Evaluation of natural radioactivity in selected soil samples collected from northern regions of Oman

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Abstract: This study explores the radioactivity concentration in soil in the Northern regions of Oman. The naturally occurring radioactive nuclides of the main series, $^{238}$U, $^{232}$Th, $^{235}$U and $^{40}$K and artificial radionuclide $^{137}$Cs have been studied by means of low level gamma spectroscopy. Majority of the studied valleys (wadis) are descending from Super Hajar Mountains either towards its east part (coastal area) or the west part (desert). Soil samples were collected from headwaters and estuaries of wadis where residences and agriculture activities exist. In soil samples, the results showed that $^{238}$U radionuclide was presented in concentrations of the range (4.5–24.0) Bq.kg$^{-1}$ while $^{232}$Th was found in the range (2.4-20.6) Bq.kg$^{-1}$. Ranges of $^{235}$U and $^{40}$K were reported as (0.6-2.5) Bq.kg$^{-1}$ and (47-296) Bq.kg$^{-1}$ respectively. Artificial radionuclide $^{137}$Cs was found in all soil samples in the range (2.8-11.9) Bq.kg$^{-1}$. The hazard parameters, dose rate (D), annual effective dose ($H_{eff}$), equivalent radium activity ($Ra_{eq}$), internal hazard index ($H_{i}$), external hazard index ($H_{ex}$) and gamma representative level index ($I_{\gamma r}$) were all calculated. The values of D, $H_{eff}$ and $Ra_{eq}$ were found in the ranges (6.6-33.5) nGy/h, (0.8-4.1)*10$^{-2}$ mSv/yr and (17.2-88.6) Bq.kg$^{-1}$ respectively. The values of $H_{i}$, $H_{ex}$ and $I_{\gamma r}$ were all less than 1. These obtained results remained below the optimum admissible values according to UNSCEAR. The results indicate that the radiation hazard from naturally and artificially radionuclides in soil samples from the area studied in this current work is not significant.

Keywords: Radioactivity- Soil – HPGe Detector- Gamma

1. Introduction

Ionizing radiation arises from radionuclides that produce ionizing $\alpha$ and $\beta$ particles and $\gamma$ rays. Two types of radionuclides are recognized, the man made radionuclides and the naturally occurring radionuclides. Higher concentrations of these radionuclides may cause threats to humans in the form of chronic lung diseases, acute, necrosis of the mouth, lung, pancreas, kidney, cancers and leukemia [1]. Under ordinary conditions; artificial radioactivity emitted from the nuclear power plants, industrial plants and research facilities has smaller impact to the overall radiation. Natural environmental radioactivity arises mainly from primordial radionuclides, such as $^{226}$Ra, $^{232}$Th, $^{40}$K and their decay products, which occur at trace levels in all ground formations [2]. Most wadis in Oman are having headwaters descend from the mountains towards the coast or deserts. Majority of the studied valleys are descending from Hajar mountains either towards its east part (coastal area) or the west one (desert). Along these valleys, sub-surface water is available. In result, these regions are agricultural regions and therefore are populous too. Consequently, this study will provide a baseline data of radiation considering 15 locations from northern regions of Oman.
1.1 Gamma (γ) Emission

When a new nucleus is formed after the decay of Alpha or Beta, it is left in an excited state with more energy than usual. This energy is lost by the emission of a gamma ray (photon). For sake of stability, the excited nucleus decays to the ground state by emitting gamma rays with released energy 0.661 MeV (figure 1). This path is followed 94.6% of the time whereas directed β decay carries the rest 5.5% [3].

![Figure (1): Decay of 137mBa by Gamma emission.](image)

A radionuclide’s activity (A) is calculated by the equation:

\[
A = - \frac{dN}{dt} = A_0 e^{-\lambda t} \quad \text{......... (1.1)}
\]

Where \( A_0 \) is the initial count rate at time (t=0), N is the number of radioactive nuclei, and \( \lambda \) is the decay const. and associated with half-life. Half-life is the time for half of the atoms of a given nuclide to decay. The half-life is a unique characteristic of each nuclide range from millions of a second to millions of years.

Naturally occurring radionuclides are distinguished to three decay chains (235U, 238U and 232Th,). The longest-lived member is 232Th, which has half-life of 1.4*10^10 years [2]. Some radioactive decays are non-series. The most important non-series radionuclide in this study is 40K, which contributes 0.012% of natural potassium. Uranium, thorium and potassium are the major radionuclides contained in the Earth’s crust [4]. Table (1) shows radionuclides, their half-lives and major radiation [5].

