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Assessment of Bioavailability of Chromium (Cr), Vanadium (V) and Uranium (U) in Wild Plants in Siwaqa Area, Central Jordan

Asma F. Bzour[1]*, Hani N. Khoury[1] and Sawsan A. Oran[2]

[1]Department of Geology, University of Jordan, P. O Box 11942, Jordan
[2]Department of Biological Sciences, University of Jordan, Jordan

Corresponding author.

Abstract

Surficial soil enriched with U-rich encrustations and green Cr-rich smectite covers large areas of Siwaqa, Central Jordan. The wide distribution of redox-sensitive elements (RSE) as chromium (Cr), vanadium (V) and uranium (U) in the top soil are related to the weathering action of alkaline surface and groundwater on the parent rocks. The bioavailability, distribution, sorption, and ecotoxicity of Cr, V, and U of the wild plants and top soils in the study area were investigated. A total number of 23 surface soil samples and 23 plant samples were collected and analyzed for the most toxic elements (Cr, V and U) by using Induced Coupled Plasma-Mass Spectrometry (ICP- MS). The average Cr, V and U values in the soil was 59, 6.6 and 67 mg/kg respectively. The transfer factors (TFs) of these elements have indicated that the highest Cr and V values (0.6 and 0.26 respectively) were in the wild plants and particularly in *Bellevalia* sp. The highest TF for U (0.25) was recorded in *Onopordum transjordanicum*. The uptake of elements by plants was dependent on the plant species and the concentration of elements in the soil. Differences between TFs values of these elements in the various plant species are related to the different characteristics of the plants. The comparative uptake of the studied RSE was affected by the numerous physical, chemical and biological conditions of the soil. The results of this work provide a valuable knowledge for understanding the bioavailability of some toxic elements in the soil and plants of Central Jordan. The results are expected to be of great help for the Jordanian Uranium Mining Company during their environmental risk assessments.

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Introduction

Chromium (Cr), Uranium (U) and Vanadium (V) are naturally occurring trace elements in rocks and soil environment resulted from pedogenetic processes of weathering (Pierzynsiki et al., 2000). These elements are important in contaminating surface and ground waters and decreasing crop production as a result of bioaccumulation and biomagnification in the food chain. Knowledge of basic chemistry, environmental and associated health effects of these heavy metals is important to understand their speciation, bioavailability, and remedial options. Heavy metals are absorbed in the soil, by initial fast reactions (minutes, hours), followed by slow adsorption reactions (days, years) by plants, redistributed into different chemical forms with varying

bioavailability, mobility, and toxicity (Shiowatana et al., 2001). Heavy metals distribution in soils takes place as a result of mineral precipitation and dissolution, ion exchange, adsorption, aqueous complexation, biological immobilization and mobilization, and plant uptake (Levy et al., 1992). Potentially toxic elements in soils and plants may come from the bedrock itself and anthropogenic sources like solid or liquid waste deposits (Wilson and Pyatt, 2007).

Plants are important components of ecosystems as they transfer elements from abiotic into biotic environments. Water, air and soils are the primary sources of elements from the environment to plants (Hamilton, 1995). All plants have the ability to accumulate essential elements from the soil and need different concentrations from these elements for growth and development. This ability also allows plants to accumulate other non-essential elements which have not known biological function (Djingova and Kuleff, 2000). Several studies have been carried out to evaluate and describe the accumulation of toxic trace elements and its impacts on the plant diversity were investigated by many authors and in different regions (Tomé et al., 2002; Chen et al., 2005).

The soils and plants contain all naturally occurring radioactive elements with half-lives comparable to the age of the earth, although their concentrations in plants may be rather low (Buck et al., 1996). The phytochemistry of honey samples and plant species were studied and the trace elements of Cu, Zn, Ni, Cr and Mn were investigated. The results have indicated that the differences in the values of trace elements were controlled by pollens where the honeybees fed (Atrouz et al., 2004). The uptake of heavy metals by plant parts is real and not due to contamination by aerosols and was best illustrated by highest Ni uptake index in *Atriplex leuceclada* (Abed and Al-Eisawi, 1994).

