Natural Radionuclides of Carbonaceous Fossil Wood Trunks in Um Bogma Formation, Sinai, Egypt

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Abstract: The Lower Carboniferous Um Bogma Formation in west-central Sinai, Egypt displays extensive and pervasive dolomitization. Organic materials especially the carbonaceous matter was found to play an important role in the distribution of the radionuclides within soils and sedimentary rocks. The Um Bogma Formation is the more important formation than the others as it hosts most of the radioactive anomalies. Ten samples were collected from carbonaceous fossil wood trunks and fragments embedded in siltstone and shales of early Carboniferous age. These samples were studied by X'Pert diffractometer for the mineral and chemical composition. The most important minerals in the samples are Goethite (Iron Oxide Hydroxide), Graphite(Carbon), Quartz (SiO_2), and Kaolinite (Aluminum Silicate Hydroxide). The samples were also analyzed for concentrations (Bq/kg). of ^{238}U, ^{232}Th series and ^{40}K by HPGe gamma spectrometer. The concentrations Bq/kg ranged from 748.07 to 1459.94, 926.75to 1845.90, 43.75 to97.13, 17.58 to 68.09, and from 684.27 to 2019.21 for ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U, and ^{40}K respectively. The $^{226}Ra/^{238}U$, $^{238}U/^{235}U$, and $^{238}U/^{232}Th$ ratios in samples ranged between1.00- 1.81, 17.00-18.50, 9.43-21.14 respectively. These variations in the concentrations of radioelements and their ratios are due to the action of the alteration processes. The Ra_{eq} ranged from 1042.00 to2045.45 Bq/kg with mean value 1550.72 Bq/kg. The Ra_{eq} in all samples exceeds the permitted limits (370 Bq/kg). Absorbed Dose Rate (nGy/h) ranged from 482.64 to 949.02 with mean value 720.53, Annual Effective Dose (mSvy/h) ranged from0.59 to 1.17 with mean value 0.88, and External Hazard Index ranged from 2.82 to5.53 with mean value 4.19. From the obtained results, the mean value of Annual Effective Dose (0.88) is higher than the permissible levels (0.48) and the calculated average value of external hazard index is higher than unity. This result points to a dangerous effect in that region for human health.
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1. Introduction

Coal is an important energy source of the nature and it is the most significant source for power generation worldwide, and coal bed methane production in most of the countries.

Fossil fuel, coal is formed by the geological action over millions of years. Plants perished in vast swamp areas. The weight of the top layers of water and dirt generated heat and put pressure on the lower layers of plant matter over a prolonged period of time. The decayed plants and animals dried and hardened to form carbon-rich coal.

Coal, like most materials found in nature, contains quantities of naturally occurring radionuclides, ^{238}U, ^{232}Th and ^{40}K. The Carboniferous sedimentary rocks in both sides of the Gulf of Suez, Egypt are found to be radioactive anomalous. (Aassy (1981) recognized trunks and stems of fossil woods in the late Carboniferous Rod El Hamal Formation with relatively high uranium content in the carbonaceous parts than in the silicified parts. In southwestern part of Sinai Peninsula, eastern part of Gulf of Suez, defined section of the early Carboniferous Um Bogma Formation is exposed. Trunks of fossil woods are noticed between the lower and middle members of Um Bogma Formation.

There are many studies on the early Carboniferous Um Bogma Formation [Agamy 1996]. Sharkawi et al.1999Agami et al, 2000, Aassy et al.2000,

The main aim of this work is to study the effect of the organic carbon (carbonaceous matter) of different radio nuclides to determine the dose emitted and the effective dose emitted from samples, and to estimate the hazard index to be compared with the world values.

Geologic Setting

The Um Bogma Formation is the more important formation than the others as it hosts most of the radioactive anomalies and it is subdivided into three members known as:

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(1) Lower Shaly-Ore Member comprises black shales with thin sandy dolomite bands and manganese-iron ore. (Sharkawi *et al.,* 1999) introduced the karstification process in this member and the formation of karstified dolostone rocks with intrakarstic product housing manganese ores and caliche nodules. Aassy *et al.* (2000) introduced

the lateritic profile section and the formation of gibbsite-bearing shale. This rock unit is highly radioactive when it is karstified and lateritized.

