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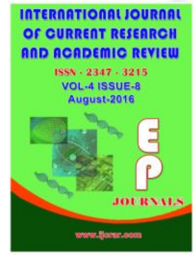

Environmental Geochemistry of Uranium in Recent Sediments of Alkhasa Valley in Kirkuk City / Northern Iraq

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KEYWORDS

Uranium, Sediments of Alkhasa Valley, Geochemistry.

A B S T R A C T

Recent sediment serves as a major reservoir for contaminants as it possesses an ability to bind various chemicals together. To safeguard the members of the public from an unwanted exposure, studies were conducted on the sediments of alkhasa valley where this valley play a very important role in agriculture of Kirkuk -Iraq. The average of uranium found in present study is (1.33 ppm) with range (1-1.6 ppm), the maximum concentration of uranium is (1.6 ppm) in last station (KSD7) and the minimum in first station (KSD1) with value (1 ppm). The results indicated safe levels of U activity in all stations.

Introduction

The Sediments may provide clues to the trace elemental concentrations of natural and anthropogenic contaminants because its important carriers of elements in the hydrological cycle (Ruiz-Fernández *et al*., 2007). Uranium (U) is one of the elements present in sediments, soils, aqueous, plants, food and living organisms in trace quantities andthe mean concentration in soil is (3ppm) (Ricardo *et al*., 2009; Bleise *et al*., 2003). The primary chemical effect of U in humans is Nephritis (kidney) (Hursh & Spoor, 1973). The enriched U is more radioactive and the depleted U is less radioactive than natural U (Weigel, 1983).

The source of U in environment are leaching from natural deposits, release in mill tailings, emissions from nuclear industry and the combustion of coal and other fuels (Dresen*et al.* 1982; Cothern and Lappenbusch 1983, Essien *et al.*,1985, Tadmor 1986), however the soil contamination with U is due to addition of phosphate fertilizers (Träber *et al*., 2015). On the other hand, large amounts of U contents are produced by the modern industry: metallurgy, oil refinery, nuclear industry, nuclear weapon tests, the use of U ammunition, ore mining, phosphogypsum waste heap as well as the manufacture and processing of fuel rods (Boryło, 2012). The sources of mine water cintamination with U, are process water from U-recovery plants, pumped water from underground mine workings that has been in contact with uraniferous ore, surplus tailings water from return water dams and storm water run-off from areas with contaminated materials such as ore piles, dumps and tailings (Funke, 1990; Pulles *et al*., 1996; Wendel, 1998).The concentration and activity of U in the environment depend on the geological features of the area, weather conditions, human, economic and industrial activities (Jwanbot *et al*., 2012).

Uranium occurs in nature in +3, +4, +5, and +6 oxidation states, however, the +4 and +6are very abundant (Zhang *et al.*, 2002). There are three isotopes for U with percentages by mole fraction and half-lives of^{234}U 0.0054% (245,500 Years), ^{235}U 0.72% (704,000,000 Years) and ^{238}U 99.27 % (4,468,000,000 Years) (Berglund and Wieser, 2011; Träber *et al*., 2015). This element has a very high density (18.95 g/cm^3, 1.7 times higher than lead density of 11.35 g/cm^3). The Metallic U has a high boiling point (4131 ^oC) and melting point (1132 ^oC), has a tensile strength similar to most steels and it is chemically very reactive (Zou *et al*., 2011).

Generally, U is found as oxides in the earth's crustsuch as uranium dioxide (UO_2) or triuraniumoctaoxide (U_3O_8) while the pitchblende mineral is the main U ore, consists primarily of uranium oxides. U is primarily (80-90%) present in the +6 oxidation state as the uranylcation (UO_2^{+2}) Speciation of uranium in soil (Ebbs *et al*., 1998).

The U contents and its mobility in aqueous systems is mainly controlled by the pH, alkalinity, the oxidation reduction potential and the type of complexing agents present, such as carbonates, phosphates, vanadates, fluorides, sulfates and silicates, etc. (Langmuir, 1997).