Top soils of Central Jordan are enriched in redox sensitive trace elements such as U, Cr, Zn, Cr, Ni, Cu, Co, As and Cd in the form of sulphides and selenides (Nassir and Khoury, 1982; Khoury and Nassir, 1982; Khoury, 2012; Fourcade et al., 2007; Techer et al., 2006; Khoury et al., 2014 and 2015). The highest concentrations of trace elements from Central Jordan are present in the altered marble, chalk marl/travertine, and top soil (Elie et al., 2007; Khoury, 2015). Limited research was carried out on the the impact of trace elements on the wild plants in central Jordan. The impact dust and heavy metals emitted from petroleum refinery on plant diversity in Tafila / Jordan was investigated (Oran and Abu Zahra, 2014; Oran and Al- Zo'ubi, 2016). Recent work on central Jordan has indicated that redox sensitive elements are present in the structure of high and low temperature minerals and are adsorbed by the organic matter. of the parent bituminous rocks (Fourcade et al., 2007; Techer et al., 2006; Khoury et al., 2014 and 2015; Khoury et al., 2016). Heavy metals such as U, Cd, As, Cr, Pb, Ni, Zn and V are also enriched in the phosphate rocks of Central Jordan (Abed et al., 2008).

The lack of knowledge of the behavior of some toxic trace elements in soils and plants in Central Jordan in addition to the planned mining activities by Jordan Uranium Mining Company (JUMCO) in the area have encouraged the authors to carry out this work., The transfer of U, Cr, and V to the wild plants in terms of sorption, toxicity and speciation will be emphasized.

Materials and methods

Geology of Central Jordan

The northern boundaries of the first and second areas of Central Jordan (Daba-Khan Al-Zabib- Siwaqa) are located 25 km and 60 km south of Amman with the first area situated between E 36° 00′ to 36° 15′ and N31° 15′ to 31° 30′ and the second area between E 35° 00′ to 36° 15′ and N 31° 15′ to 31° 30′. Fig. 1a shows the location map of the studied area. The studied area was mapped in detail by the Natural Resources Authority (NRA) (Barjous, 1986; Jaser, 1986) and the geology, stratigraphy and sedimentology were described in details by Powell (1989) and Powell and Moh'd (2011).

Fig. 1c is a generalized geological map of the study area (Barjous, 1986; Jaser, 1986; Khoury et al., 2014). Most of the outcropping rocks are of Upper Cretaceous age. The outcropping rocks of Central Jordan are sedimentary in origin and the exposed bed rock ranges in age from Turonian (Upper Cretaceous) to Eocene (Lower Tertiary). Outcrops in central Jordan illustrate the presence of three main rock types: Bituminous marl; varicolored marble (pyrometamorphic rocks); travertine and top soil. The Bituminous Marl Unit overlies the Phosphorite Unit and underlies the varicolored marble and all are of Maestrichtian – Lower Paleocene age (Blake and Ionides, 1939; Quennel, 1956; Burdon, 1959; Barjous, 1986; Jaser, 1986; Khoury et al., 2014). In central Jordan, unusual redox-sensitive elements (RSE) cover large areas and are mainly associated with the varicolored marble, Pleistocene-Recent travertine and top soil. Yellow uranium encrustations and green Cr-rich

smectites are also associated with the varicolored marble (pyrometamorphic rocks) and the underlying bituminous marl (oil shale) and phosphorites (Khoury, 2006; Khoury and Abu-Jayabb, 1995; Khoury et al., 1984; Khoury et al., 2014). The mineralogy of surficial top soil in central Jordan and RSE source rocks, transport conditions, and deposition processes were explained in detail by Khoury et al. (2014). The geochemistry of uranium and vanadium of the mineral phases was not investigated in detail. The general chronological sequence of the different lithological units is illustrated in Fig. 1d. The Upper Cretaceous to Tertiary rocks in Central Jordan were deposited at the margin of the Tethys shelf-sea in environments ranging from super-tidal to deep sub-tidal (Bender, 1986). Transgression took place during Cenomanian times and marine sedimentation continued until the Late Eocene, despite fluctuations in sea level. Gentle folding, block faulting and possible strike- slip faulting were related to continued tectonic movement on the Jordan Rift structure which is located 60 Km to the west of the study area (Bender, 1986; Powell, 1989; Powell and Moh'd, 2011).