(2) Middle Marly Dolostone-Siltstone Member is also karstified and lateritized and consists of marl with siltstone and gibbsite-bearing siltstone. Its thickness is 6–8 m and moderately radioactive.

(3) Upper Dolostone Member is unconformably overlying the karstified and lateritized soil profile and consists of bedded dolostone with thin shale interbeds. The dolostone beds are present as step-like forms and in some parts are not deposited and laterally validated to grey clay stone (Aassy Ibrahim , et al, 2011).

2. Samples and Sample Preparation

Ten samples are collected from different locations of the carbonaceous fossil wood trunks and fragments of Um Bogma Formation in Talet Seleim area, southwestern Sinai, Egypt as shown in Fig (1). These samples are crushed to 200 mesh and put in 200 ml polyethylene containers for gamma measurements. The crushed samples were stored for four months to reach secular equilibrium between ^{238}U and ^{232}Th and their progenies.

Fig. 1: Location Map of the Collected Samples

Measurements

The dried sample were studied by X'Pert diffractometer for the chemical and mineral composition, gamma measurements were taken by a closed-end coaxial gamma-ray detector (p-type) made of high purity germanium (HPGe) in a vertical configuration (Pop Top-cryostat configuration) cooled with liquid nitrogen. The used HPGe EG&G Ortec Model GEM-50210-P has a full width of half maximum (FWHM) of 0.8 keV at the 122 keV gamma transition of ^{57}Co and 1.9 keV at the 1.33 MeV gamma transition of ^{60}Co. The detector has a photo-peak relative efficiency of about 50% of the 3"×3" NaI(Tl) crystal efficiency.

The Ortec 4001M Minibin & Power Supply connects to the Ortec 659 0-5kV detector bias supply and the Ortec Model 570 amplifier through the two pins. A power ground return, a high-quality signal ground, and ±12V and ±24V. The power supply furnishes four standard DC voltages; ±12V at 1A and ±24V at 0.5A. The detector preamplifier used in this study was an Ortec Model 257P. The analog-to-digital converter was an EG&G Ortec TRUMP^{TM}-8k/2k card controlled by the MAESTRO^{TM} Model A65-B32 software installed on a PC computer operating under Windows-98.

Energy 63.3 keV of ^{234}Th was taken for measuring the ^{238}U concentrations (Bq/kg). ^{234}Th is short-lived and progeny of ^{238}U, therefore, secular equilibrium was assumed between ^{238}U and ^{234}Th and thus ^{234}Th

Was assumed to represent actual ^{238}U activities . For ^{226}Ra concentrations (Bq/kg) were calculated from energies of 295.1 and 351.87 keV of ^{214}Pb and

609.31 keV of ^{214}Bi. For the ^{232}Th concentrations (Bq/kg) energies of 338.4 keV and 911.2 keV from ^{228}Ac and 583 keV and 2614.4 keV from ^{208}Tl were taken. For ^{235}U concentration (Bq/kg) energy of 143.8 keV was taken. For ^{40}K concentration (Bq/kg) the 1460.8 keV was taken for measuring the concentrations. All concentrations were measured in Bq/kg.

Determination of activity concentrations in Bq/kg was calculated using the equation (1) (Amrani.and Tahtat.,2001).

$$A = \frac{C}{M \beta \varepsilon} \quad (1)$$

Where: C is the net peak area of specific gamma ray energy (count per second).

M is the mass of the samples (kg).

β is the transition probability of gamma-decay.

ε is the detector efficiency at the specific gamma-ray energy.

Exposure to radiation has been defined in terms of the radium equivalent Ra_{eq} in Bq/kg which is calculated from equation (2) (Tufail *et al* 2006).

$$Ra_{eq} = C_{Ra} + (C_{Th} \times 1.43) + (C_K \times 0.077) \quad (2)$$

Where:

C_{Ra}, C_{Th} and C_K are the concentrations Bq/kg for radium, thorium and potassium respectively.

