Natural Radioactivity, Major and Trance Elements Measurements of Surface soil from Al-Madinah Al-Monawarah, Saudi Arabia.

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Abstract: Nine surface soil samples were collected from several localities around Al-Madinah Al-Monawarh and one from the city itself. Samples were analyzed for Aluminum (Al), Iron (Fe) and Calcium (Ca) concentrations in percent as major elements constituents. Lead (Pb) and Arsenic (As) in ppm as trace elements. The concentrations range for Al from (4.48 - 7.65 %), Fe from (3.08 - 4.92 5 %), Ca from (1.66 - 10.60 %), Pb from (14 -27 ppm) and As from (10.5 -30.7 ppm). Major, minor and trace minerals were analyzed, they found to be in the order Quartz, Albeit and Calcite as major minerals. Gamma-ray spectrometer based on HPGe crystal was applied for the concentrations in Bq/kg dry weight, ^{238} U which ranges between LDL and 12.3, ^{226}Ra from 7.01 and 15.55 Bq/kg., ^{228}Ra ranges from 5.23 to 21.8 Bq/kg and ^{40}K concentrations range from 64.6 to 754.2 Bq/kg. The man-made ^{137}Cs was detected in the nine samples and LDL in Al- Madinah Al- Monawarah sample. The radium equivalent Bq/kg and the absorbed dose rate nGy/h were calculated for each sample. It was found that the values of the absorbed dose are in the accepted range recorded by EPA (Environmental Protection Agency).

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1. Introduction

Study of radioactivity levels and concentrations of natural and man-made radionuclides have been done all over the world. In Egypt Ibrahiem *et al* (1993), gave a base map for the radioactivity in the Delta and middle Egypt, they measured the absorbed dose one meter above the ground in each point. Also; Ibrahiem *et al* (1995), studied sediments and surface area of Nasser lake area by both neutron activation analysis and gamma-ray spectroscopy techniques. Amaral (2000), in Portugal studied gamma-ray spectrum and dose rate In Situ, also studied the type and composition of the different rocks, as well as the mechanical, chemical and biological properties for each type of soil. Melo *et al* (2000), studied the severe internal dose in Brazil from the high radioactivity concentrations due to the concentrations of uranium and thorium in the studied area. Morton *et al* (2006), measured the natural radioactivity concentrations 232Th, 238U and 226Ra series and ^{40} K, as well as the man made ^{137}Cs. Also; they studied the salinity level in the soil from the series of the black plateau resulting from the flood of Virgin River, south-east of Nevada state USA. Florou *et al* (2007), studied the effect of the external dose intake for areas of high radioactivity levels in three islands from Greece of volcanic origin. In those areas many geothermal springs gives gases as carbon mono-oxide, carbon dioxide as well as radon. **Baykara, O. and Dogru M**. (2000). studied 72 soil samples from the northern and eastern regions of Anatoly of Turkey for the concentrations of radioactivity and dose rate. Santor *et al* (2009), analyzed 78 soil samples for the concentrations of ^{226}Ra and ^{228}Ra, Saidou *et al* (2011), studied a site for uranium mining in Cameron to plot a base line map for Poli province, they found that the concentrations of radionuclides and the absorbed dose are slightly higher than the world average.

The aim of this work is to study the radioactive hazards in Al- Madinna Al- Monawarh and the environs around.

2. Geology of The Study Area

The study area lies east of the Red sea between 24° 22' 27" and 24° 32'16" latitude north and 39° 31' 36" and 39° 43' 11" longitude east and elevation 625m above the sea level.

Rock types in the study area are :

Volcanic rocks, Silicic volcanic rocks, pyroclastic rocks, Sandy rocks and, Sedimentary rocks, from the erosion of volcanic rocks, Breccia Andesitic, Basalt, Sandy regions, Limestone, Harrat areas, Felsic rocks, Quaternary deposits Harrat Khyber and Harrat Hirmah. Al-Madina also contains valleys from sand and clay, and sediments from erosion of granite, (Saudi Geological Survey, 2010).

Sampling and Samples Preparation

The study area was divided to 10 parts representing the center, northern, north eastern,

western and southern localities of Al- Madinah Al-Monawarh (Fig.1). A sample was collected from each location. Samples were collected from the wadi (Valley) deposits by a template 30x30x15 cm. Samples were packed in a polyethylene bags, then labeled. Remnants of plants, weeds and rocks were removed, then soil samples were grinded, sieved with a 1mmx1mm mesh sieve, mixed for homogeneity. Samples were dried to 80 °C not to lose the volatile ^{137}Cs or the natural polonium. A 640 cc of the dried sample were weighed then stored for one month in a polyethylene Marinelli beaker, for gamma-ray spectrometry, to reach secular equilibrium between ^{226}Ra and ^{232}Th and their progeny. 10 gm of the dried sample were used for the analysis by atomic absorption for the Al, Fe, Ca, Pb and As concentrations. Also; 10 gm for XRD to identify the mineral composition. Sampling was done obeying methods adopted by Her Majesty's Office, the UK Atomic Energy Authority and UK Nirex Ltd, (RADREM, 1980).