Table 1: Radionuclides, their half-lives and major radiation
2. Materials and Methods

2.1 Soil samples Collection and Preparation

The sultanate of Oman occupies most of the south east corner of Arabian Peninsula. The distribution of natural radioactivity in the environment depends basically on the geological and geographical properties. In this study, decision was made to collect samples of soil from east and west of western Hajar Mountains. The sampling area considered in this study located at west of north Al Hajar Mountains where the influence of dust storms on the Arabian Peninsula takes place. Heavy rain drifts soil from mountains to the wadis and plain. Disturbed soils cause a decrease in the net surface radiation. Furthermore, the Ophiolites which are igneous rocks constitute the bulk of the geographical configuration of the northern regions of Oman. In terms of natural radioactivity; it is well known that igneous rocks are strongly enriched in thorium and uranium [6]. Samples were collected from 15 different locations. The locations are shown on the map in figure 2.

| Radionuclide  | Half-life | Major Radiation |
|---------------|-----------|-----------------|
| Thorium-228   | 1.91 y    | α,γ             |
| Thorium-232   | 1.40E10 y | α,γ             |
| Uranium-234   | 2.46E5 y  | α,γ             |
| Uranium-235   | 7.04E8 y  | α,γ             |
| Uranium-238   | 4.47E9 y  | α,γ             |
| Radium-226    | 1.60E3 y  | α,γ             |
| Radium-228    | 5.75 y    | β,γ             |
| Radon-222     | 3.82 day  | α,γ             |
| Lead-210      | 22.2 y    | α,γ,β           |
| Potassium-40   | 1.25E9 y  | β,γ             |
| Cesium-137    | 30.1 y    | β,γ             |
Figure (2): Map shows the locations of collected samples from northern regions of Oman.

In the Lab, soil samples were dried in a slow-airflow at room temperature for several days, and then they were sieved through a 1000 μm mesh sieve in order to homogenize the sample and remove organic materials and stones. Half kilogram of each sample was weighed on a digital balance. Later they were sealed in standard geometry containers (0.5 litter Marinelli beakers). The samples were kept for 4 weeks so that equilibrium would take place between $^{226}\text{Ra}$ and its short lived progenies [3].

3. Results and Discussions

Although thorium has higher activity concentrations in world averages according to UNESCAR [7], the results of the present study showed that the activity concentrations of uranium were higher than thorium in all samples. If the ratio is of $^{232}\text{Th}/^{40}\text{K}$ in the soil samples is (>0.5) (figure 3), this will ensure the absence of heavy minerals in the study area and vice versa. This indicates the insoluble state of thorium ion $\text{Th}^{4+}$. In contrast, uranium was found in $\text{U}^{6+}$ oxidation state which is soluble. This interprets the larger concentrations of uranium paralleling with thorium [8].
Figure(3): $^{232}$Th/$^{40}$K Activity ratios of soil samples

However, the values measured remain below the optimum worldwide average values and appropriately comparable to concentrations reported from different countries (Table 2).

Table (2): A comparison of activity concentrations in soil between this study and different studies reported in other countries.

| Country                  | Activity concentration(Bq.kg$^{-1}$) | References |
|--------------------------|--------------------------------------|------------|
|                          | $^{232}$Th | $^{40}$K | $^{226}$Ra |                  |
| Present study            | 2.4 - 20.6 | 47.2 - 224.3 | 10 - 43 | Present study    |
| Oman, Musandam           | 4.5       | 86.4     | 25.4     | [9]             |
| Oman, Musandam           | 9.95      | 158.21   | -        | [10]            |
| AlQasseem, Saudi Arabia  | 2.5-3.9   | 212-915  | 1.4-35.3 | [11]            |
| Riyadh, Saudi Arabia     | 7-25      | 89 - 320 | 11 – 30  | [12]            |
| Jedda, Saudi Arabia      | 7.4       | 369      | 9.3      | [13]            |
| Ad-Dahna, Saudi Arabia   | 15.8 - 36.7 | 285.3 - 533.2 | 16.2 - 30.6 | [14]        |
| Yemen                    | 18 - 113  | 64 – 1667 | 16.6 – 84.4 | [1]           |
| World-wide               | 45        | 420      | 25       | [6]             |
3.1 Variation of $^{238}$U series activities

The stability in $^{238}$U series was checked by plotting a linear fit between $^{238}$U and $^{226}$Ra (figure 4), which shows a linear regression for their activity concentrations. It showed a statistically correlation with a correlation coefficient of $(R=0.77)$ and with $P < 0.0004$, where $P$ is the probability of rejecting the truth. However the correlation $(R=0.77)$ is not significant and indicate the result gotten before that uranium is less stable and more soluble (+6 state) in the study area. This explained the two upper shifted points in figure (4).

![Figure (4): Linear regression of the activity concentrations of $^{238}$U versus $^{226}$Ra for all soil samples.](image)

3.2 Variation of $^{232}$Th series activities

Activity concentrations were calculated in soil for the significant radionuclides recognized in $^{232}$Th decay series (see figure 5), which are $^{228}$Ac, $^{212}$Pb and $^{212}$Bi.

