{eq}$ (370 Bq kg$^{-1}$). This index value must be less than unity in order to keep the radiation hazard insignificant; i.e. the radiation exposure due to the radioactivity from construction materials is limited to 1.0 mSv$^{-1}$. Then, the external hazard index can be defined as:

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

Where, $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the specific activities of $^{226}$Ra, $^{232}$Th and $^{40}$K in Bq kg$^{-1}$, respectively. ($H_{ex}$ must be less than unity) showing little risk of external hazard to workers handling the sediments.

**The annual gonadal dose equivalent AGDE**

The active bone marrow and the bone surface cells are considered as the organs of interest by UNSCEAR, 1988. Therefore, the AGDE due to the specific activities of $^{226}$Ra, $^{232}$Th and $^{40}$K was calculated using the following formula Mamont-Ciesla et al., 1982.
AGDE (µSv/y) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_{K}

Where, \( A_{Ra}, A_{Th} \) and \( A_{K} \) are the specific activities of \(^{226}\text{Ra}, ^{232}\text{Th} \) and \(^{40}\text{K} \) in Bq kg\(^{-1} \), respectively.

Transfer factor (TF)

The soil-to-plant (TF) quantifies the transfer of radionuclides from soil to plant when uptake by plant roots. From the observed activity concentrations of the radionuclide in the plant and in the corresponding soil, the TF values were calculated according to the following equation Beretka and P.J. Mathew, 1985.

\[
TF = \frac{\text{Activity in plant ( Bq kg}^{-1}\text{ dry weight )}}{\text{Activity in soil ( Bq kg}^{-1}\text{ dry weight )}}
\]

RESULTS AND DISCUSSIONS

Natural Radionuclides Concentrations

For surficial soil samples, the specific activities of \(^{226}\text{Ra} \) (\(^{238}\text{U} \) series), \(^{232}\text{Th} \) series, \(^{40}\text{K} \) and \(^{137}\text{Cs} \) are presented in Fig.2. The average activity concentration along the study area were found to be 19.38 (3.35 to 61.09) Bq/kg of dry weight, 10.87 (1.39 to 24.28) Bq/kg of dry weight, 245.25 (132.46 to 531.19) Bq/kg of dry weight and 1.48 (0.67 to 2.52) Bq/kg of dry weight of \(^{226}\text{Ra} \) (\(^{238}\text{U} \) series), \(^{232}\text{Th} \) series, \(^{40}\text{K} \) and \(^{137}\text{Cs} \), respectively. Thorium and uranium in the soil are contained mainly in resistant heavy minerals such as monazite, zircon and xenotime; potassium is present in the light mineral fraction, such as potash–felspar, mica and glauconite, from which it is slowly converted into soluble forms by weathering processes, Eisenbud, M. 1973.

The obtained results showed that, the lowest values were found in sandy soil, while the highest values were found in sandy loam soil. This can be attributed to the difference in grain size texture since the clayey soil show higher levels than the sandy soils. This is due to the fact that the increases in the surface area cause a higher adsorptive capacity than for the bigger grain size soils as shown in Fig(3). Thus, the concentrations of specific activities of radionuclides increase as the particle size decrease, because of the increase in surface area per unit of mass. The clay minerals are composed mainly of plate-like particles of secondary aluminum silicates and have a negative charged surface, so they have the ability to attract cations that can exchange with the cations adsorbed on the clay surface El-Sharkawy, 2000.

A slight variation in the radioactivity content of the soil can be observed due to the atmospheric deposition, type of soil, soil formation and the sand transport process and geomorphology. Rainfall may result in more effective leaching and/or transport of uranium from soil to sea water. The amount of radionuclides in soil samples are also affected by the amount and composition of the organic matter content, adsorption kinetics and the pH of the medium. Chemical analysis showed higher content of CaCO3 as shown in Fig (4).

On the basis of the physico-chemical analysis of soils which comprised predominantly of sand, silt and clay with the order of magnitude following sand%, clay%, silt%, these soils can be classified as loamy sand and sandy-clay using USDA classification, Miller S, 1994. This is an indication that the radionuclides are probably associated with the same particulate and that they are quickly co-precipitated on the surface of suspended matter. Desorption of radium from estuarine sediments and suspended soil is enhanced by an increase in salinity, Moore W, et. al, 1998 or by the combined effects of low pH and high salinity, Lauria DC, et. al, 2004. A comparison of radionuclide activities in the sediment of the study area and in other coastal and aquatic environments is given in table (1).