Besides the radium equivalent activity, the total air absorbed dose rate (nGy h^{-1}) in the outdoor air at 1 m above the ground due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq/ kg) dry weight was calculated using the equation (3) (UNSCEAR 2000; Veiga et al. 2006)

$$D(nGyh^{-1}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \quad (3)$$

Where:

C_{Ra}, C_{Th}, and C_K are the specific activities (concentrations) of ^{226}Ra, ^{232}Th and ^{40}K in Bq /kg respectively.

By using a conversion factor of 0.7 SvGy^{-1} and outdoor occupancy factor of 0.2 (people spend about 20% of their life outdoor) the Annual Effective Dose (in mSvy-1) received by population can be calculated using equation :

$$D_{eff}(mSvy^{-1)} = D(nGh^{-1}) \times 8{,}766\ h \times 0.7(SvGy^{-1}) \times 0.2 \times 10^{-6} \quad (4)$$

Where:

D (nGh^{-1}) is the total air absorbed dose rate in the outdoor.

8,766 h is the number of hours in 1 year.

10^{-6} is conversion factor of nano and milli.

From Ra_{eq} an external hazard index, H_{ex} can be computed by the supposition that the maximum value allowed for the index is 1, that corresponds to Ra_{eq} = 370 Bq/kg. Thus it can be calculated in Bq/kg by (Beretka and Mathew 1985):

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \quad (5)$$

3. Results and Discussions

Table (1) represents the results obtained by X'Pert diffractometer . The most important minerals in the samples are Goethite (Iron Oxide Hydroxide), Graphite(Carbon), Quartz (SiO_2), and Kaolinite (Aluminum Silicate Hydroxide).

Table (1): The mineral compositions of the samples analyzed by X'Pert diffractometer**, (Leet *et al*., 1982, and Mineral Data, *2012***

Reference code	Mineral name	Chemical Composition	Description
003-0251	Goethite	$Fe^{3+}O(OH)$ Iron Oxide Hydroxide	Common in iron ore deposits; one of the commonest minerals formed under oxidizing conditions as a weathering product of iron-bearing minerals.
025-0284	Graphite	C Carbon	Metamorphosed lime stones, organic-rich shales, and coal beds.
005-0490	Quartz	$Si O_2$ Silicon Oxide	Sedimentary, metamorphic, and igneous rocks and Very common mineral found world wide
003-0058	Kaolinite	$Al_2Si_2O_5(OH)_4$ Aluminum Silicate Hydroxide	Secondary mineral derived from the weathering of aluminum-silicate minerals

In the samples, the natural radionuclides (^{238}U, ^{226}Ra, ^{232}Th, ^{235}U, ^{40}K,) were detected. The obtained concentrations of naturally occurring radionuclides in the analyzed samples are presented in Table (2).

Table (2): The activities concentrations Bq/.kg dry weight of the ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U and ^{40}K for themeasured samples.

Samp. No.	^{238}U	^{226}Ra	^{232}Th	^{235}U	^{40}K	$^{226}Ra/^{238}U$	$^{238}U/^{235}U$	$^{238}U/^{232}Th$
1	924.76±33.73	926.75±2,65	43.75±4.30	50.60±1.90	684.27±18.02	1.00	18.28	21.14
2	1171.54±40.14	1247.64±3.05	75.21±4.30	68.09±1.17	1624.98±23.51	1.07	17.21	15.58
3	1071.68±35.37	1248.25±4.06	62.74±1.73	58.99±1.80	1369.20±29.01	1.16	18.17	17.08
4	748.07±29.12	1353.77±2.91	47.93±1.60	40.81±1.92	1018.94±24.17	1.81	18.33	15.61
5	1459.94±50.67	1845.90±3.10	63.77±4.60	17.58±2.24	1407.22±28.13	1.26	17.58	15.03
6	916.04±42.94	1213.31±3.01	97.13±2.47	50.15±1.83	2019.21±30.54	1.32	18.23	9.43
7	921.31±41.95	1179.93±3.02	94.82±4.30	54.22±1.80	1848.89±28.35	1.28	17.00	9.72
8	1092.07.±54.62	1633.23±3.20	65.54±4,60	60.52±2.10	1381.51±29.23	1.50	18.05	16.66
9	942.20±46.23	1456.86±3.20	72.03±4.51	51.01±2.01	1886.47±26.59	1.55	18.47	13.08
10	1113.14±42.61	1258.02±1.85	76.18±3.83	60.16±1.84	1614.21±27.91	1.13	18.50	14.61