Bioclimatic and vegetation regions of Central Jordan

Central Jordan is classified as arid Mediterranean bioclimate; cool, warm and very warm varieties (Al-Eisawi, 1996). The largest area of Central Jordan is located in arid Mediterranean warm variety bioclimatic zone and located in Irano-Turanian vegetation region, while the north west of Central Jordan is in Mediterranean vegetation region and a very small part of south east of central Jordan located in Saharo-Arabian vegetation regions Fig. 1b. The vegetation is mostly herbs, shrubs and bushes with no trees (Al-Eisawi, 1996).

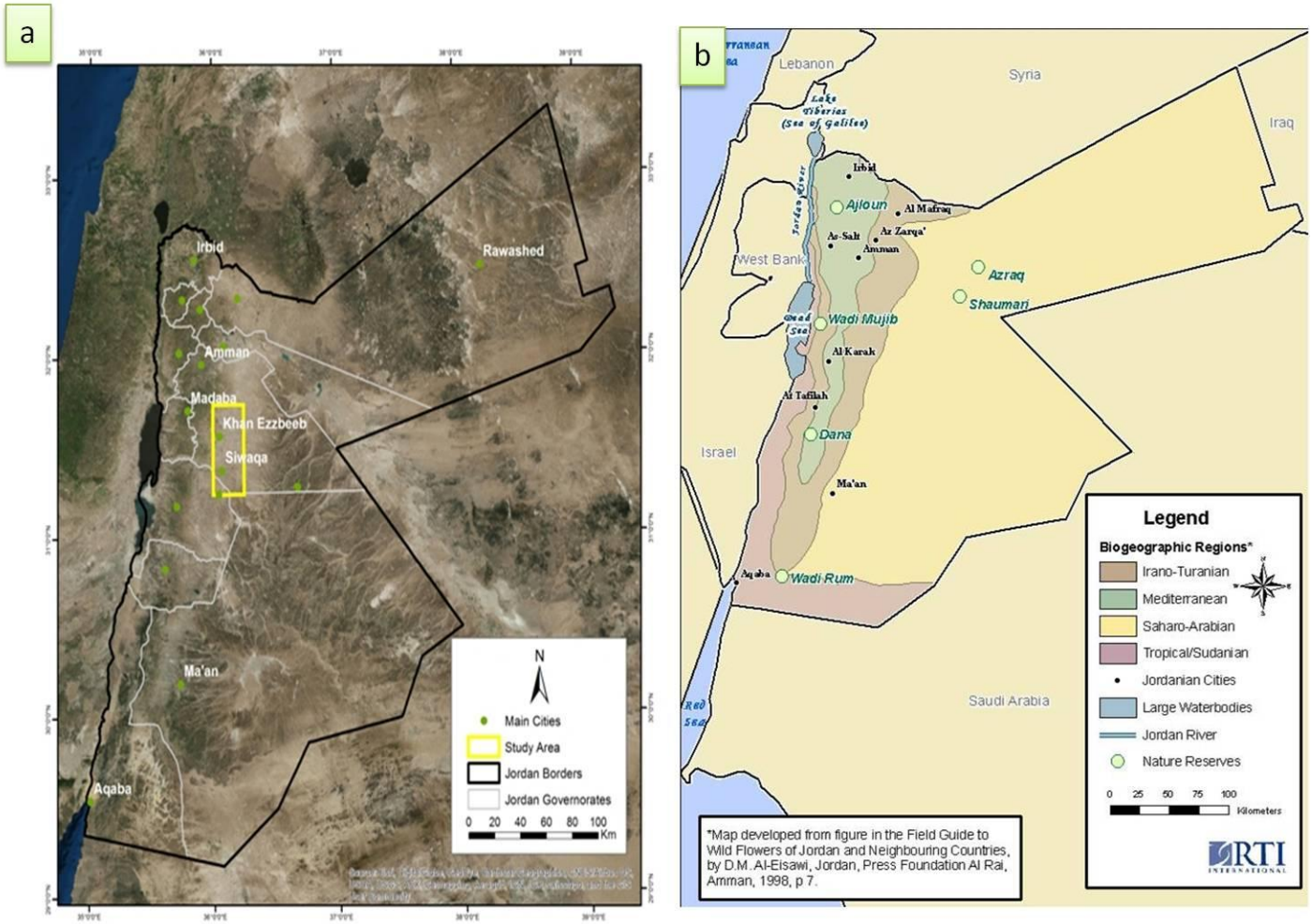

Fig. 1: (a) Location map of Jordan showing the study area/ Central Jordan; (b) Location map of the study area showing the vegetation regions of Central Jordan (after Al-Eisawi, 1996);