The behavior of Uin soil dependupon adsorption, while the adsorption of Uin clay is more than in both silt and sand respectively. The mobility of Uis reverse and its concentration in soil increases with decreasing sand content and increasing clay content due to the adsorption on the surface of particles rather than ionic-type or intrafoliar retention (Birke *et al*.,2009; Bird and Evenden, 1996). The adsorption and transport of U to sediments allows for accumulation in stream sediments (Brierly, 1981). Retention of Uinsuspension stream sediment decreases as oxidant and alkaline conditions increase while its retention decrease by U highly found as Carbonate due to an increase in the solubility of the element (dominant carbonated uranyl complexes) (Bird and Evenden, 1996) where the transport of U-bearing tailings particles by wind and water erosion and the waterborne transport of dissolved Uranium are the main mechanisms of contaminated stream sediment by U (Winde and Sandham, 2004).

The impact of fertilizer-derived uranium on soils and water is likely to be dependent on several factors, including soil properties (pH, moisture content, mineralogy and texture), fertilizer composition, and water chemical content, therefore, likely to be spatially variable (Zielinski *et al*., 1997). The aims of this study is to determine the U content in sediment samples collected throughout alkhasa valley for recent sediments of Kirkuk-Iraq using the Inductively coupled plasma – Mass Spectrometry (ICP-MS) in order to evaluate the contamination level of U in the study area.

Materials and Methods

Description of study area

The study area, Kirkuk city (the geographical position being (N44°43'00"–44°32'00") and (E 35°50'00"– 35°38'00") is located in Northern Iraq (Fig.1).The altitude above sea level is (367 m), the area of Kirkuk Government equal around (9676 km^2) and represent the ratio about 2.2% of Iraq.

Sampling Sites and Chemical analyses

The Recent Sediment samples were collected in November-2015 using an auger technique, it were collected at one depth between (0-10 cm) and the samples represented by symbols, (KSD1-KSD7).

The samples were oven-dried, pulverized and submitted to a screening process using 200 mesh sieves. The powder material of Recent Sediment samples was stored in plastic sacks. The concentration of Uranium in Recent sediment was determined by employed assayed by the ICP-MS techniques.

Statistical Analysis

Correlation analysis results

The data obtained from analytical methods were treated statistically using SPSS software (Statistical Package for the Social Sciences) (version 22 for windows). Descriptive data analysis was performed, including the calculation of mean, SD,the concentration range of U. Pearson correlation matrix was performed to identify the relationship between the trace metals.

The result of correlation for recent sediment in Kirkuk city (table 2) and (appendix1) are summarized in (table 1)

The very high positive correlation between Uwith (Ta and K_2O) byr value (0.904 and 0.896) respectively, then high positive correlation of Uwith (TiO_2, Y and Nb) byr value (0.822, 0.793 and 0.753) respectively and The good correlation of U with $Na_2O$with value (0.673) (figure 2) because according to Goldschmidtthis elementsare found in one group (Lithophile elements)of classification elements (Goldschmidt, 1954) while the good correlation of Uwith (p and P_2o_5) by value (0.74 and 0.68) respectively because one of the main sources of Uare from fertilizers that’s have high values of phosphor (Träber *et al*., 2015). The good negative correlation of U with (Ni and Co) by value (-0.79 and -0.783) respectively because the source of this element is different with source of U in the environments.

Result and Discussion

Environmental geochemistry of uranium in Recent Sediment

The recent sediment in Kirkuk city are found inalkhasa valley from north to south of Kirkuk, it's one of the environments rich by the toxic element, one of this element is U, there are two type of pollutant source (natural) resulting from source rock and anthropogenic source from manufactory, waste water of house, air precipitation, burial of waste and from agriculture land (Jianshu *et al*., 2014).

Elevated concentration of uranium can be related to U milling and mining sites (Morrison and Cahn 1991), nuclear fuel and nuclear weapons production sites (Riley *et al*., 1992), combustion of coal and oil, in particular, when no proper combustions gas cleaning systems are installed, and the application of phosphate fertilizers (Barisic *et al*., 1992; Zielinski *et al*., 2006)

The present study of pollution show the concentration of uranium in recent sediment, the average concentration of uranium in alkhasa valley is (1.33 ppm) with range (1-1.6 ppm) and stander deviation (0.35) (table3 and figure3), the maximum concentration of uranium is (1.6 ppm) in last station (Near industrial quarter) because its station the last point of alkhasa valley in Kirkuk city that's mean after pollution and concentrate the pollutant of city and the pollutant of industrial quarter put in this valley as well as the waste water from hoses (Schnug, 2005; Wendel, 1998) whileaddition of waste water to agriculture land lead to increase value of U in this environment. The minimum concentration of uranium in first station (Near bridge Rahim Iaoh) in value (1 ppm) because its station before basing alkhasa valley from Kirkuk city that's mean before putting the waste tailing and other pollutants (pollution) (Jwanbot *et al*., 2012; Wendel, 1998).