The ^{238}U concentrations measured from ^{234}Th ranged between 748.07±29.12 and 1459.94±50.67 Bq/kg. Values of ^{226}Ra concentrations ranged between 926.75±2,65 and1845.90±3.10 Bq/kg. It seemed, that the ^{226}Ra activity concentrations were higher than that measured for ^{238}U with ($^{226}Ra/^{238}U$) ratios more than unity. This disequilibrium in the ^{238}U series because the difference in chemical and physical properties of the two elements in the series **(**Ivanovich and Harmon, 1982) and some processes led to migration of uranium from the parent rocks and sediments. ^{232}Th series concentrations ranged between 43.75±4.30 and 97.13±2.47 Bq/kg. Ratios of $^{238}U/^{232}Th$ ranged between 9.43and 21.14 which means enrichment of ^{238}U than ^{232}Th. ^{235}U activity concentrations ranged between 17.58±2.24 and 68.09±1.17Bq/kg. The ratios ^{238}U /^{235}U ranged between 17.00 and 18.50 which showed general deviation from natural ratio (21). ^{40}K concentrations ranged from 684.27±18.02 and 2019.21±30.54Bq/kg.

Table (3) shows the Radium Equivalent (Bq/kg), Absorbed Dose Rate (nGy/h), Annual Effective Dose, (mSvy/h) External Hazard Index for samples.

Ra_{eq} values ranged between1042.00 and 2045.45 Bq/ kg, with mean values 1550.72 Bq/ kg, which are higher than the recommended maximum value of 370 Bq/ kg. The annual effective dose equivalent (mSv/ y) in air was calculated using the values of the absorbed dose rate. The average values of annual effective dose for all studied samples were 0.88. This value is higher than 0.48 mSv/y recommended by UNSCEAR . This means that these samples are not safety for human beings from the environmental radiation point of view. The calculated average value of external hazard index is higher than unity. This result points to a dangerous effect in that region for human health.

Table (3): The Radium Equivalent (Bq/kg), Absorbed Dose Rate (nGy/h), Annual Effective Dose, (mSvy/h) External Hazard Index for samples .

Samp. No.	Ra_{Eq} Bq/kg)	D(nGy/h)	D_{eff} mSv/y)	H_{ex}
1	1042.00	482.64	0.59	2.82
2	1480.31	688.46	0.85	4.00
3	1443.40	670.72	0.82	3.90
4	1500.77	696.17	0.85	4.06
5	2045.45	949.02	1.17	5.53
6	1507.69	702.00	0.86	4.07
7	1457.89	678.20	0.83	3.94
8	1833.33	850.78	1.04	4.95
9	1705.12	793.92	0.97	4.61
10	1491.25	693.40	0.85	4.03
Min.	1042.00	482.64	0.59	2.82
Max.	2045.45	949.02	1.17	5.53
Mean	1550.72	720.53	0.88	4.19

Conclusions

The lower Carboniferous sedimentary rocks of Um Bogma Formation in southwestern Sinai was found to have some carbonaceous trunks and stems. These carbonaceous bodies are embedded in siltstone, shales and sandstone.

