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IMPACTS OF URANIUM AND SOME HEAVY ELEMENTS CONTENTS ON THE GROUND WATER QUALITY OF EL-MISSIKAT AREA, EASTERN DESERT, EGYPT

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ABSTRACT

Uranium beside some heavy elements contents in some rock, ground and well water samples of El-Missikat area, Eastern Desert, Egypt have been analyzed to decide the water suitability in El-Missikat area for human and animals uses. For this purpose, the uranium analysis has been performed using different techniques include UV/VIS Spectrophotometry, N_2-Laser Fluorimetric (UA-3) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) techniques. In this work, ten different samples varied between rocks and water samples were collected from El-Missikat locality (four rock samples, three ground water samples beside three well water samples) and chemically analysed. The obtained results indicated that, the average concentrations of uranium and heavy elements (Pb and Cd) determined by ICP-MS in the water samples were higher than the recommended permissible limits. The acheived results show that, the well water has risky levels of uranium and heavy elements contents which severly impacts the health of users. The ptesent study recommends that, the well water in the Missikat area should not be used as drinking water source.

INTRODUCTION

Uranium as a metallic element is not found in the earth crust but usually occurs in ore minerals as, carnotite, uraninite and pitch-blende. The uranium in the earth's crust has been estimated to be around 3 mg/kg (Padam et al., 1996). Naturally uranium isotopes, are U^{238} (99.27%), U^{235}(0.72%) and U^{234}(0.005%), while the other isotopes are synthesized (Wall and Krumholz, 2006. Uranium found in various oxidation state as U(VI) in the oxidizing medium and U(IV) in the reducing medium (Koulouris et al., 2000). Chemically uranium associates with oxygen as, uranyl ion (UO_2^{2+}) which easily solubilizes by the ground or rain waters. The uranium solubilization depends on the water's chemical profile beside its oxidation state and is higher under oxidizing conditions than reducing conditions (Mehra et al., 2007; Mirza et al., 2013; Sunday et al., 2013 and Abojassim and Mohamed, 2017). On the other hand, the heavy metals even at ppm levels introduced waters through mining and industrial enterprises as tanneries, painting, batteries, fertilizer is hazardous species (Yadav et al., 2019). Heavy metals as, Co, Cr, Cu, Mg, Mn, Zn, Ni and Pb species are accumulate in human bodyand cause different diseases as, liver failure, headache, neurotoxicity or even death (Furness,2017, Lawal, 2017). Therefore, it is crucial to re-

move those species using technologies including ion exchange (Dabrowski et al., 2004), adsorption (Shahat et al., 2015), precipitation (Fu and Wang, 2011), coagulation (Al-Shannag et al., 2015), electrochemical (Ebbers et al., 2015), photochemical (Crespo-Otero and Barbatti, 2011) and membrane separation (Huang et al., 2017), ultra-filtration (Huang et al., 2016), surfactant micelles (Huang et al., 2017) beside nanoparticles (Chen et al., 2017). Among those methods, adsorption is regarded the most popular, economic and effective method for the heavy metals removal (Lei et al., 2017).

Based on the biodegradability, uranium beside heavy metals are not found in their nature, which heap up in vital human organs and exert progressively growing toxic actions (ASTDR, 1999 and Donia et al., 2009). Naturally the radioactive nuclides enter the human body through two main pathways, by inhalation of the radioactive gases like radon, and ingestion of primordial radionuclides like K^{40}, Th^{232}, $U^{238}$and Ra^{226}. The survey results indicated that, uranium in soils or phosphate fertilizers can be transfer to water, plants, food supplements and then to animals and humans beside is introducing the human body through ingestion with the drinking water as part of the food chain (Mehra et al., 2007, Mirza et al., 2013 and Sunday et al., 2013). Uranium causes many cancer diseases during concentration in specific human organs specially bones, liver and blood (Mehra et al., 2007). The greatest risk cause by uranium is the likelihood of damage to the kidney structure then causes acute renal failure (Sunday et al., 2013). Practically, long-run intake of uranium makes an increase in the risks of contracting damage in addition cardiovascular diseases (Aswood et al., 2017), whereas the experimental evidence indicated that, the respiratory and reproductive systems are also affected by the uranium exposure (WHO, 2004). Lariviere et al., (2013) found a strong correlation between the uranium concentrations in drinking water and in human bones. The elevated uranium levels in the drinking water have been associated with many epidemiological studies such as leukemia, stomach and urinary track cancer as well as increased diastolic and systolic blood pressure (Auvinen et al., 2002 & 2005 and Zamora et al., 2009). For these purposes, measurements the radioactivity in water sources have been performed and takes attention in many parts of the world, mostly for assessment of the doses and risk resulting from consuming water (Oliveira et al., 2001, Bhalara et al., 2014). The levels of uranium as radioactive species in ground waters are depend mainly on the uranium bearing rocks (Ajayi and Adesida, 2009).