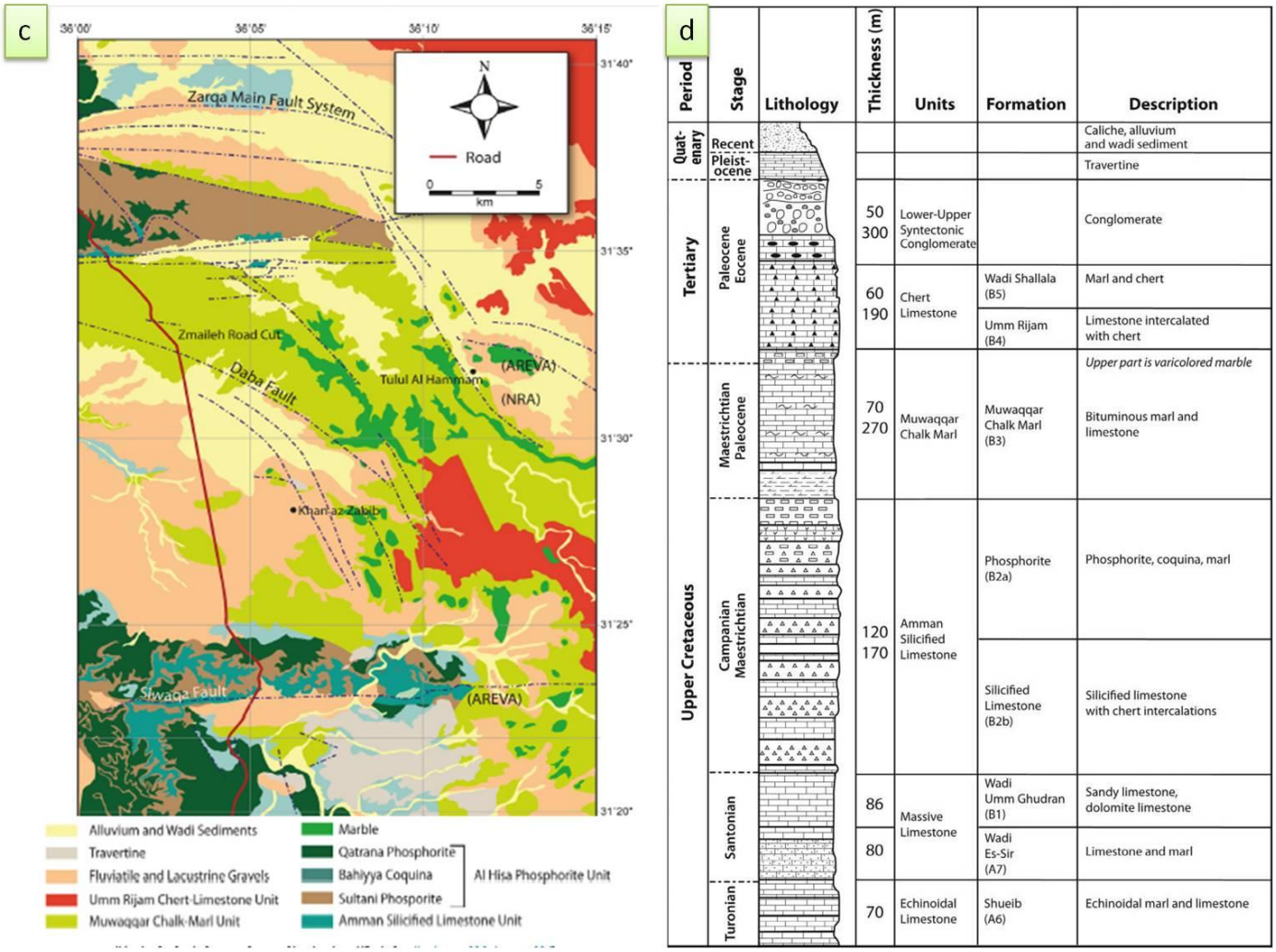

Fig. 1: (c) Geological map of central Jordan (modified after Barjous, 1986; Jaser, 1986; Khoury et al., 2014); (d) Simplified geologic section of central Jordan (modified after Barjous, 1986; Jaser, 1986; Khoury et al., 2014).