The concentrations of radionuclides ^{238}U, ^{235}U, ^{232}Th, ^{226}Ra and ^{40}K were measured in ten collected samples from these carbonaceous trunks using HPGe

detector. The samples collected to represent different grades of carbonization. The results showed that there is a variation in ^{238}U activity concentrations and this variation is related to variation in carbon content according to the difference color and tone.The ratios $^{238}U/^{235}U$ clarified low deviation from the natural one (21.7). the ^{226}Ra activity concentrations were higher than that measured for ^{238}U with ($^{226}Ra/^{238}U$) ratios more than unity. This disequilibrium in the ^{238}U series because the difference in chemical and physical properties of the two elements in the series and some processes led to migration of uranium from the parent rocks and sediments. Ratios of $^{238}U/^{232}Th$ ranged between 9.43and 21.14 which means enrichment of ^{238}U than ^{232}Th.

The average values of $Ra_{eq,}$ annual effective dose, and external hazard index are higher than the recommended maximum values. These results point to a dangerous effect in that region for human health.

References

1. Aassy, I. E.; Ahmed, F. Y.; Afifi, S. Y. and El Shamy, A. S. (2000): "Uranium in laterites, Southwestern Sinai, Egypt". *First Seminar on Nuclear Raw Materials and Their Technology, Cairo, Egypt, 1-3 Nov., p. 1-20.*
2. Aassy Ibrahim E, El Galy Mohamed M., Nada Afaf A., El Feky Mohamed G., Abd El Maksoud Thanaa M., . Talaat Shadia M, . Ibrahim Eman M, 2011, Effect of alteration processes on the distribution of radionuclides in uraniferous sedimentary rocks and their environmental impact, southwestern Sinai, Egypt, *Radioanal Nucl Chem 289:173–184*.
3. Aassy I.E., Ahmed FY, Afifi SY, El Shamy AS 1999, Uranium in laterites, Southwestern Sinai, Egypt, *In: First seminar on nuclear raw materials and their technology, Cairo, Egypt, 1–3*
4. *Nov 1999, pp 1–20.*
5. Aassy, I.E., 1981, Structural and Radiometric Studies of Wadi Araba Area, Easter Desert,Egypt, *Ph.D. Thesis, Mansoura University, Mansoura, Egypt, 297 p.*
6. Amrani D., Tahtat M.,2001, Natural radioactivity in Algerian building materials, *Applied Radiation and Isotopes 54 , 687-689.*
7. Beretka J , Mathew P. J 1985, Natural radioactivity of Australian building materials industrial wastes and byproducts, *Health Phys. 48:87–95*
8. Agami N. L , Ibrahim E. H. , Odah H. H., 2000, Sedimentary Origin of the Mn-Fe Ore of Um Bogma, Southwest Sinai: Geochemical and Paleomagnetic, *Economic Geology, v. 95 no. 3 p. 607-620.*
9. Agamy NL, 1996, Geology and radioactivity studies on the Paleozoic rock units in the Sinai Peninsula, Egypt. *PhD thesis, Fac. Sci., Mansoura Univ., p 302.*
10. Ivanovich, M. and Harmon, R.S. (1982): "Uranium Series Disequilibrium, Applications to Environmental Problems". *Clarendon Press, Oxford 1982, pp. 571.*
11. Leet L., Judson Sh., Kauffman M., 1982, Physical Geology, sixth edition, *Englewood Cliffs, New Jersey0763*Sharkawi M, El Aref M, Abdel Mottelib A 1989, Manganese deposits in Carboniferous paleokarst profile, Um Bogma region, west central Sinai, *Egypt. Miner Deposita 25:34–43.*
12. Mineral Data, *2012, webmineral.com.*
13. Tufail , M. , Nasim Akhtar., M. 2006, Measurement of terrestrial radiation for assessment of gamma dose from cultivated and barren saline soils of Faisalabad in Pakistan, *Radiation Measurements, 41, pp. 443-451.*
14. UNSCEAR (2000), United Nations Scientific Committee on the Effects of Atomic Radiation, ''Sources and Effects of Ionizing Radiation''. *Report to General Assembly, with Scientific Annexes United Nations. United Nations, New York*.

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