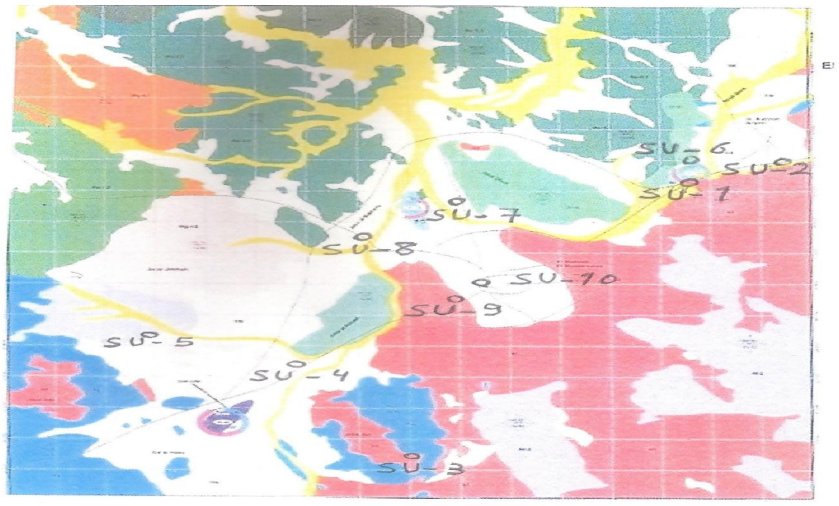

Fig. (1) Sample location in the studied area

Analytical Techniques

An X-ray diffraction spectrometer model Burker XRD D8 Advance was applied for the minerals' identification. Also; an inductively coupled plasma atomic absorption spectrometer of A-Analyst 700 model Perkin Elmer OPTIMA 4000 DV series, was used for the concentrations of Pb and As ppm and Al, Fe and Ca %. A gamma ray spectrometer based on a HPGe crystal of the vertical type Canberra model number GC2520, cryostat Canberra model 7500SL, FWHM 1.06 keV for the transition 122 keV ^{57}Co and 2.0 keV for the transition 1332.5 keV of ^{60}Co, peak to Compton ratio 53:1, relative efficiency 27.1%, were applied for the concentrations of the natural ^{232}Th and ^{226}Ra series and ^{40}K and the manmade ^{137}Cs. Multichannel analyzer of 8K ADC (analogue to digital converter), Genie 2000 program, was used for spectrum analysis.^{152}Eu in 640cc Marinilli beaker and natural KCl with three different concentrations were used for measurements of absolute calibration, as well as ^{226}Ra point source normalized to the same configuration.

Energies (keV) of 295.2 (20.1) and 351.9 (38.3) ^{214}Pb and 609.3 (49.9), 1120.3 (16.2) and 1764.5 (16.0) ^{214}Bi were used for the ^{226}Ra series. 338.4 (13), 911.16(30.3), and 968.97(18.3) ^{228}Ac and 727.25 (8.1) ^{212}Bi, also 583.02(33.2) and 2614.48(35.9) ^{208}Tl for the ^{232}Th series (Saito & Moriuchi, 1985). 1460.8(10.7) for the ^{40}K and 661.65 (89.9) for the ^{137}Cs (Holden; 2003).

3. Result and Discussion

Table (1) represents XRD analysis results it shows the major, minor, trace and minerals. The major minerals are: the quartz (SiO_2) which is dominant, albite ($NaAlSi_3O_8$), then calcite ($CaCO_3$), while the microcline is the least. The minor mineral is the clinochlore [$(MgFe^{2+})17Si_{20}O_{54}(OH)_6$].

Table (2) gives concentrations of Al, Fe and Ca % and Pb and As ppm. The high Al content interpret the presence of albite and microcline minerals, while the high Ca indicates the presence of Calcite in the studied samples. The stable lead ^{208}Pb, Pb and ^{207}Pb) ranged from 14.00 to 27.00 ppm which may not be radiogenic Pb as the relation with either ^{226} Ra or^{228} Ra are not clear (Table 2 and3). Calcium,

(^{40}Ca, ^{42}Ca, ^{43}Ca, ^{44}Ca, ^{46}Ca and ^{48}Ca), they are all stable, ranged from 1.66 to 10.60%, the highest percent is noticed in sample SU-3 in which Calcite is the dominating mineral (Table-1). Arsenic (^{75}As) ranged from 10.50 to 30.70 ppm. Iron (^{54}Fe,^{56}Fe,^{57}Fe and ^{58}Fe) ranged from 3.08 to 4.92% which represents moderate concentrations and reflected in the presence of clinochlore (Mg, Fe^{2+}, Al) $(SiAl)_2O_5(OH)_4$ mineral as minor in all of samples except SU-4 in which the iron is represented in the trace minerals magnetite (Fe_3O_4) and augite (Mg, Fe^{2+}). From the previously mentioned types of minerals and the major elements(Al, Fe, and Ca) distributions in the studied samples, it can be strongly conclude that the studied soil sediment are mirror- image to the exposed rocks in the studied area.