Indices of Pollution

In general, soil and recent sediment contamination may be considered appreciable when concentrations of an element in soils were two-three times greater than themean background levels (Logan and Miller, 1983). Metal pollution in soil poses a serious threat to the human health and safety of agricultural products. Evaluation for distribution and remediation of heavy metal pollution is the most concerned (Deng Hong-gui *et al*., 2012). In present study, the geo accumulation index (I_{geo}), contamination factor (CF) and enrichment factor (EF) was applied to assess U contamination in Recent sediments located within Kirkuk city.

Index of Geo-Accumulation(I_{geo})

The geo-accumulation index (*I*geo) was used to assess U pollution in Recent sediment of Kirkuk city. It is expressed by the following equation (Müller, 1969):

$$Igeo = log2\left(\frac{Cm}{1.5\ Bn}\right) \ldots\ldots\ldots\ldots\ldots\ldots\ 1$$

where *Cm* = measuredtotal concentration of metals (U) (n) in soils and Recent sediment ($\mu g \cdot g^{-1}$); *Bn*= geochemical background values of metals (U) (n)($\mu g \cdot g^{-1}$) (Turekian and Wedepohl, 1961) the average of uranium in earth's crust is (1.7 ppm); 1.5 = the background matrix correction factor due to lithogenic effects (Loska *et al*., 1997; Gonzáles-Macías *et al*., 2006; Chen *et al*, 2007).The I_{geo} scale consists of seven grades (0-6) ranging from uncontaminated to very highly contaminated (Table 4).

The results of the I_{geo} of the U investigated in this present study are presented in (Table7) also Show the type of Description of Recent Sediment is class one (Uncontaminated to moderately contaminated) for all samples for Recent Sediment ofalkhasa valley by U pollution but respectively along to alkhasa valley from North to south of Kirkuk city to industrial quarter (last station study) where showing high grade of pollution.

Contamination Factor (Cf)

The assessment of soil and Recent Sediment contamination was carried out using the contamination factor. A contamination was described the contamination of a given toxic substance in a basin by (Hakanson, 1980). Contamination factor (C*f*) is ratio of the concentration of the element (U) in samples to pre-industrial reference value for the element (Uranium). It is expressed by the following equation

$$Cf = \left(\frac{Cm\ Sample}{Cm\ background}\right) \ldots\ldots\ldots\ldots\ldots .2$$

Where the C_m sample is the concentration of metal (U) in present study and C_m background is the average background of metal (U) in earth's crust according to (Turekian and Wedepohl, 1961) the average of uranium in earth's crust is (1.7 ppm).The *Cf* scale consists of Four grades ranging from low contamination to very highly contaminated (Table 5).

The results of the Contamination Factor of the Uranium investigated in this present study are presented in (Table 7) describe the degree of contamination in Recent Sediment of alkhasa valley is (CF < 1) (low contamination) unless in near industrial quarter is in class (1 ≤ *CF* < 3) with discretion (Moderate) its station the last point of alkhasa valley in Kirkuk city that's mean after pollution and concentrate of the pollutant of city and the pollutant of industrial quarter put in this valley as well as the waste water from hoses and founding the waste tailing and other pollutants (Schnug, 2005; Wendel, 1998; Jwanbot *et al.*, 2012).

Enrichment Factor (EF)

The enrichment factor is the relative abundance of a chemical element in a soil compared to the bedrock (Hernandez *et al.*, 2003) which is a powerful tool to distinguish between anthropogenic and naturally occurring sources of heavy metals. EF technique is used in the area of soil, sediments, solid wastes and atmospheric aerosols to determine the degree of modification in the composition (Pekey, 2006).