In this study, the radiological parameters such as: radium equivalent activity, absorbed dose rate, internal hazard index, external hazard index, annual gonadal equivalent dose and transfer factor were calculated and compared with reported data for all samples from the study area. For surficial soil
samples, the Ra\textsubscript{eq} was evaluated and their values ranged from 15.55 to 95.88 Bq/kg with an average value 52.09 Bq/kg these values lower than the limit of 370 Bq kg\(^{-1}\) adopted by the Organization for Economic Cooperation and Development \textit{NEA-OECD, 1979}. The absorbed dose rate in air was evaluated and their values ranged from 9.73 to 50.07 nGy/h with an average value 28.55 nGy/h, it is clear that all the absorbed dose rate values in the studied area were lower than the permissible level corresponding to 55 nGy h\(^{-1}\) \textit{UNSCEAR, 2000, and El-taher, 2011}, the internal hazard index was evaluated and its values ranged from 0.018 to 0.429 with an average value 0.15 the calculated internal and external hazard values less than unity showing little risk to workers handling the surficial soil and the annual gonadal effective dose was evaluated and its values ranged from 60.54 to 318.12 µSv/y with an average value 182.36 µSv/y.

\textbf{Table (1)}: Summary of activity concentrations of \textsuperscript{226}Ra, \textsuperscript{232}Th and \textsuperscript{40}K (Bq kg\(^{-1}\)) in sediment samples in Egypt and in worldwide

<table>
<thead>
<tr>
<th>Country</th>
<th>Location</th>
<th>Ra-226</th>
<th>Th-232</th>
<th>K-40</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Worldwide</td>
<td></td>
<td>40</td>
<td>35</td>
<td>370</td>
<td>UNSCEAR, 1993</td>
</tr>
<tr>
<td>Worldwide</td>
<td></td>
<td>32</td>
<td>45</td>
<td>420</td>
<td>UNSCEAR, 2000</td>
</tr>
<tr>
<td>Egypt</td>
<td>Suez canal</td>
<td>4.9–20.2</td>
<td>3.3–35.4</td>
<td>59–368</td>
<td>El Tahawy et al., 1994</td>
</tr>
<tr>
<td>Egypt</td>
<td>Red Sea</td>
<td>24.6</td>
<td>31.4</td>
<td>428</td>
<td>El Mamony et al., 2004</td>
</tr>
<tr>
<td>Egypt</td>
<td>Nasser lake</td>
<td>14.4–22</td>
<td>18.4–24.4</td>
<td>222–326</td>
<td>Khater et al., 2005</td>
</tr>
<tr>
<td>Egypt</td>
<td>River Nile sediments</td>
<td>3.8–34.9</td>
<td>2.9–30.1</td>
<td>112–313</td>
<td>El-Gamal et al., 2007</td>
</tr>
<tr>
<td>Egypt</td>
<td>Brullus lake sediments</td>
<td>14.3</td>
<td>20</td>
<td>312</td>
<td>El-Reefy et al., 2010</td>
</tr>
<tr>
<td>Egypt</td>
<td>Red Sea Soil</td>
<td>2.6–45.4</td>
<td>2.4–45.3</td>
<td>81–748</td>
<td>Salama, 2012</td>
</tr>
<tr>
<td>Egypt</td>
<td>Al-Almein- Alam Elrum</td>
<td>16.6</td>
<td>11.8</td>
<td>291</td>
<td>El sayed et al 2015</td>
</tr>
<tr>
<td>Egypt</td>
<td>Hamraween sediments</td>
<td>388</td>
<td>6.9</td>
<td>55.9</td>
<td>El shabasy, 2015</td>
</tr>
<tr>
<td>Egypt</td>
<td>Shalateen-halayeb</td>
<td>19.4</td>
<td>10.9</td>
<td>245.3</td>
<td>Present work</td>
</tr>
</tbody>
</table>

\textbf{Fig. (2)}: Activity (Bq/kg of dry weight) of \textsuperscript{226}Ra (\textsuperscript{238}U) series, \textsuperscript{232}Th series and \textsuperscript{40}K in surficial soil samples.
For plant samples, the specific activities (Bq/kg of dry weight) of $^{226}$Ra ($^{238}$U) series, $^{232}$Th series, and $^{40}$K are presented in Fig (5). The average activity concentrations were found to be 8.59 (1.48 to 21.57), 11.2 (2.64 to 25.44) and 254.66 (69.73 to 450.58) Bq/kg of dry weight respectively.

The $\text{Ra}_{eq}$ was evaluated and their values ranged from 13.39 to 68.65 Bq/kg with an average value 31.09 Bq/kg. The absorbed dose rate in air was evaluated and their values ranged from 7.59 to 37.58 nGy/h with an average value 18.55 nGy/h. The internal hazard index was evaluated and their values ranged from 0.048 to 0.214 with an average value 0.11, the external hazard index was evaluated and their values ranged from 0.038 to 0.192 with an average value 0.09, the annual gonadal effective dose was evaluated and their values ranged from 48.71 to 239.77 µSv/y with an average value 119.63 µSv/y.