Table (1) Identified minerals in different soil samples

Sample No.	Major	Minor	Trace
SU-1	Quartz, Albite	Microcline, Calcite Chlinochlore	Ludlokite, Biotite, Augite, Magnetite
SU-2	Quartz, Albite	Calcite, Chlinochlore	Geigerite, Microcline, Reevesite, Biotite, Magnetite
SU-3	Calcite, Quartz	Albite, Chlinochlore	Magnetite, Tinaksite, Dundasite, Geigerite
SU-4	Quartz, Albite	……..	Microcline, Calcite, Kaolinite, Faugasite, Augite, Magnetite
SU-5	Quartz, Albite, Microcline	Chlinochlore	Calcite, Volkovskite, Magnetite
SU-6	Quartz, Albite	Chlinochlore	Calcite, Augite, Magnetite, Pargasite, Biotite, Ludlokite
SU-7	Quartz, Albite	Calcite, Chlinochlore	Reevesite, Biotite, Magnetite, Pargasite
SU-8	Quartz, Albite	Chlinochlore, Calcite	Pargasite, Augite, Biotite, Magnetite
SU-9	Quartz, Albite	Calcite, Chlinochlore	Biotite, Augite, Pargasite, Microcline
SU-10	Quartz, Calcite, Albite	Chlinochlore	Pargasite, Biotite, Tunisite, Offeretite, Magnetite

The distribution of natural and man – made radionuclides are shown in table (3). For the $^{238}U - ^{226}Ra$ series concentrations of protactinium-234m (^{234m}Pa) ranged from LDL to 12.3, while ^{226}Ra from 7.01 to 15.55Bq/kg. This shows disequilibrium in the series which is very clear from the ratios of ^{226}U/^{238}Ra. These ratios are less than one in six samples (SU-1, 3, 4, 5, 8 and 9) and higher than one in four samples (SU-2, 6, 7 and 10). This may be due to difference in chemical behaviors between ^{238}U and ^{226}R. For the $^{232}Th - ^{228}Ra$ series concentrations ranged from 5.23 to 1.8, disequilibrium can't be observed in the series.

Table (2) Results of some major and trace elements in the different soil samples.

Elements	Al	Fe	Ca	Pb	As
DL.	**0.25**	**0.05**	**0.05**	**1.00**	5.50
Units	**%**	**%**	**%**	**Ppm**	Ppm
SU – 1	6.98	4.72	3.75	19.00	10.50
SU – 2	7.65	4.68	4.98	18.00	12.30
SU – 3	4.48	3.08	10.60	14.00	29.10
SU – 4	6.18	4.60	2.78	16.00	30.70
SU – 5	6.84	3.08	1.60	16.00	17.26
SU – 6	6.61	4.60	3.48	16.00	17.30
SU – 7	6.79	4.68	3.51	19.00	13.41
SU – 8	6.73	4.92	3.28	15.00	12.20
SU – 9	6.80	4.32	5.04	27.00	15.70
SU – 10	6.04	4.14	8.00	25.00	28.60

The main reasons for disequilibrium are the differences in chemical and physical properties of the elements in the series, weathering, radon as a gas can escape from the sample, also the emission of beta or alpha particles may lead the residual nucleus leaving the crystal recoil. ^{40}K concentrations ranged from 64.64 Bq/kg (2.2%) (sample SU-3) to 754.2 (25.5%) in sample SU-5 (Table 3). The concentrations of ^{40}K reflects the minerals domination. The lowest^{40} K in sample SU-3 agree with Albeit and clinochlore as minor minerals and Quartz with Calcite as major minerals. The highest ^{40}K concentrations in (sample

SU-5) coincide with Albite and microcline as major minerals. ^{137}Cs was found in four samples (SU-2, 3, 6, 7) with concentrations around 3.0 Bq/kg dry weight, these samples represent the eastern half of Al-Madinah Al- Monawarah, while the western side (SU-4, 5, 8, 9) show lower concentrations of ^{137} Cs. The central part (SU-10) show no ^{137} Cs concentration.

Table (3) Concentrations of radionuclides for the natural $^{238}U - ^{226}Ra$ and $^{232}Th - ^{228}Ra$ series, ^{40}K and the man-made ^{137}Cs in Bq/kg dry weight.