In this study, the uranium mineralization at El-Missikat area occurrences is represented essentially by pitchblende and uranophane as secondary uranium products with sulphides and fluorites as associated gangue minerals (Attawiya, 1984, Ahmed, 1991, Amer et al., 2005 and Abu-Deif and El-Tahir, 2008). The uranium mineralization (mainly uranophane) is associated with jasperoid veins occur within shear zones (Raslan, 2008).

El-Missikat area has residents (Bedouin and Miners) and animals which need safe water sources for the daily uses. Due to the scarcity of water recourses in El-Missikat area, the population tends to use the wells, rainwater and the pools of flood water as drinking water source. El-missikat area had been geologically studied by several authors (Abu Deif and El-Tahir, 2008, Omar, 2010, Arafa and Omar, 2011 and Omar et al., 2012).

The present work deals with the field geology and some chemical methods to show the impact of uranium, cadmium and lead as heavy elements on the quality of floor water and ground water of El-Missikat area. In addition to determines the uranium and some heavy elements contents (Pb and Cd) of rocks and some water samples which passing through the shear zone of El-Missikat area to assess the suitability of the water sources for human uses. It is also aimed to assess the impact of

the mineralized shear zone in the studied area on the quality of the collected water samples. In the present study three ground water, three well water samples beside four rock samples were collected and analyzed in Nuclear Materials Authority (NMA) and Chemical Warfare Management laboratories.

FIELD GEOLOGY AND SAMPLING

Gabal El-Missikat is one of the most important uranium occurrences in the Eastern Desert, Egypt. It lies at a distance of about three kilometers to the south of landmark Km. 85 on the Qena-Safaga highway (Fig.1). The uranium mineralization of the study area is connected to smoky and jasperoid siliceous veins in reactivated tension fractures of shear zones crossing the granite in ENE direction and dipping between 60-70º toward SE (Bakhit, 1978).

Geologically, the rock units of El-Missikat area is arranged from the oldest to the youngest as the following; granitic gneiss, amphibolites, calc-alkaline granitoids, alkaline granites, Nubian sandstone and Wadi alluvial deposits, (Fig.2). These rocks are dissected by several types of dykes in different directions with presence of pegmatite and fluorite occurrence.

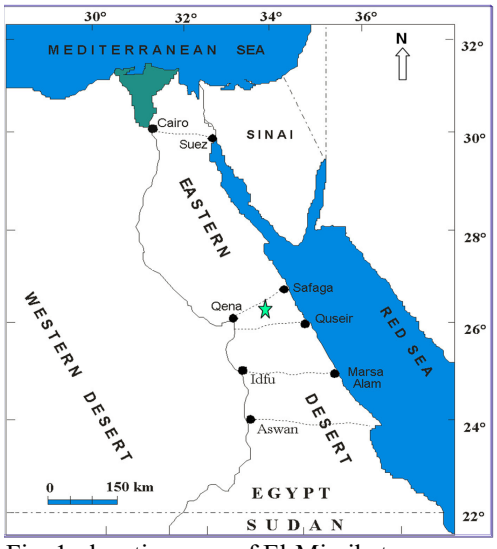

Fig. 1: location map of El-Missikat area

MATERIALS AND METHODS

Samples Collection

The collected rock samples are named R_1, R_2, R_3 and R_4 and ground water samples are G_1, G_2 and G_3 while W_1, W_2 and W_3 are the well water samples as indicated in Table (1) and plotted on Figure (3). The water samples were collected and filtered through 0.45µ filter and then acidified to store in 100 ml standard polyethylene vials and packed for chemical analyses.

