Activity Concentrations of Natural Radionuclides in Sedimentary Rocks from North of Arabian Shield (Hail), Saudi Arabia

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Abstract: Sedimentary rock geological and radioactivity studies are important for their use as raw materials in the construction industry.Six samples were collected from south of Hail at the east and north of the Arabian Shield of Saudi Arabia. Their coordinates between Lat. N: 26^{0}04$^'$09.1 to N:28^0 59$^'$ 01.3$^{''}$ and long. E:43^0 35$^'$16.1$^{''}$to E:45^{0}19$^'$59.3$^{''}$. Samples were analyzed by XRD for the mineral constituents. X-RD results give the major, minor and trace minerals, the major mineral is calcite (CaCo3), Dolomite ($CaMg(CO_3)_2$), and Quartz(SiO_2), with minor and trace concentrations. Also, the dried samples were analyzed by atomic absorption spectrometer for the Al, Ca, K, Bi, Pb, andTh, concentrations in ppm and/or percent.**Results** give concentrations in percent and ppm ranging from 0.10 to 0.68, 3.16 to 33.53, and 0.13 to 0.31 in %, and <10, <7.5 to 1756.38, and <1 in ppm respectively. Using high-resolution gamma-ray spectroscopy, the activities concentrations Bq/.kg dry weight for the ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U and ^{40}K, ranged from 176.10$\pm$0.07 to 222.86$\pm$0.21, 28.34$\pm$0.11 to 231.04$\pm$0.05, 08.66$\pm$0.17 to 137.84$\pm$0.04, 09.35$\pm$0.03 to 12.69$\pm$0.03, 78.27$\pm$0.23 to 202.21$\pm$0.04 with mean values 199.18$\pm$0.17, 109.01$\pm$0.07,37.96$\pm$0.10, 11.47$\pm$0.02, and 123.57$\pm$0.13 respectively.For theRa$_{Eq}$ (Bq/kg) ranged from 59.193 to 438.615 with mean value 172.810 which is lowr than *370Bq/kg*, the permissible limit **(UNSCEAR, 2000).** The mean values of the annual effective dose (D_{eff}(mSv/y) was found to be less than one (0.096) which is within the worldwide mean values($<$1for D_{eff} (mSv/y) (UNSCEAR, 2000).

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Key words: Dolomite rock, Sand stone, high-resolution gamma-ray spectroscopy, Arabian Shield of Saudi Arabia

1. Introduction

The sedimentary rocks to the east and north of the Precambrian Proterozoic Arabian Shield cover almost two-third of the total area of Saudi Arabia. The younger rocks in Saudi Arabia belong to the Paleozoic (540-250 Ma), Mesozoic (250-65 Ma), and Cenozoic (65 Ma to Recent) (collectively referred to as Phanerozoic cover), and crop out as relatively flat lying beds of sedimentary rocks such as sandstone, siltstone, limestone, and evaporates (salt deposits). The youngest deposits in the region include coral limestone and unconsolidated sand, silt, gravel, and sabkha, which accumulated in the sand seas of Ar Rub al Khali and An Nafud, filled dried-up lake beds and wadis, and fringed the coastlines. Sedimentary geological and radioactivity studies are important for their use as raw materials in the construction industry (bricks, ceramics, cement, fillers, etc.), and for its a science used to examine rock formations. It helps to determine the types, classification, criteria, geologic age of rocks, their contents of Phanerozoic fossils **(Saudi Geological Survey (SGS), 2012).**

The concentrations and distributions of natural radionuclides occurring in sedimentary rock samples from Eastern Desert and Nile Valley in Egypt, were measured. The sediments have already been investigated in the geological and mineralogical aspects, this study is necessary to investigate the natural radioactivity in order to complete their classification **(ARABI et al, 2006).**

Higher radiation levels are associated with igneous rocks, such asgranite, and lower levels with sedimentary rocks. There are exceptions, however, as someshales and phosphate rocks have relatively high content of radionuclides (**Tzortzis et al,2003**).