Table.1 Pearson correlation matrix showing (U and elements) relationship for AlkhasaRecent sediment of Kirkuk city-northern Iraq (n = 7)

	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5
U	**0.256**	**0.398**	**- 0.01**	**-0.321**	**0.31**	**0.673 ***	**0.876****	**0.822***	**0.896 ****
	MnO	**Cr**	**OM**	**PH**	**L.O.I**	**Mo**	**Cu**	**Pb**	**Zn**
U	**0.125**	**-0.179**	**-0.022**	**0.083**	**0.025**	**-0.16**	**-0.054**	**0.540**	**0.625**
	Co	**Ni**	**As**	**Te**	**Th**	**Sr**	**Cd**	**Sb**	**V**
U	**-0.783***	**-0.79***	**0.417**	**-0.055**	**0.379**	**0.365**	**0.548**	**0.490**	**-0.101**
	P	**La**	**Ba**	**W**	**Zr**	**Ce**	**Sn**	**Y**	**Nb**
U	**0.862***	**0.666**	**0.135**	**0.471**	**0.494**	**0.143**	**0.218**	**0.793***	**0.753***
	Rb	**Sc**	**Li**	**Ta**	**Hf**				
U	**0.382**	**-0.091**	**-0.169**	**0.904****	**0.49**				

****. Correlation is significant at the 0.01 level (2-tailed)**
***. Correlation is significant at the 0.05 level (2-tailed)**

Table.2 average and range as well as standard deviation of uranium and other elements in recent sediment ofalkhasa valley in Kirkuk-Iraq.

Elements	Samples of Khasa Sediment			Elements	Samples of Khasa Sediment		
	Mean	Range	S.D		Mean	Range	S.D
U ppm	1.27	1 - 1.6	0.21	Th ppm	5.29	4.9 - 5.6	0.24
Sio_2 %	42.12	40.13 - 43.45	1.10	Sr ppm	308.43	250 – 360	42.84
Al_2o_3 %	9.03	8.33 - 9.54	0.57	Cd ppm	0.34	0.2 - 0.5	0.10
Fe_2o_3 %	4.81	4.14 - 5.2	0.41	Sb ppm	0.69	0.5 - 0.8	0.11
Mgo %	3.95	3.04 - 4.84	0.60	V ppm	93.71	91 – 97	1.98
Cao %	20.14	18.98 - 21.42	0.96	P ppm	0.06	0.043 - 0.065	0.01
Na_2o %	0.51	0.31 - 0.88	0.20	La ppm	20.36	19.4 - 21.7	0.77
K_2o %	1.54	1.38 - 1.76	0.13	Cr ppm	223.86	183 – 246	21.43
Tio_2 %	0.6	0.56 - 0.66	0.04	Ba ppm	277.29	255 – 301	18.26
P_2o_5 %	0.16	0.13 - 0.18	0.02	W ppm	0.73	0.6 - 0.8	0.076
Mno %	0.10	0.1 - 0.11	0.01	Zr ppm	44.73	36.8 - 51.6	4.44
OM %	3.58	2.98 - 5.63	0.92	Ce ppm	40.57	38 – 43	1.72
PH	7.37	7.17 - 7.56	0.14	Sn ppm	1.97	1.3 - 3.6	0.80
Loi %	21.23	20.1 - 22.1	0.80	Y ppm	15.47	14.7 - 16.2	0.62
Mo ppm	1.04	0.8 - 1.4	0.25	Nbppm	8.11	7.5 - 8.5	0.32
Cu ppm	31.13	24.6 - 54.8	10.68	Ta ppm	0.46	0.3 - 0.6	0.11
Pb ppm	16.47	10.4 - 22.1	4.90	Scppm	10.71	10 – 11	0.49
Zn ppm	69.29	60 - 85	9.55	Li ppm	27.4	25.5 - 28.6	1.20
Ni ppm	133.49	123.6 - 145.6	6.73	Rbppm	48.94	46.1 - 52.4	1.98
Co ppm	18.16	17.2 - 19.9	0.97	Hfppm	1.47	1.1 - 1.7	0.20
As ppm	7.43	7 - 8	0.53	Teppm	1.93	1.5 - 2.4	0.35

Table.3 concentration of (U) in recent sediment of alkhasa valley of Kirkuk city-northern Iraq.