Analytical Procedures

The rock samples were subjected to major and trace elements contents analysis (Shapiro and Brannock, 1962). For this purpose, one gram from the fine rock samples was dissolved in a mixture of acids include, hydrochloric (10 ml), nitric (5 ml) and hydrofluoric (10 ml) till dryness and re-dissolved the resultant cake with 10 ml diluted HCl (10%) then completed up to volume with double distilled water. The obtained solution was directed to different analytical techniques. Oxides included, TiO_2, Fe_2O_3, MnO and P_2O_5 were determined spectrophotometrically while Na and K ions were determined photometrically. Calcium and magnesium species were estimated titrimetrically against 0.01M EDTA solution in presence of Eriochrome and murexide indicators at pH, 10 and 12 respectively. For Si and Al oxides analysis 0.1g fine ore sample is fused with 1g of NaOH in a nickel crucible on Bunsen flame. The fused sample is re-dissolved in 10 ml HCl (1:1) and closed up to a measuring volume for SiO_2 and Al_2O_3 estimation spectrophotometrically at 640 and 475 nm respectively. The uranium concentrations in rocks, water samples were analyzed in all experiments spectrophotmetrically at 650 nm using Arsenazo III dye as a complexing agent (Merczenko and Balcerzak, 2000). All colorimetric ex-

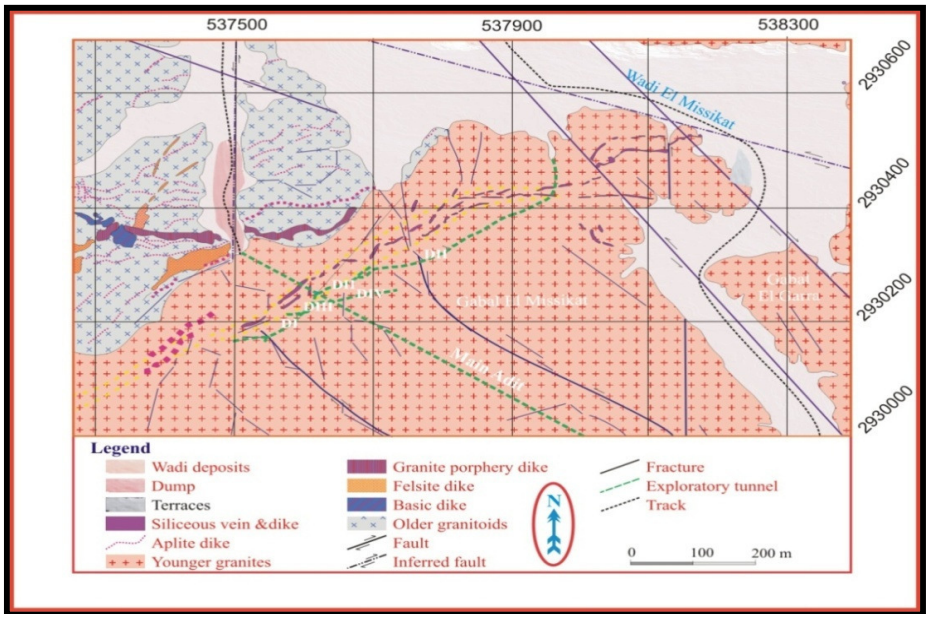

Fig. 2: Geological map of El-Missikat area (Omar et al., 2012)