In sedimentary rocks such as sandstone, limestone, and non-carbonaceous shale, most of the radionuclides are in the detrital particles **(Johnson, 1979).**

WadiNaseib area of Egypt is mainly covered by sedimentary rocks of Paleozoic age. Some of the Paleozoic sediments in the southwestern part of Sinai, including the study area, are of great importance especially from the mineralogical and radioactivity points of view. They host several types of mineral deposits, some transitional metals **Galy et al, 2008).** The radioactivity in soil are primary comes from U, Th and their progenies and also from the natural K, and represents the main external exposure to the environment **(Hamzah et al,2011).**

The aim of this study is to analyze the sedimentary rock samples at the east and north (south of Hail) of the Arabian Shield of Saudi Arabia by XR-D spectrometer and atomic absorption spectrometer and to determine the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U, and ^{40}K by using gamma rays spectrometer with HPGe detector. This study will also calculate the radium equivalent and then estimate the annual effective dose and the external hazard index of the study area.

Experimental techniques

Geological setting:

Sedimentary rocks formed when eroded particles of other rocks have been deposited (on the ocean floor, stream/lake beds, etc) and compacted, or by the precipitation of minerals mineraloids from water. Sedimentary rocks contain important information about the history of the Earth. They contain fossils, the preserved remains of ancient plants and animals. All buildings and public structures require sedimentary rock in their construction, so measuring the activity concentrations of radionuclides in these rocks are very important for the environment. Six samples were collected from south of Hail as shown in figure 1.

Sample 4 K.do contains Dolomite rock. Dolomite is very significant in the petroleum business because it forms underground by the alteration of calcite limestone. This chemical change is marked by a reduction in volume and by recrystallization, which combine to produce open space (porosity) in the rock strata.

Sample5 Tudo contains of limestone. Limestone is a sedimentary rock consisting of more than 50% calcium carbonate (calcite - $CaCO_3$). There are many different types of limestone formed through a variety of processes. Uses of limestone are: base for cement; as dimension stone for decoration of walls and floors; in the production of lime fertilizer, paper, petrochemicals, pesticide, glass etc.

Sample 6 F.ss includes Sand stone which is a sedimentary rock formed from cemented sand-sized clasts. The cement that binds the clasts can vary from clay minerals to calcite, silica or iron oxides. It is used as: if soft then generally of no use; if hard then can be used as aggregate, fill etc. in the construction and roading industries; dimension stone for buildings, paving, etc. (Saudi Geological Survey,2012).

Fig. 1 Map of the samples' locations

Sample preparation

Six samples were collected from south of Hail at the east and north of the Arabian Shield of Saudi Arabia. Samples were grounded, sieved by 1mm x 1mm, then dried to 95°C for 24 hours in order not to lose the volatile polonium or cesium. The dried fine grained samples were packed in polyethylene Marinelli beakers for gamma spectroscopy, and then stored for up to four months to reach secular equilibrium between ^{238}U and ^{232}Th and their progenies

Measurements

Ten grams of the dried samples were analyzed by XRD spectrometer model Burker XR-D D8 Advance for the mineral constituents, also 10 grams of the dried samples were analyzed by atomic absorption spectrometer model AAnalyst 700 for the Al, Ca, K, Bi,Pb, and Thconcentrations in ppm and/or percent. Samples were analyzed for concentrations of ^{238}U, ^{232}Th series, ^{235}U, and ^{40}K using the gamma spectrometer based on Canberra hyper pure germanium detector "HPGe" coaxial detector with relative efficiency of 20% and FWHM 4.2 keV at 1461 keV, the measurements were done for a time period of twenty four hours.

For sedimentary rocks,^{238}U was calculated from a gamma-ray line of energy 63.29keV, of ^{234}Th. Gamma-ray lines of energies 295.09, 351.87, 609.31, 1120.27, and 1764.49 keV resulting from the decay of daughters ^{214}Pb and ^{214}Bi radionuclides (which they are in secular radioactivity equilibrium) were used to determine the activity concentrations of ^{226}Ra. The gamma-ray lines at338.42, 911.07, 968.97, 583.10, and 2614.48 kev from the decay of short half life daughters ^{228}Ac and ^{208}Tl were used to determine the activity concentrations of ^{232}Th sespectively (since there is secular radioactivity equilibrium in ^{232}Th series); while the143.8and 1460.80 transitions were used to determine the activity concentrations of ^{235}U and ^{40}K respectively.

The specificactivity (A) in Bq/Kg for each detected nuclide was calculated using the following equation:(Amrani &Tahtat., 2001).

$$A = \frac{c}{M\,\beta\,\epsilon} \quad (1)$$

Where: c is the net counting rate of a specific gamma ray (count per second)

M is the mass of the samples (kg)

$\beta$is the transition probability of gamma-decay

$\epsilon$is the detector efficiency at the specific gamma-ray energy.