![Figure (5): Variation of the concentrations of radionuclides in $^{232}$Th series](image)
This study shows that thorium series in R=0.98) is more stable than uranium series (R=0.77) and it is less soluble. The stability between uranium series and thorium series in soil was checked by plotting a linear fit (Figure 6) between the activity concentrations of $^{238}$U and $^{232}$Th radionuclides. The correlation shows low significance with R=0.83.

![Figure (6): Linear regression between uranium and thorium series for all soil samples.](image)

3.3 Variation of $^{137}$Cs activity

The activity concentrations of the artificial nuclide $^{137}$Cs from soil samples showed some of the values to be bit higher than the values obtained from other studies conducted in neighboring countries as shown in table (3). The investigated area, Hajar Mountains are very high mountains and peaking at about 3000 meters above sea level. The released cesium in the air blend with clouds accumulated on the top of Hajar Mountains. Later these amounts of $^{137}$Cs are deposited on the wadis by rain. However, the influence on humans from such low levels of $^{137}$Cs is still insignificant.

| Study area          | Activity concentrations of $^{137}$Cs in soil (Bq.Kg\(^{-1}\)) | References |
|--------------------|---------------------------------------------------------------|------------|
| Northern Oman      | 2.8-11.9                                                      | Present study |
| Oman, Musandam     | 1.7                                                           | [9]         |
| Kuwait              | 4.45                                                          | [16]        |
| Iraq                | 15.9-70.9                                                     | [17]        |
| Jordan              | 1.3                                                           | [18]        |
| Pakistan            | 3.5                                                           | [19]        |
| Saudi Arabia        | 1.7                                                           | [20]        |
| Saudi Arabia        | 4.5                                                           | [21]        |
4. Assessment of hazards

Exposure to radiation can be defined in terms of many parameters, such as absorbed dose rate, annual effective dose, radium equivalent activity, Gamma representative level and internal and external hazard indices.

The external absorbed dose rate in air at a height of about 1 m above the ground was calculated by:

$$D (\text{nGy. h}^{-1}) = 0.0417 A_R + 0.462 A_{Ra} + 0.604 A_{Th}$$ …….. (1)

According to the annual effective dose received by a member in mSv.y^{-1} unit, the values obtained for the water samples were calculated using equation:

$$H_R (\mu \text{Sv.y}^{-1}) = 8760 \times 0.2 \times 0.7 \times 10^{-6} \times D$$ …………… (2)

The radium equivalent activity (Raeq) was calculated by equation:

$$R_{aeq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_K$$ ………………………………………….. (3)

The external, internal and gamma representative level hazard indices which are on concern due to their impact on people health were calculated using:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1$$ ………………………………………….. (4)

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1$$ …………… (5)

$$I_{yr} = \frac{A_U}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \leq 1$$ ………… (6)

formulas respectively. Where $A_{Ra}$, $A_{Th}$ and $A_K$ are the activities of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in Bq.kg^{-1}, respectively [22]. The calculations as shown in table (4) ensured that the values of these indices are less than the unity, the optimum acceptable limit in globe. Hence these areas can be considered as risk free zones from human health perspective.

Table (4): Hazard parameters of soil samples conducted from different studies.

| Country               | D (nGy/h) | $H_R$ ($^{*}10^{-2}$) (µSv/yr) | $R_{aeq}$ (Bq.kg^{-1}) | $H_{ex}$ | $H_{in}$ | $I_{yr}$ | References |
|-----------------------|-----------|--------------------------------|------------------------|----------|----------|---------|------------|
| Present Study         | 6.6-33.5  | 0.8-4.1                        | 17.2-88.6              | 0.07-0.34| 0.04-0.19| 0.10-0.53| Present study |
| Oman, Musandam       | -         | 2.2                            | 38.5                   | -        | -        | -       | [9]         |
| Oman, Musandam       | -         | 2.5                            | 40.8                   | -        | -        | -       | [10]        |
| Al-Qasseem, Saudi Arabia | 35.2    | 4                              | 68.1                   | 0.21     | 0.18     | 0.54    | [11]        |
| Ad-Dahna, Saudi Arabia | 51.4    | 3.2                            | 106                    | -        | 0.29     | -       | [21]        |
| Basrah, Iraq         | 49.35     | -                              | 105.6                  | 0.38     | 0.29     | -       | [15]        |
| World-wide           | 57        | 7.0                            | 128.7                  | <1       | <1       | <1      | [6]         |
In most locations, soil samples activity concentrations parents’ radionuclides and their daughters were lower than the recommended worldwide limits except for six locations activity concentrations of $^{226}$Ra were found to be higher than the worldwide average. Artificial radionuclide $^{137}$Cs was found in all soil samples as a result of the global fallout contamination from nuclear weapon testing and nuclear accidents around the world.

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