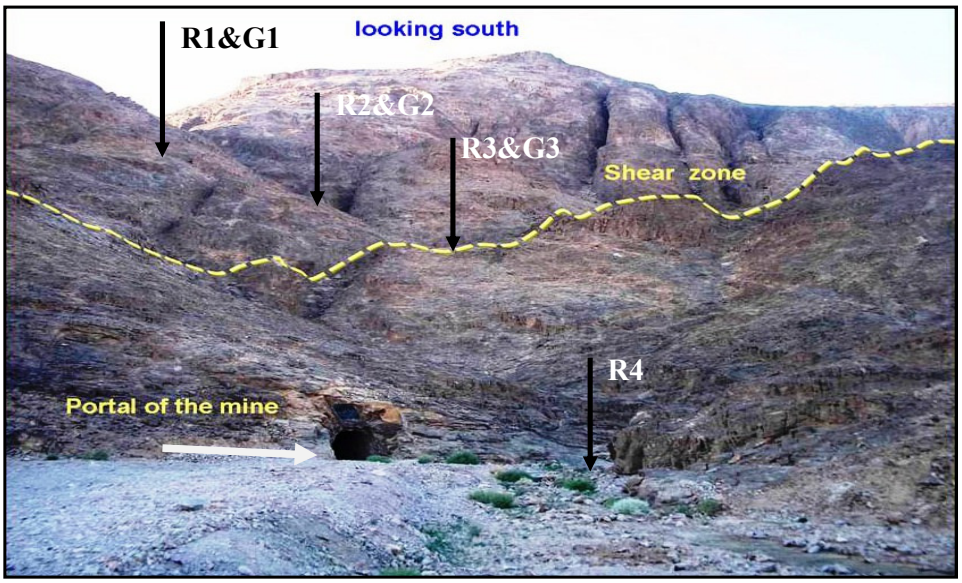

Fig. 3: Shear zone of El-Missikat area with the samples locations
(R: Rock samples G: Ground water samples)

Table 1: The studied samples in El-Missikat area

Sample type	Sample code
Rocks	R_1
	R_2
	R_4
Shear zone	R_3
Ground water	G_1
	G_2
	G_3
Water wells	W_1
	W_2
	W_3

periments were applied using Lambada UV/VIS spectrophotometer (Perkin-Elmer, USA) while pH values of water samples were adjusted with Digimed DM-21 (UK) pH meter. For control of instrument analysis stability, JG2as an international granite standard rock sample was continuously analyzed during the work.

Instrument optimization for stable signal, high sensitivity and low interference was performed daily on the work. Reference water with certified uranium content in all water samples analysis, the uranium concentration was less than 0.005 µg l^{-1} in addition blank, drift control and duplicate samples were analyzed at regular time intervals throughout the samples analysis. The uranium concentrations in all experiments were emphasized using N_2-Laser Fluorimetric technique (UA-3). A suitable sample volume (10-50 µl) was mixed with a buffer solution by the ratio of 2/3, completed up to 7 ml with double distilled water in quartz cell. The fluorescence of the measured standard uranyl solution in the cell was compared with the sample and internal standard measurements (Robbins, 1978, Tikoo and Murty, 1980). All the uranium analysis data were confirmed using ICP-MS Egilant Technologist 7700. On the other hand, the trace elements were determined using X-ray fluorescence [PHILIPS X'Unique II spectrometer]. For the studied rocks, mineral identification of representative technological sample (R_3 sample) was detected using PHILIPS PW 3710 X-ray diffraction (XRD).

RESULTS AND DISCUSSION

The chemical composition of the studied rocks (R_1, R_2, R_3 and R_4) Table (2) shows that, all samples have high percentage of SiO_2, Al_2O_3 and low percentage of Fe_2O_3, MgO, CaO and Na_2O while K_2O shows intermediate percentage. On the other hand, the trace elements contents (Table 3) and show that the concentrations of Cr, Zn, Zr, Rb, Y, U and Th are varied from low to moderate concentrations levels. These results indicated that ground water of the study area have high contents of uranium, thorium, lead, zinc, chromium and cadmium compared to the international permissible limits in the drinking water.

The mineral identification at the shear zone (R_3) using XRD analysis indicated that, the principle mineral constituents are quartz (SiO_2) and kaolinite [$AlSi_2O_5(OH)_4$] (Fig.4). From Table (3), it is observed that the concentration of uranium in the shear zone (R3) is 202 mgl^{-1}, indicated that no uranium mineral but uranium is mostly adsorbed upon the kaolinite mineral.

The low uranium content in well water samples cannot be detected spectrophotmetrically, but they detected by using ICP-MS analytical technique. On the other hand, the uranium contents in R_1, R_2, R_3 and R_4 rocks samples determined by using ICP-MS technique were 157, 180, 212 and 146 mg l^{-1} respectively. Based on the obtained uranium values it is noted that, the highest value is related to the shear zone in El-Missikat area, sample R_3 (212 mg l^{-1}) which matched well with field observations. The uranium contents in ground water samples (G_1, G_2 and G_3) show 27, 16 and 36 mg l^{-1} respectively in which the highest value

Table 2: Major oxides content (wt %) of the studied rock samples of El-Missikat area