Field and laboratory work

Field trips were conducted extensively to Siwaqa area Central desert of Jordan. Fresh plant specimens and soil samples were collected for every plant species available at the time of sampling and were placed in plastic bags. Global positioning system (GPS) records were made from the sampling sites during the period of the study. Sample locations were chosen randomly referring to the availability of plant species and sampling was based on ease of access, adequate vegetation growth to provide vegetation samples. Plant specimens were identified based on: Flora Palaestina (Parts 1, 2, 3 and 4) (Zohary, 1966; Feinbrun, 1986). Field Guide to Wild Flowers of Jordan and Neighboring Countries (Al-Eisawi, 1998).

Siwaqa area is dry with scare vegetation; wild plants and green grass cover the landscape during Spring time that is used by locals for grazing and as folk medicine (Fig. 2a). A number of 23 soil samples and 23 plant samples were collected. Fig. 2b shows a general view of the landscape in Siwaqa area indicating the scarcity of vegetation cover. The top soil is porous and is mainly composed of calcite and gypsum. Secondary green Cr-rich smectite and yellow uranium minerals are common features filling voids and planes of weakness. Yellow secondary uranium minerals (source of U and V) together with green Cr- rich smectites are almost always associated together in the top soil and the underlying travertine and altered marble (Figs. 2c and 2d). Fig. 3 (a-h) are selected photographs of some typical wild plants from Siwaqa area.

Plant samples were carefully brush- washed with tab water and again washed and rinsed with distilled water to remove externally adhered metals and dust from the

surface of the plants. The plants were oven dried at 100ºC overnight. The samples were ground in a small coffee grinder. Finally, the samples were ground into fine powder by using Mill mix.

Soil samples were oven dried at 100ºC overnight, then sieved through 63 microns mesh screen into fine powder. The samples were characterized using chemical methods. All the analytical work was carried out in the laboratories of the Department of Geology, University of Jordan and the Jordan Atomic Energy Commission (JAEC).

Inductively coupled plasma mass spectrometry (ICP-MS) analyses were carried out in the Chemical Analysis Section, chemical and physical analysis laboratories, Nuclear Fuel Cycle Commission (NCC- CPAL), Jordan Atomic Energy Commission (JAEC). The ICP- MS (ELAN® DRC-e, PerkinElmer SCIEX) was used to measure the amount of toxic trace elements in plants and soil particles. All samples were sent via Jordanian Uranium Mining Company (JUMCO) to ALS Arabia Co. Ltd. Jeddah, Saudi Arabia to be analyzed.

The TF was used as an index for the accumulation of trace elements by plants or the transfer of elements from soil to plants in mg/kg (Whicker et al., 1999; Yanagisawa et al., 1992). The transfer factor TF is defined as a factor used to describe the amount of element which is expected to be transferred to plant from soil. It is also defined as the ratio of specific activities in plant parts and soil (in Bq/kg dry weight plant part divided by Bq/kg dry weight soil).