Radium equivalent activity, $Ra_{Eq}$is used to estimate thehazard posed by different concentrations of radionuclides in materials. Equation (2) was used to determine Ra_{eq}(Bq/.kg) (Tufail & NasimAkhtar, 2006):

$$Ra_{eq}(Bq/.kg) = C_{Ra} + (C_{Th} x\ 1.43) + (C_K x0.077) \quad (2)$$

Where:C_{Ra}, $C_{Th}$and C_K are the concentrations Bq/kg for radium, thorium and potassiumrespectively.

The total air absorbed dose rate D(nGy/ h) 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)

$$D(nGy/h) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K (3)$$

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 SvG/y and outdoor occupancy factor of 0.2 (people spend about 20% of their life outdoor) the Annual Effective Dose (in mSv/y) received by population can be calculated using equation:

$$\mathbf{D_{eff}(mSv/y) = D(nGy/h)\times 8{,}766\ h \times 0.7(SvG/y) \times 0.2 \times 10^{-6}} \quad \mathbf{(4)}$$

Where:D(nGy/h) 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.

3. Results and discussions

As shown in table (1), the X-RD results give the major, minor and trace minerals. Results show that the major mineral is calcite (CaCo3), Dolomite ($CaMg(CO_3)_2$), and Quartz(SiO_2), with minor and trace concentrations.

Table (2) represents concentrations of Al, Ca, K, Bi, Pb, andTh, measured by Atomic AbsorptionAnalyzer. Results give concentrations in percent and ppm ranging from 0.10 to 0.68, 3.16 to 33.53, and 0.13 to 0.31 in %, and <10, <7.5 to 1756.38, and <1 in ppm respectively.

Table (3) represents the activities concentrations Bq/.kg dry weight of the ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U and ^{40}K for the samples.

Of all the 6 samples measured in this study, 6 F.ss. appears to have the highest concentrations of ^{238}U, also, 6 F.ss has the highest concentrations of ^{232}Th. Whereas the concentrations of ^{40}K vary with the type of the rocks. Three values for ^{235}U detected.

For the ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U and ^{40}K, the activities concentrations Bq/.kg dry weight ranged from 176.10$\pm$0.07 to 222.86$\pm$0.21, 28.34$\pm$0.11 to 231.04$\pm$0.05, 08.66$\pm$0.17 to 137.84$\pm$0.04, 09.35$\pm$0.03 to 12.69$\pm$0.03, 78.27$\pm$0.23 to 202.21$\pm$0.04 with mean values 199.18$\pm$0.17, 109.01$\pm$0.07,37.96$\pm$0.10, 11.47$\pm$0.02, and 123.57$\pm$0.13 respectively.

For theRa_{Eq} (Bq/kg) ranged from 59.193 to 438.615 with mean value 172.810 which is lowr than *370Bq/kg*, the permissible limit **(UNSCEAR, 2000).** The mean values of the annual effective dose (D_{eff}(mSv/y) was found to be less than one (0.096) which is within the worldwide mean values($<$1for D_{eff} (mSv/y) **(UNSCEAR, 2000).**

Table (1): The mineral constituents of six samples of sedimentary rocks analyzed by XRD spectrometer, (Leet*et al*., 1982, and Mineral Data, 2012