Oxide / Rock Samples	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	MgO	CaO	Na_2O	K_2O	P_2O_5	L.O.I*
R_1	72.8	22.2	0.005	0.43	0.14	0.49	0.13	1.44	N.D	2.2
R_2	70.7	22.1	N.D*	0.35	0.10	0.51	1.6	2.55	0.005	1.9
R_3	71.9	22.8	N.D	0.42	0.13	0.50	0.14	1.93	0.01	2.5
R_4	72.5	22.1	0.01	0.23	0.10	0.53	0.10	0.67	N.D	3.9

L.O.I*: Total loss of ignition at 1000 °C N.D*: Not detected

Table 3: Trace elements content of the studied rock samples of El-Missikat area

Trace elements	Rock sample, (mg l^{-1})			
	R_1	R_2	R_3	R_4
Cr	61	60	71	67
Ni	9	7	10	11
Cu	10	14	12	14
Zn	62	64	65	64
Zr	102	111	108	104
Rb	534	530	538	535
Y	54	57	55	53
Ba	33	38	36	37
Pb	21	22	20	23
Sr	4	5	3	6
Ga	16	19	18	17
Nb	20	16	18	16
U	162	182	202	141
Th	60	32	20	15
Cd	10	20	9	5

was obtained in G_3 sample which related to the shear zone (Table 4). High uranium content in the water samples especially G_3 might be due to the leaching of uranium from the surrounding rocks especially at the shear zone in the study area. For the well water samples (W_1, W_2 and W_3), the uranium contents were around 0.2 mg l^{-1} (Table 4).

The drinking water guidelines relating to the radiological quality of drinking water. No change has been made to the recommended screening levels of 0.5 Bq/L for gross alpha and 0.5 Bq/L gross beta (corrected for potassium-40) and there is no change to the 1 mSv/year reference value (ICRP, 1993).

However, uranium is known to occur at higher concentrations, frequently in smaller supplies. For example, uranium concentrations of up to 700 µg/l have been found in private supplies in Canada (Moss et al., 1983;

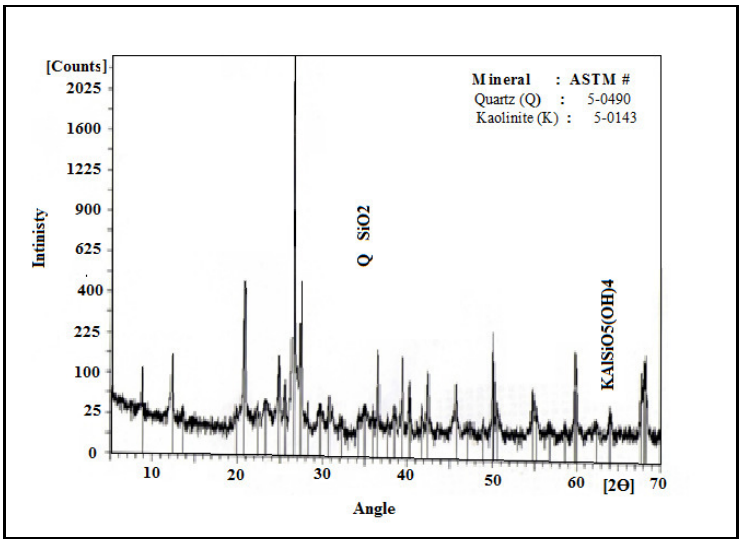

Fig. 4: XRD pattern of the shear zone rock sample (R_3) of El-Missikat area

Table 4: Uranium analysis comparison in rocks and water samples of El-Missikat area

Sample	Uranium concentration (mg l^{-1})		
	UA-3	Spectrophotometry	ICP-MS
R_1	153	168	157
R_2	179	188	180
R_3	220	202	212
R_4	152	140	146
G_1	31	24	27
G_2	19	20	16
G_3	43	30	34
W_1	0.20	N.D	0.221
W_2	0.19	0.01	0.201
W_3	0.21	N.D	0.221

N.D.: Not detected & detection limit is 0.1 mg l^{-1}

Moss, 1985). A study in Finland examined a population receiving drinking-water containing uranium with a median concentration of 28 µg/l (Kurttio et al., 2002). In a study of 476 Norwegian groundwater samples, 18% had uranium concentrations in excess of 20 µg/l (Frengstad et al., 2000). Concentrations in excess of 20 µg/l have been reported in groundwater from parts of New Mexico, USA (Hakonson-Hayes, Fresquez & Whicker, 2002), and central Australia (Hostetler, Wischusen & Jacobson, 1998; Fitzgerald et al., 1999).