Names of sites	Symbol	U ppm
Near bridge Raheem Awe	KSD1	1
Near bridge alqala	KSD2	1.1
Near bridge alweladah	KSD3	1.2
Near bridge Girnata	KSD4	1.5
Near the forth bridge	KSD5	1.2
Neare bridge domez	KSD6	1.3
Near industrial quarter	KSD7	1.6
Mean	1.33	
Range	0.8 – 1.8	
S.D.	0.35	

Table.4 classification of Geo-accumulation index (Müller, 1969)

Geo-accumulation Index (I_{geo})		
Value	**Class**	**Description**
Igeo = 0	0	Uncontaminated
0 <Igeo ≤ 1	1	Uncontaminated to moderately contaminated
1 <Igeo ≤ 2	2	Moderately contaminated
2 <Igeo ≤ 3	3	Moderately to strongly contaminated
3 <Igeo ≤ 4	4	strongly contaminated
4 <Igeo ≤ 5	5	Strongly to extremely contaminated
Igeo> 5	6	Extremely contaminated

Table.5 Classification of contamination factor (Hakanson,1980).

Contamination Factors (CF)	
CF classes	**CF Description**
CF < 1	low contamination
$1 \leq CF < 3$	Moderate
$3 \leq CF < 6$	Considerable
$CF \geq 6$	Very high

Table.6 classification of Enrichment Factor (Mmolawa *et al*., 2011)

Enrichment Factors (EF)	
EF classes	**EF Description**
$EF < 2$	Deficiently to minimal enrichment
$2 \leq EF < 5$	Moderate enrichment
$5 \leq EF < 20$	Significant enrichment
$20 \leq EF < 40$	Very high enrichment
$EF \geq 40$	Extremely high enrichment

Table.7 The results of the Igeo, Cf and Ef in Recent study for Recent Sediments for alkasa valley Kirkuk city-northern Iraq.

Name of Site	symbole	U ppm	I_{geo}	CF	Sc ppm	Ef
Near bridge Raheem Awe	KSD1	1	0.118	0.588	14.8	0.855
Near bridge alqala	KSD2	1.1	0.129	0.647	14.4	1.035
Near bridge alweladah	KSD3	1.2	0.141	0.705	14.3	1.026
Near bridge Girnata	KSD4	1.5	0.177	0.882	13.2	1.411
Near the forth bridge	KSD5	1.2	0.141	0.705	14.8	1.026
Neare bridge domez	KSD6	1.3	0.153	0.764	14.2	1.112
Near industrial quarter	KSD7	1.7	0.200	0.941	13.1	1.454
Mean	M.	1.271	0.150	0.747	14.11	1.116

Appendix.1 Concentration of elements in alkhasa valley Kirkuk city-Iraq

Samples / Elements	KS1	KS2	KS3	KS4	KS5	KS6	KS7
Uppm	1	1.1	1.2	1.5	1.2	1.3	1.6
Sio2 %	42.55	42.78	41.45	42.72	41.74	40.13	43.45
Al2o3 %	9.54	8.42	8.33	9.44	8.54	9.54	9.43
Fe2o3 %	4.3	4.88	5.05	4.14	5.2	5.09	4.98
Mgo %	4.84	3.04	4.21	3.32	4.04	4.26	3.94
Cao %	19.41	18.98	20.53	19.11	20.66	21.42	20.9
Na2o %	0.31	0.62	0.38	0.57	0.48	0.31	0.88
K2o %	1.38	1.4	1.54	1.55	1.56	1.57	1.76
Tio2 %	0.56	0.57	0.59	0.6	0.58	0.64	0.66
P2o5 %	0.13	0.15	0.16	0.17	0.16	0.15	0.18
Mno %	0.1	0.11	0.1	0.1	0.11	0.1	0.11
Om %	3.12	3.23	5.63	3.56	2.98	3.21	3.3
PH	7.36	7.56	7.17	7.3	7.26	7.44	7.48
Loi %	20.3	21.6	21.7	22.1	20.9	21.9	20.1
Moppm	0.8	0.9	1.2	0.9	1.4	1.3	0.8
Cuppm	26.4	24.8	27.1	24.6	54.8	29.6	30.6
Pbppm	10.4	12.2	13.8	14.3	22.1	21.2	21.3
Znppm	60	62	71	62	66	79	85
Nippm	145.6	135.5	134.7	132.7	128.9	133.4	123.6
Coppm	19.9	18.2	17.4	17.3	18.8	18.3	17.2
Asppm	7	7	8	7	7	8	8
Thppm	4.9	5.2	5.5	5.3	5.6	5.1	5.4
Srppm	250	298	311	262	360	319	359
Cdppm	0.2	0.3	0.3	0.4	0.5	0.3	0.4
Sbppm	0.6	0.7	0.5	0.7	0.8	0.7	0.8
Vppm	94	92	93	91	95	97	94
P %	0.043	0.052	0.058	0.06	0.059	0.058	0.065
Lappm	19.4	20.3	20.7	20.4	20.5	19.5	21.7
Crppm	246	235	210	222	234	183	237
Bappm	275	255	287	256	301	271	296
Wppm	0.6	0.7	0.8	0.7	0.8	0.7	0.8
Zrppm	36.8	46.3	43.8	44.7	43.4	51.6	46.5
Ceppm	40	41	42	39	41	38	43
Snppm	1.4	1.3	1.7	1.6	3.6	1.8	2.4
Yppm	14.7	14.9	15.7	16.1	15.8	14.9	16.2
Nbppm	7.5	8	8.1	8.2	8.4	8.1	8.5
Tappm	0.3	0.4	0.4	0.6	0.5	0.4	0.6
Scppm	11	10	11	10	11	11	11
Lippm	28.4	27	26.4	25.5	28.6	27.3	28.6
Rbppm	46.1	47.3	52.4	48.9	49.5	48.8	49.6
Hfppm	1.1	1.5	1.4	1.4	1.6	1.7	1.6
Teppm	1.8	2.4	1.5	2.3	1.8	2.1	1.6