Samp No.	Major	Minor	Trace
1-Tu	CALCITE$(CaCO_3)$	QUARTZ(SiO_2)	CLINOCHLORE$(MgFe^{2+})_5Si_3Al_2O_{10}(OH)_8$, DOLOMITE$(CaMg(CO_3)_2)$, GYPSUMCa$(SO_4)\bullet 2(H_2O)$, MONTMORILLONITE $Na_{0.2}Ca_{0.1}Al_2Si_4O_{10}(OH)_2(H_2O)_{10}$, OFFRETITE$K_{1.1}Ca_{1.1}Mg_{0.7}Al_{5.2}Si_{12.8}O_{36}\bullet 15.2(H_2O)$, PARGASITE $NaCa_2Mg_3Fe^{2+}Si_6Al_3O_{22}(OH)_2$
2-Li	DOLOMITE$(CaMg(CO_3)_2)$	CALCITE$(CaCO_3)$	ALBITE$(NaCaAl\ Si\ _3O_8)$,BIOTITE$(K(MgFe^{2+})_3AlSi_3O_{10}(OH\ F)_2)$,CLINOCHLORE$(MgFe^{2+})_5Si_3Al_2O_{10}(OH)_8$, MONTMORILLONITE $Na_{0.2}Ca_{0.1}Al_2Si_4O_{10}(OH)_2(H_2O)_{10}$, PARGASITE $NaCa_2Mg_3Fe^{2+}Si_6Al_3O_{22}(OH)_2$, QUARTZ$(SiO_2)$, ROGGIANITE $Ca_2Be(OH)_2Al_2(Si_4O_{13})\bullet 2.4(H_2O)$
3-Ka	CALCITE$(CaCO_3)$, QUARTZ(SiO_2)	DOLOMITE$(CaMg(CO_3)_2)$	ALBITE$(NaCaAl\ Si\ _3O_8)$, CLINOCHLORE$(MgFe^{2+})_5Si_3Al_2O_{10}(OH)_8$,GYPSUMCa$(SO_4)\bullet 2(H_2O)$, MAGNETITE$(Fe^{3+}{}_2Fe^{2+}O_4)$, NONTRONITE $Na_{0.3}Fe^{3+}{}_2Si_3AlO_{10}(OH)_2\bullet 4(H_2O)$,PALYGORSKITE $Mg_{1.5}Al_{0.5}Si_4O_{10}(OH)\bullet 4(H_2O)$,PARGASITE $NaCa_2Mg_3Fe^{2+}Si_6Al_3O_{22}(OH)_2$
4- K.do	CALCITE$(CaCO_3)$, DOLOMITE$(CaMg(CO_3)_2)$	QUARTZ(SiO_2)	ALBITE$(NaCaAl\ Si\ _3O_8)$, BIOTITE$(K(MgFe^{2+})_3AlSi_3O_{10}(OH\ F)_2)$, CLINOCHLORE$(MgFe^{2+})_5Si_3Al_2O_{10}(OH)_8$, GYPSUMCa$(SO_4)\bullet 2(H_2O)$, MAGNETITE$(Fe^{3+}{}_2Fe^{2+}O_4)$, MONTMORILLONITE $Na_{0.2}Ca_{0.1}Al_2Si_4O_{10}(OH)_2(H_2O)_{10}$, PALYGORSKITE $Mg_{1.5}Al_{0.5}Si_4O_{10}(OH)\bullet 4(H_2O)$
5-Tudo	DOLOMITE$(CaMg(CO_3)_2)$	ALBITE$(NaCaAl\ Si\ _3O_8)$, CALCITE$(CaCO_3)$	AUGITE$(Ca,Na)(Mg,Fe,Al,Ti)(Si,Al)_2O_6$, BIOTITE$(K(MgFe^{2+})_3AlSi_3O_{10}(OH\ F)_2)$, CLINOCHLORE$(MgFe^{2+})_5Si_3Al_2O_{10}(OH)_8$, GYPSUMCa$(SO_4)\bullet 2(H_2O)$, MONTMORILLONITE $Na_{0.2}Ca_{0.1}Al_2Si_4O_{10}(OH)_2(H_2O)_{10}$, NONTRONITE $Na_{0.3}Fe^{3+}{}_2Si_3AlO_{10}(OH)_2\bullet 4(H_2O)$, PARGASITE $NaCa_2Mg_3Fe^{2+}Si_6Al_3O_{22}(OH)_2$, QUARTZ$(SiO_2)$
6-F.ss.	QUARTZ(SiO_2)	CALCITE$(CaCO_3)$	AUGITE$(Ca,Na)(Mg,Fe,Al,Ti)(Si,Al)_2O_6$,ALBITE$(NaCaAl\ Si\ _3O_8)$, BIOTITE$(K(MgFe^{2+})_3AlSi_3O_{10}(OH\ F)_2)$, CLINOCHLORE$(MgFe^{2+})_5Si_3Al_2O_{10}(OH)_8$DOLOMITE$(CaMg(CO_3)_2)$, HALITE$(NaCl)$,MAGNETITE$(Fe^{3+}{}_2Fe^{2+}O_4)$, MONTMORILLONITE $Na_{0.2}Ca_{0.1}Al_2Si_4O_{10}(OH)_2(H_2O)_{10}$, NONTRONITE $Na_{0.3}Fe^{3+}{}_2Si_3AlO_{10}(OH)_2\bullet 4(H_2O)$, PARGASITE $NaCa_2Mg_3Fe^{2+}Si_6Al_3O_{22}(OH)_2$, SCOLECITE $CaAl_2Si_3O_{10}\bullet 3(H_2O)$

Table (2): Concentrations of Al, ,Ca, K, Bi, Pb, andTh, for the samples measured byAtomic Absorption Analyzer.