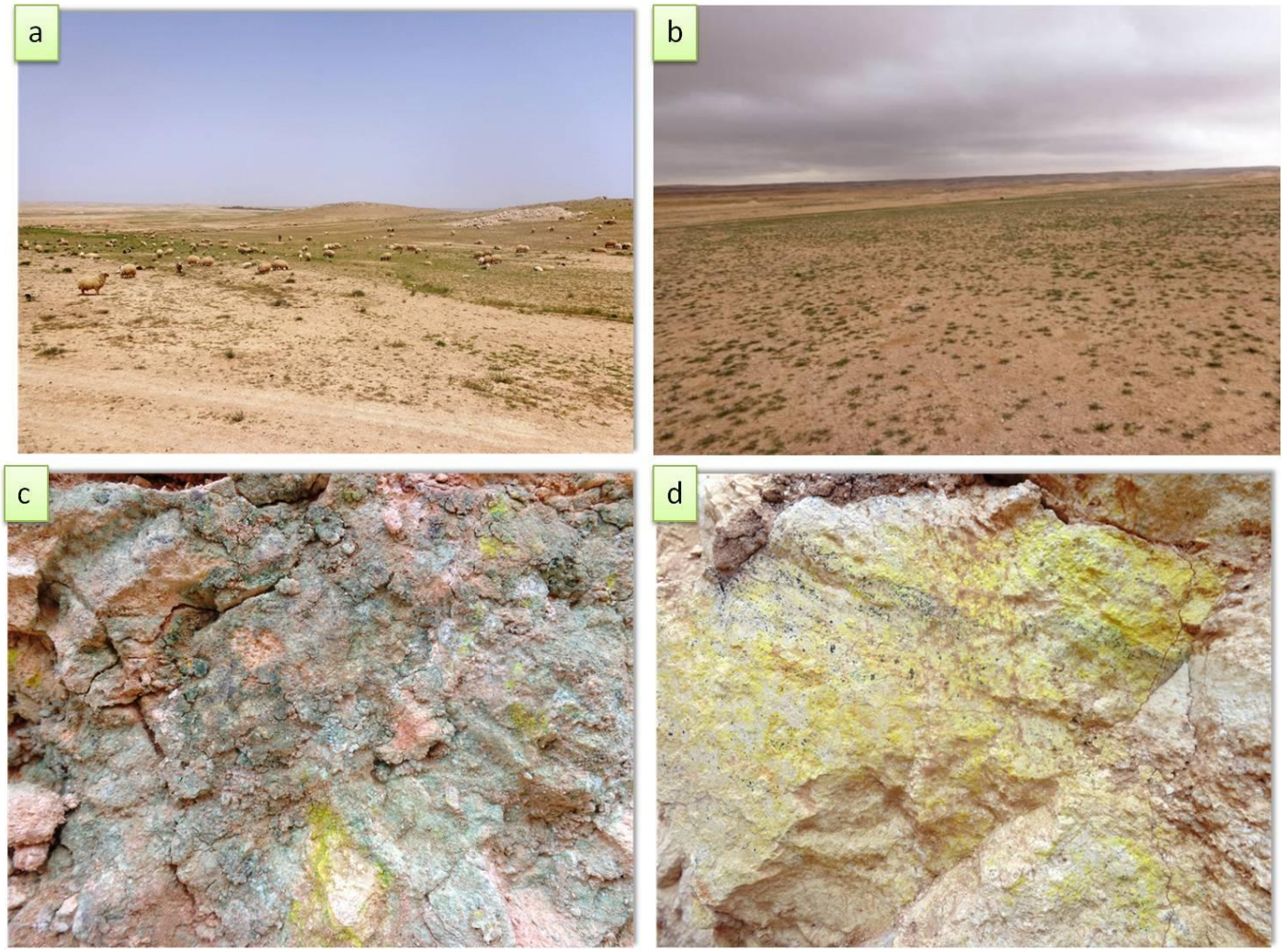

Fig. 2: (a) Landscape of the study area showing grazing by herbavious animals; (b) A Photograph showing a general view of the landscape indicating the scarcity of vegetation cover; (c) A photograph illustrates the presence of secondary encrustations of uranium and Cr-rich smectites in the altered marble/top soil; (d) A phtograph of yellow secondary uranium minerals filling porous rocks.