Fig.1 Map showing the recent sediment in Kirkuk city-Iraq

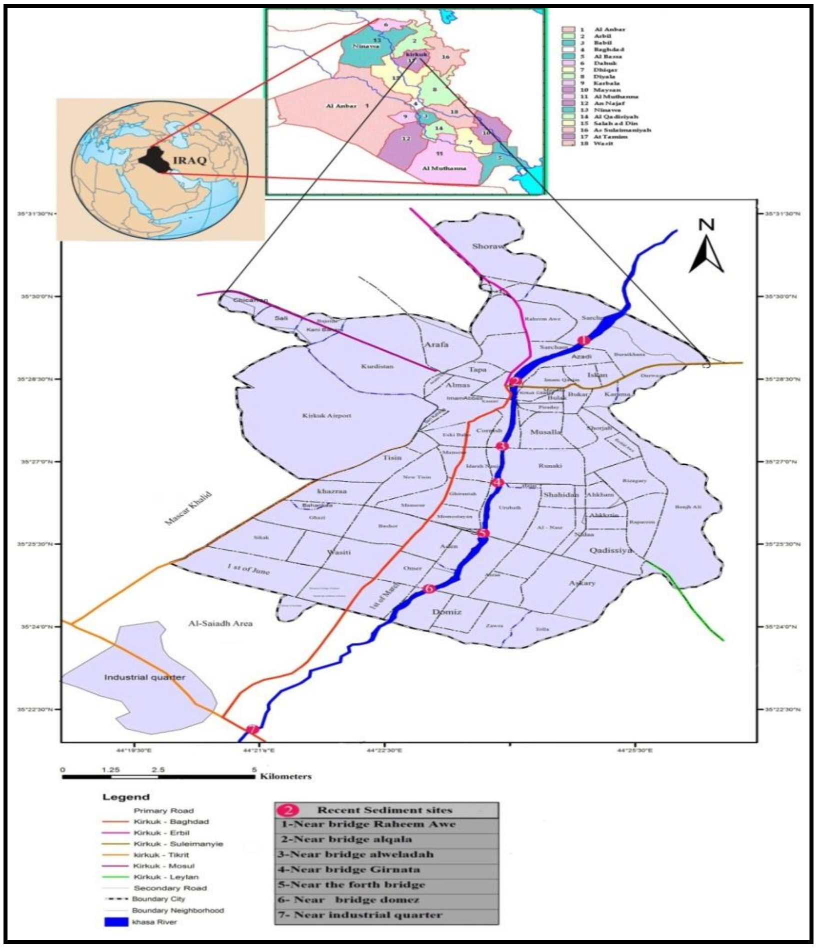


Fig.2 Chart showing the high correlation of U with other elements

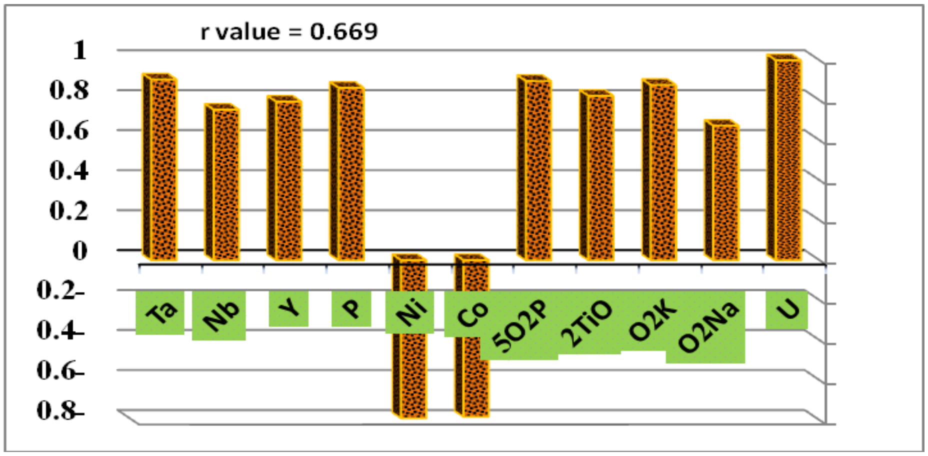
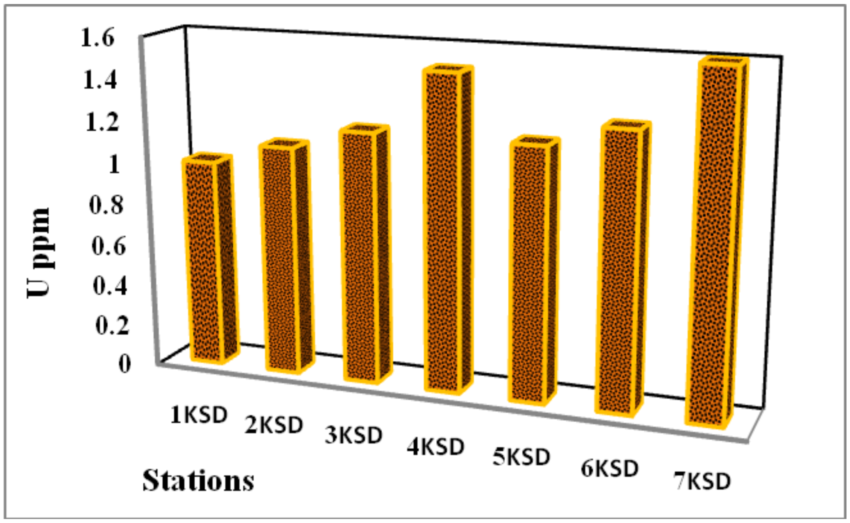


Fig.3 Chart showing the content of U in Recent sediment of alkhasa valley of Kirkuk city-northern Iraq



The enrichment factor was calculated using the formula originally introduced by (Buat-Menard and Chesselet, 1979) as shown in the following equation:

$$EF = \frac{\left(\frac{Cn}{Cref}\right)\text{sample}}{\left(\frac{Bn}{Bref}\right)referens} \ldots\ldots\ldots 3$$

Where the Cn sample) is the concentration of the examined chemical element (U) in the examined environment, *Cref* (sample) is the concentration of the examined chemical element (U) in the reference environment according to (Turekian and Wedepohl) equal to (1.7 ppm), *Bn*is the concentration of the reference chemical element (Scandium) in the examined environment (Bargagli *et al*., 1995; Bergamaschi *et al*., 2005; Bhuiyan *et al*., 2011) (table.6) and *Bref* is the concentration of the reference element (Scandium) in the reference environment according to (Turekian and Wedepohl) equal to (16 ppm).The *Ef* scale consists of Five grades ranging from Deficiently to minimal enrichment to Extremely high enrichment (Table 6).

The results of the Enrichment Factor of the Uranium investigated in this present study of Recent sediment of alkhasa Valley are presented in (Table 7) for all samples are in class *EF* < 2 is Deficiently to minimal enrichment.

Conclusions

The high grade of pollution with Uranium for recent sediment of alkhasa valley in Kirkuk city found Near industrial quarter (1.6 ppm) because its position the last point of alkhasa valley in Kirkuk city that's mean after pollution and concentrate of the pollutant of city and the pollutant of industrial quarter put in this valley as well as the waste water from houses and urban environment. The indices of pollution that's used are (I_{geo}, Cf and Ef) where these indices show the grade of pollution by uranium along of alkhasa valley in Kirkuk city where also show the high grade in the last station (Near industrial quarter) in same reason.

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