Samp. No.	Elements	Al	Ca	K	Bi	Pb	Th	
	D L	0.05	0.05	0.05	10.00	7.50	1.00	4.10
	Units	%	%	%	ppm	ppm	ppm	Bq/kg
1-Tu		0.31	33.53	0.24	<10	16.77	<1	<4.10
2- Li		0.10	20.41	0.18	<10	1756.38	<1	<4.10
3-Ka		0.38	25.73	0.31	<10	<7.5	<1	<4.10
4 -K.do		0.33	23.67	0.25	<10	298.43	<1	<4.10
5-Tudo		0.12	20.36	0.19	<10	<7.5	<1	<4.10
6 -F.ss.		0.68	3.16	0.13	<10	89.69	<1	<4.10

Table (3): The activities concentrations Bq/.kg dry weight of the ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U and ^{40}K for the samples.

Samp. No.	Lat. and Long.	^{238}U	^{226}Ra	^{232}Th	^{235}U	^{40}K
1-Tu	N:28^{0}41'20.6" E:44^{0}40'58.5"	176.10±0.07	28.34±0.11	14.61±0.18	ND	129.36±0.35
2- Li	N:28^0 59' 01.3" E:43^0 35'16.1"	189.62±0.18	75.66±0.03	15.27±0.06	ND	78.27±0.23
3-Ka	N:28^{0}42'08.4" E:43^0 52'22.7"	240.10±0.16	66.85±0.06	33.65±0.04	12.36±0.03	202.21±0.04
4- K.do	N:28^{0}47'51.1" E:43^{0}45'41.0"	189.10±0.18	135.04±0.12	17.71±0.11	09.35±0.03	110.89±0.03
5 -Tudo	N:28^{0}54'03.4" E:43^0 42'06.5"	177.31±0.19	117.14±0.08	08.66±0.17	ND	84.80±0.08
6-F.ss.	N:26^{0}04'09.1" E:45^{0}19'59.3"	222.86±0.21	231.04±0.05	137.84±0.04	12.69±0.03	135.89±0.03
Min.		176.10±0.07	28.34±0.11	08.66±0.17	09.35±0.03	78.27±0.23
Max.		222.86±0.21	231.04±0.05	137.84±0.04	12.69±0.03	202.21±0.04
Mean		199.18±0.17	109.01±0.07	37.96±0.10	11.47±0.02	123.57±0.13

Table (4):TheRa$_{Eq}$ (Bq/kg), D(nGy/h), and D$_{eff}$(mSv/y) For sedimentary rocks.

Samp. No.	Ra$_{Eq}$Bq/kg)	D(nGy/h)	D$_{eff}$(mSv/y)
1-Tu	59.193	27.221	0.033
2- Li	103.523	47.387	0.058
3-Ka	130.540	59.500	0.073
4- K.do	168.904	77.632	0.095
5-Tudo	136.053	62.826	0.077
6 -F.ss.	438.615	195.567	0.240
Min.	59.193	27.221	0.033
Max.	438.615	195.567	0.240
Mean	172.810	78.33	0.096

Conclusion

Three techniques, the analysis by X-RD, AtomicAbsorption Analyzer. and High-resolution γ-ray spectroscopy were applied to study sedimentary rock samples, The obtained results show that the major mineral is calcite (CaCo3), Dolomite (Ca Mg$(CO_3)_2$), and Quartz(SiO_2), the concentrations in percent for Ca are higher than Al and K, while the concentrations in ppm for Pb appear high. The activities concentrations Bq/.kg dry weight of the ^{238}U are higher than ^{226}Ra, ^{232}Th, and ^{40}K. The mean value of Ra$_{Eq}$ (Bq/kg) and The mean values of the annual effective dose (D$_{eff}$(mSv/y) were found to be within the worldwide mean values(370 Bq/kg and <1for D$_{eff}$ (mSv/y) (UNSCEAR, 2000).

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