Based on the radioactive decay chain, uranium isotopes include U^{234}, U^{238}beside Ra^{226} are radiotoxic and carcinogenic species and constitute the most abundant radionuclides in waters (Sidhu and Breithart, 1998). The geological characteristics of soil proved that uranium in the oxidizing aqueous conditions, the groundwater in contact with uranium deposits, well water samples were collected to known uranium mineralized area near Abu-Zenima, west central Sinai to examine the applicability of utilizing the hydrogeochemical technique for detection uranium mineralization in similar arid areas (El-Rayes & Arnous,2015). while in the reducing conditions (absence of oxygen) uranium precipitates and forms secondary uranium deposits (Cothern and Rebers, 1990). On the other hand the concentrations of lead and cadmium in the ground and well water samples are ranged from 1.9 to 6.1 and from 1.1 to 34 mgl^{-1} respectively (Table 5). The pH values of the studied waters have been observed to be in neutral to alkaline limits (6.6-8.2), while the total dissolved solids (TDS) ranged from 420 to 605 mg l^{-1}. Based on the safe limit of TDS for drinking water (600 mg l^{-1} WHO, 2011), the TDS of the studied water samples except G_3 sample (605 mg l^{-1}) were below the permissible limit. The international limits recommend 1.9, 25 and 7µg l^{-1} for uranium, lead and cadmium respectively which mean that, the well water in the studied location are not suitable for human uses based on the high cadmium content especially in G_2 sample (34 µg l^{-1}). Misund et al., (1999) demonstrated that, the leaching of rock by water and coagulation in wells resulted in higher uranium contents in the ground water. The large spread of uranium in ground water could be attributed to the redox sensitivity, the variety of rock uranium contents and its solubility in the oxidizing environment.

Table 5: Cadmium and lead contents (mgl^{-1}) beside pH and TDS values in the studied water samples

Sample	Cd	Pb	pH	TDS
G_1	2.3	5.3	7.32	501
G_2	34	3.2	7.13	420
G_3	1.5	6.1	7.01	605
W_1	1.2	2.4	7.02	560
W_2	1.1	1.9	7.11	576
W_3	0.99	2.2	6.91	465

In spite the European Union (EU, 1998) has not define the maximum allowed uranium level, World Health Organization (WHO, 2011) set 30µg l^{-1} as a maximum uranium limit guideline for drinking water whereas the same concentration level set by the US Environmental Protection Agency (EPA, 2003). Ten years later, the WHO (2008) guidelines set the maximum uranium allowed level in drinking water is 15µg l^{-1}, whereas 10 year before it was much lower (2µg l^{-1}, WHO, 1998). Germany to date recommends that, the allowed uranium in drinking water should not exceeds 2µg l^{-1} (EFSA, 2009, Ljung et al., 2011 and Stalder et al., 2012). Finally, the recommended permissible limits concentration level in drinking water in some countries around the world are shown in Table (6) and graphed on Figures (5&6). Data indicated that, the uranium contents in the studied drinking water samples not comparable with the uranium contents in drinking water of some international countries.

CONCLUSIONS

El Missikat area has several rock units, the most important one is the shear zone which has high concentrations of uranium, thorium and some heavy elements (lead and cadmium). Different analytical techniques including, N_2-Laser Fluorimetric, inductively coupled plasma-mass spectrometry (ICP-MS), Spectrophotometry and X-ray Fluorescence are performed on the rocks, ground and well water samples.

The obtained data show that the uranium concentrations in the rocks, ground and well water samples were ranged from 152-220, 19-43 and around 0.2 $\mu g l^{-1}$ using UA-3 technique, while the uranium concentrations using spectrophotometric technique were 140-202, 20-30 $\mu g l^{-1}$ and undetected values for rocks, ground and well water samples respectively. The uranium contents determined by us-

Table 6: Range of uranium concentrations in drinking water worldwide

Country	Uranium concentrations, $\mu g\ l^{-1}$	References
Canada	0.05 – 4.21	
Argentina	0.04 – 11.0	ICRP, (1993)
Kuwait	0.02 – 2.48	
Sweden	0.02 – 470	Selden et al., (2009)
USA	0.03 – 0.15	Fisenne and Welford, (1986)
Iran	1.0–10.9	Alirezazadeh and Garshasbi, (2003)
Finland	0.02 – 6000	UNSCEAR, (2000)
Turkey	0.2 – 17.62	Kumru, (1995)
Norway	<0.02 – 170	Banks et al., (1995)
Egypt	0.06 – 33.06	EPA, (2003), EPA, (2008)
Egypt	0.201-0.221	Present study