Sample code.	Concentration Bq/kg dry weight					Conc.%
	U-Ra series		**Th series**	**K-40**	**Cs-137**	Natural-K
	^{214m}Pa	**Ra-226**	**Ra-228**			
SU-1	LDL	7.75 ±0.057	6.84 ±0.078	262.69 ±1.42	0.94 ±0.078	8.875 ±0.048
SU-2	12.1±0.56	10.54 ±0.051	13.95 ±0.09	255.30 ±0.95	3.09 ±0.099	8.63 ±0.032
SU-3	4.6±0.51	7.71±0.05	6.46 ±0.055	64.64 ±0.033	3.6 ±0.16	2.183 ±0.011
SU-4	3.56±0.29	10.08 ±0.044	18.79 ±0.13	301.32 ±1.14	2.51 ±0.085	10.18 ±0.039
SU-5	LDL	15.55 ±0.064	21.84 ±0.13	754.21±2.31	1.7 ±0.06	25.48 ±0.078
SU-6	11.42 ±0.52	7.01 ±0.037	8.21±0.054	328.36 ±1.22	3.13 ±0.095	11.093 ±0.041
SU-7	12.3±0.75	10.21 ±0.066	14.86 ±0.13	312.40 ±1.57	3.181 ±0.133	10.554 ±0.053
SU-8	LDL	8.45±0.063	8.02 ±0.099	409.74 ±2.28	1.484 ±0.102	13.842 ±0.077
SU-9	4.8±0.47	8.72±0.062	5.23 ±0.054	265.92 ±1.42	0.91 ±0.071	8.984 ±0.048
SU-10	9.7±0.7	7.60±0.045	6.16 ±0.058	209.38 ±1.09	*LDL	7.074 ±0.037

*LDL: Lower than Detection Limit

The Radium equivalent Ra_{eq} is calculated from equation (1) Tufail *et al* (2006) :

$$\mathbf{Ra_{eq} = A_{Ra} + (A_{Th} \times 1.43) + (A_K \times 0.077)} \quad (1)$$

Where; A_{Ra}, A_{Th} and A_K are concentrations Bq/kg for radium, thorium and potassium respectively.

The absorbed dose nGy/h is given by the equation (2) Quindos *0*, (2004):

$$\mathbf{D = C_{Ra} A_{Ra} + C_{Th} A_{Th} + C_K A_K} \quad (2)$$

Where: C_{Ra}, C_{Th} and C_K are the conversion factors Bq/kg to nGy/h for radium, thorium and potassium respectively.

Table (4) represents the values of the Ra_{eq} and the absorbed dose. The Ra_{eq} Bq/kg dry weight ranged from 21.93 to 104.83, less than 370 adopted by EPA for the permissible value. Using the conversion factors from Bq/kg to nGy/y Qunidos *et al* (2004) the adsorbed dose one meter above the ground, ranged from 10 nGy/h (>1mmGy/y) to 52.2 nGy/h (o.46 mGy/y), it is within the permissible value given by EPA and UNSCEAR (2000).

From the previously mentioned results, it can be concluded that the studied localities around Al Madinah Al Monawarah are hazardless except for the fall out that comes from anywhere. Al- Madinah Al-Monawarah itself is safe.

Table (4) the radium equivalent Bq/kg and the absorbed dose nGy/h for the different samples.

Sample Code	Ra_{eq} Bk/kg	Absorbed dose nGy/h
SU-1	37.76	18.79
SU-2	50.15	23.89
SU-3	21.93	10.05
SU-4	60.15	28.49
SU-5	104.86	52.17
SU-6	44.03	22.07
SU-7	55.53	26.72
SU-8	51.47	26.10
SU-9	36.67	18.43
SU-10	32.53	16.04

Conclusions

Al- Madinah Al- Monawarah as an Islamic major seasonal destination city encourages the scientists to carry out their researches in different branches of sciences. This work is one of those branches which is connected with the radioactivity and hazards for its surface soil as well as the surrounding areas. The results of this work indicated that the study area is safe for the radiological levels, for either to live or to cultivate if the type of soil is suitable. The only hazard is coming from the fall out ^{137}Cs especially in the eastern parts of the studied area.

Table 5 shows that the present results are within the published values.

Table (5) Comparison of isotopes concentrations in Bq/kg dry weight concentrations in the present work and some published results.

Reference / Nuclide-Series	Present work	Ibrahiem *etal.* 1993	Ibrahiem *et al.*, 2003	Al-Garni Z., 2008
U-238−Ra-226	7.01-15.55	5.2-63.7	31-55	9.2-18.1
Th-232−Ra-228	5.23-21.8	2.5-95.6	2.4-3.2	9.5-22.6
K-40	64.6-754.2	29-653	65-1046	378.2-592.6

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