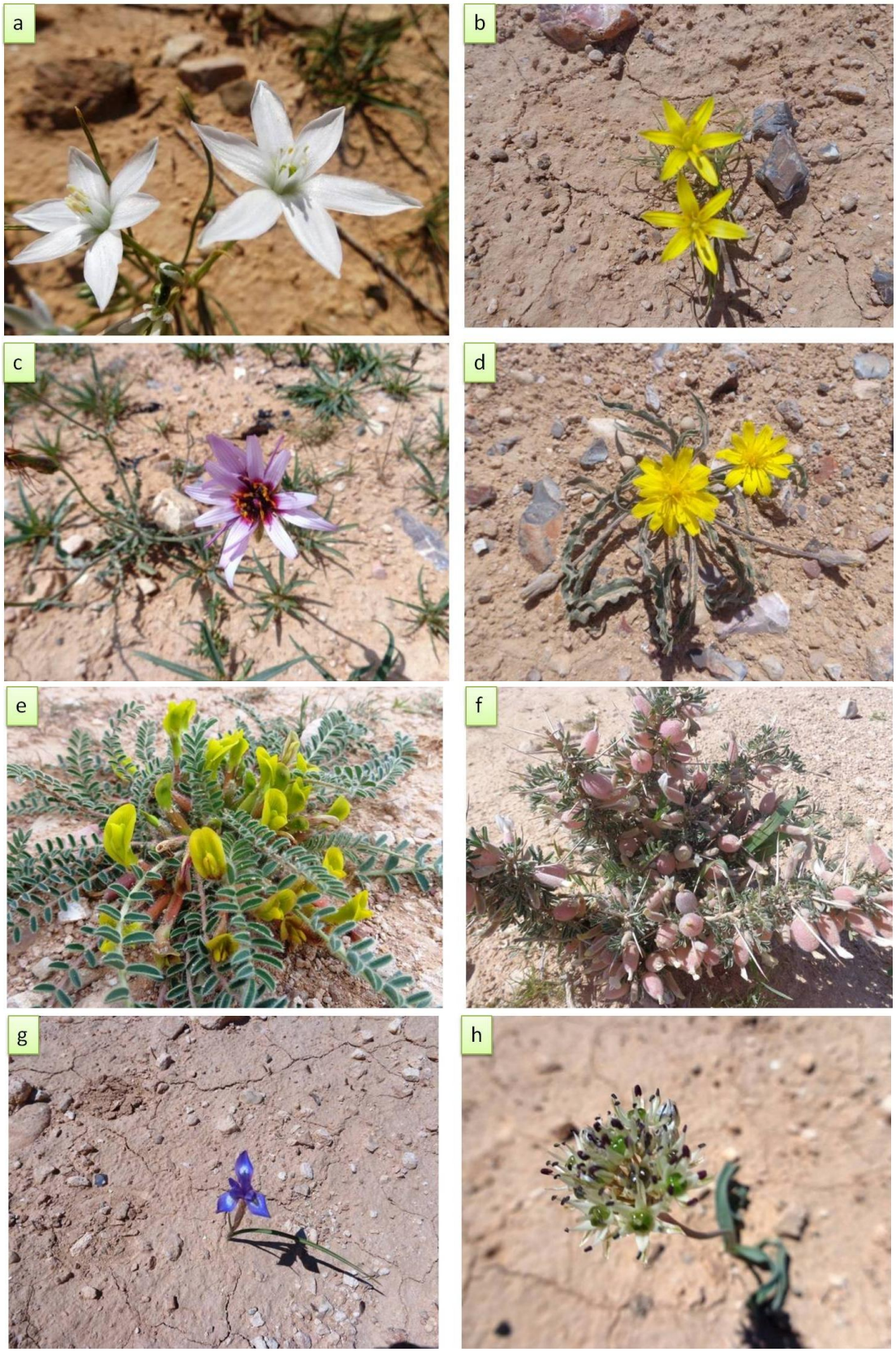

Fig. 3: Photographs of wild plants (a) *Ornithogalum trichophyllum*; (b) *Gagea reticulate*; (c.) *Scorzonera papposa*; (d) *Scoronozera schweinfurthii*; (e) *Astragalus sparsus*; (f) *Astragalus spinosus*; (g) *Gynandriris sisyrinchium;* (h) *Allium rothii.*

Results and discussion

In homogeneous unusual surface RSE (U, V and Cr) hosted by the top soil covers large areas in central Jordan. The secondary uranium mineralization is restricted to the permeable fractured and porous zones. The combustion of the bituminous marl led to the formation of the in central Jordan (Khoury et al., 2014). The action of circulating highly alkaline water that accelerated the leaching process of the redox sensitive trace elements such as U, Cr and V. Such an alkaline oxidizing environment (pH~12.7) is currently active in Maqarin area, north Jordan (Khoury, 2012 and 2014). The uranyl-vanadate minerals strelkinite and/or tyuyamunite ($Na_2(UO_2)_2(V_2O_8).6H_2O$ and $Ca(UO_2)_2(VO_4)2{\cdot}5\text{-}8H_2O$) and Cr-rich smectite were precipitated from highly alkaline solutions during the dry periods after the precipitation of the thick travertine deposits (Khoury et al., 2014).

Mass Spectrometry analysis (ICP- MS) results

The mass spectrometry analysis (ICP- MS) results are given in Table 1. The table includes the concentration results of U, V, and Cr in the soil and plant samples in addition to the calculated transfer factors.

Table 1. Trace elements concentrations (mg/kg) in soil, plant and their TFs of Siwaqa area.