The transfer factor has been evaluated as the ratio of concentration (Bq/kg) of an isotope in plant to concentration in surficial soil on a dry weight basis. The transfer factors were shown to be extremely variable depending on vegetation type, various soil parameters and environmental factors as
well as with the different parts of the plant. The transfer factors of $^{238}\text{U}$ were found to be in the range of 0.1–2.8, $^{232}\text{Th}$ was found in the range of 0.14–3.6 and $^{40}\text{K}$ was found to be in the range 0.3–2.6. The uptake of radionuclides by plant roots constitutes the main pathway for the migration of radio-cesium from soil to humans, via food chain. Comparing $^{40}\text{K}$ and $^{137}\text{Cs}$ concentrations in each plant revealed an inverse relationship for these radioisotopes. Correlating the concentrations of $^{40}\text{K}$ and $^{137}\text{Cs}$ were most effective in identifying plants that have a high affinity for $^{137}\text{Cs}$ uptake. Thus, higher $^{40}\text{K}$ soil activity concentration leads to a lower $^{137}\text{Cs}$ plant uptake Rmadan, et al., 2012.

For water samples, Values of pH, dissolved oxygen (DO), total dissolved solid (TDS), and electrical conductivity (EC) (mS/cm) were measured for sea water and the average values were found to be 8.15, 38242, 6.86 and 59.77, and for desalinized water were 6.83, 515, 7.68 and 0.80 respectively.

The concentrations, in mg/L, of the major cations (Ca$^{2+}$, Mg$^{2+}$, and K$^+$, Na$^+$) and anions (Cl$^-$, SO$_4^{2-}$, and HCO$_3^-$) in sea water samples were determined and their concentrations ranged from 439, 1683, 262 and 15100 to 441, 1741, 280 and 15100 mg/L with mean values of 440, 1711, 271 and 15229 mg/L for Ca$^{2+}$, Mg$^{2+}$, and K$^+$, Na$^+$ cations, respectively. The anions concentrations of Cl$^-$, SO$_4^{2-}$, and HCO$_3^-$ ranged from 29164, 4300 and 332 mg/L to 30115, 4485 and 337 mg/L with mean values of 29616, 4373 and 334 mg/L, respectively.

The concentrations, in mg/L, of the major cations (Ca$^{2+}$, Mg$^{2+}$, and K$^+$, Na$^+$) and anions (Cl$^-$, SO$_4^{2-}$, and HCO$_3^-$) in desalinized water samples were determined and their concentrations ranged from 19, 19, 38 and 70 to 23, 21, 41 and 78 mg/L with mean values of 21.33, 20.0, 39.67 and 74.67 mg/L for Ca$^{2+}$, Mg$^{2+}$, and K$^+$, Na$^+$ cations, respectively. The anions concentrations of Cl$^-$, SO$_4^{2-}$, and HCO$_3^-$ ranged from 269, 31 and 23 mg/L to 275, 36 and 29 mg/L with mean values of 271.33, 33.67 and 26.0 mg/L, respectively.

The ionic sequence of abundance for the ions indicates follow the order: Na$^+$$>$ Mg$^{2+}$$>$ Ca$^{2+}$$>$ K$^+$ for cations and Cl$^-$ $>$ SO$_4^{2-}$$>$ HCO$_3^-$ for anions. The ion sequence reflects sodium chloride chemical water type. The concentrations of the major ions are related to and strongly correlated with the result of water conductivity, evaporation, water dilution and circulation.

![Diagram](chart.png)

**Fig. (5):** Activity (Bq/kg of dry weight) of $^{226}\text{Ra}$ ($^{238}\text{U}$ series), $^{232}\text{Th}$ series and $^{40}\text{K}$ in plant samples
The average activity concentration of $^{226}$Ra ($^{238}$U) series, $^{232}$Th series and $^{40}$K in sea water were found to be 3.20 (1.99 to 3.91), 1.93 (1.24 to 2.45) and 15.10 (8.67 to 23.25) Bq/L, and for desalinized water were found to be ND, ND and 13.49 (8.67 to 17.85) respectively. The activity concentration of $^{40}$K in water increases with increasing water, where the highest activity of $^{40}$K measured in sea water samples was 23.25 Bq/L having the highest electric conductivities. The lowest $^{40}$K activity was 9.32 Bq/L, with an average value was 16.70 Bq/L, but for desalinized water the highest activity of $^{40}$K measured were 17.85 Bq/L that have the highest electric conductivities. The lowest $^{40}$K activity were 8.67 Bq/L, with an average value were 13.49 Bq/L respectively.

CONCLUSIONS

The average activities of natural radionuclides in the study area for all environmental samples have been measured along the south eastern Red Sea coast, of Egypt. These results are in agreement with those reported in UNSCEAR, 2000. Comparisons of radionuclides activities in the study area, other coastal and in aquatic environments in Egypt and worldwide were given. The absorbed dose rate values were lower than the permissible level corresponding to 55 nGy h$^{-1}$ UNSCEAR, 2000. The radium equivalent activity Raeq is lower than the limit of 370 Bq kg$^{-1}$ adopted by the Organization for Economic Cooperation and Development NEA-OECD, 1979. The calculated internal and external hazard values were less than unity, and also the annual gonadal effective dose showing little risk to workers handling the sediments. The results of measurements will serve as base line data and background reference level for Egyptian coastlines.

REFERENCES