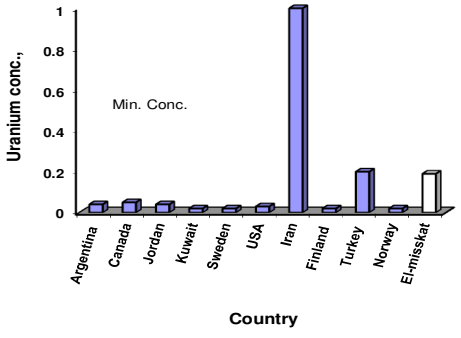

Fig. 5: Minimum uranium concentration ($\mu g\ l^{-1}$) in drinking water worldwide

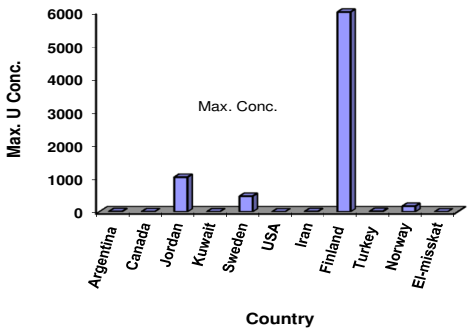

Fig. 6: Maximum uranium concentration ($\mu g\ l^{-1}$) in drinking water worldwide

ing ICP-MS technique were 146-212, 16-34 and around 0.202 μgl^{-1} for rocks, ground and well water samples respectively which nearly matched with values obtained by using the UA-3 technique.

The obtained results by the different applied techniques concluded that there is high concentration levels of uranium in the studied ground water compared with the international permissible limit for uranium in drinking water (1.9 μgl^{-1}). The high uranium content in ground water of the study area is mainly derived by leaching of the uranium-rich rocks of the shear zone in El-Missikat area.

On the other hand, the recommended international permissible limits of lead and cadmium are 25 and7 μgl^{-1} respectively, while their limits in the studied water samples indicated that, water of the study area is not suitable for human uses, especially the G_2 ground water sample which contains 34 μgl^{-1}cadmium content.

Generally, the ground and well water in El-Missikat area are so dangerous for the human due to the impact of the shear zone on ground and well water radioelement and trace metal contents, which subsequently, causes cancers and kidney damage for the users.

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تأثير اليورانيوم وبعض محتويات العناصر الثقيلة على جودة المياه الجوفية بمنطقة المسيكات، الصحراء الشرقية، مصر

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من خلال تلك الدراسة تم تحليل اليورانيوم وبعض تركيزات العناصر الثقيلة في بعض عينات الصخور والمياه الأرضية ومياه الآبار بمنطقة المسيكات بصحراء مصر الشرقية لتحديد مدى ملائمة تلك المياه للاستخدامات البشرية والحيوانية. ولهذا الغرض تم إجراء تحليل اليورانيوم باستخدام تقنيات مختلفة تشمل قياس الطيف الضوئي للأشعة المرئية / فوق البنفسجية (UV/VIS) وقياس الطيف الضوئي بالليزر (N2 UA-3) وقياس الطيف الكتلي للبلازما المقترنة بالحث (ICP-MS). في هذا العمل تم جمع عشر عينات مختلفة من الصخور والمياه من موقع الدراسة (أربع عينات صخرية، ثلاث عينات للمياه الجوفية، وثلاث عينات من آبار المياه). أشارت نتائج التحليل المتحصل عليها إلى أن متوسط تركيزات اليورانيوم والعناصر الثقيلة (الرصاص والكادميوم) في عينات المياه كانت أعلى من الحدود المسموح بها ١,٩ و ٢٥ و ٧ ميكروغرام/لتر لأيونات U و Pb و Cd على التوالي. أوصت نتائج التحليل بأن عينات آبار المياه تشكل خطراً صحياً اعتماداً على المستويات التي تم الحصول عليها من اليورانيوم وتركيزات العناصر الثقيلة وتقرر الدراسة أنه لا يمكن استخدام آبار المياه في منطقة المسيكات كمصدر لمياه الشرب.