Location 1	Soils			Plant name	Plants			TF		
Sample ID	Cr	U	V		Cr	U	V	Cr	U	V
1	44.7	3.17	54.2	*Bassia eriophora*	12.35	0.537	8.5	0.3	0.17	0.16
2	48.4	4.93	57.5	*Arthocnemum mucronatum*	3.51	0.182	2.9	0.07	0.04	0.05
3	47.8	4.81	59.2	*Astragalus spinosus*	13.45	0.487	9.7	0.3	0.1	0.16
4	49.2	3.55	56.5	*Onopordum transjordanicum*	22.9	0.882	14.5	0.5	0.25	0.25
5	42.2	6.9	62.8	*Bellevalia* sp.	26.6	1.365	16.6	0.6	0.2	0.26
6	51.1	7.09	65.7	*Astragalus* sp.	10.4	0.62	7.2	0.2	0.1	0.11
7	56.3	6.62	67.1	*Matthiola* sp.	5.88	0.423	5.5	0.1	0.06	0.08
8	70.1	2.12	61.1	*Allium rothii*	1.19	0.05	0.7	0.02	0.024	0.01
9	66.6	1.83	52	*Scorzonera schweinfurthii*	3.14	0.141	2.4	0.05	0.08	0.046
10	55.1	3.02	84.4	*Senecio flavus*	9.22	0.596	12.1	0.2	0.2	0.14
11	52.2	3.76	94.1	*Peganum harmala*	11.95	0.357	7.1	0.2	0.1	0.075
12	65.5	3.87	63.3	*Onopordum transjordanicum*	16.7	0.539	7.5	0.25	0.14	0.12
14	81.5	24.8	100.5	*Hordeum glaucus*	11.75	3.78	6	0.1	0.15	0.06
26	75.3	30.2	115.5	*Malva sylvestris*	14.5	3.11	12.6	0.2	0.1	0.12
27	66.2	34.5	120	*Hordeum glaucus*	11.95	3.03	7.4	0.2	0.1	0.06
28	61.5	39.5	132.5	*Onopordum transjordanicum*	19.35	7.38	15.8	0.3	0.2	0.12
29	60	56.8	142	*Hyoscyamus desertorum*	16.6	5.29	17.7	0.3	0.1	0.125
31	108	18.4	96.1	*Reichardia tingitana*	8.47	1.72	6.3	0.1	0.1	0.065
32	56.1	8.68	58.5	*Matthiola* sp.	5.44	0.844	2.5	0.1	0.1	0.04
33	56.7	4.4	59.2	*Allium rothii*	13.55	0.912	11	0.2	0.2	0.2
34	60.2	6.31	71.9	*Gymnarrhena micrantha*	8.92	0.904	5.1	0.15	0.14	0.07
35	59.1	8.91	73.5	*Maresia pygmaea*	8.09	0.682	4.6	0.1	0.08	0.06
36	68.8	6.64	76.6	*Gynandriris sisyrinchium*	12.1	0.589	6.4	0.2	0.1	0.08
Mean	**60.98**	**12.64**	**79.31**		**11.65**	**1.497**	**8.265**	**0.2061**	**0.1232**	**0.107**
STADE	**14.19**	**14.59**	**26.63**		**6.153**	**1.844**	**4.759**	**0.1375**	**0.0574**	**0.06507**
Std. Error of Mean	**2.958**	**3.042**	**5.553**		**1.283**	**0.3844**	**0.9923**	**0.02867**	**0.01197**	**0.01357**
Minimum	**42.2**	**1.83**	**52**		**1.19**	**0.05**	**0.7**	**0.02**	**0.024**	**0.01**
Median	**59.1**	**6.62**	**67.1**		**11.95**	**0.682**	**7.2**	**0.2**	**0.1**	**0.08**
Maximum	**108**	**56.8**	**142**		**26.6**	**7.38**	**17.7**	**0.6**	**0.25**	**0.26**
Number of values	**23**									

The abundance of Cr in the Earth's upper crust averages 100 mg/kg (Pendias and Pendias, 2000). Chromium enters the air, water, and soil mostly in the chromium (III) and chromium (VI) forms. In air, chromium compounds are present mostly as fine dust particles which eventually settle over land and water. The main reason of the toxicity of Cr (VI) is that one of the reduction products of Cr (VI) is Cr (V). Chrome (V) is a known as carcinogenic and will lodge in any tissue to form cancerous growths. WHO (2007) and Pendias and Pendias (2000), recommended that 75 mg/kg Cr in soil is critical concentration to be toxic. WHO (2007) recommended that 1.3 mg/kg is the toxic limit to plant species. The concentrations of Cr in soils of Siwaqa area range between 42.2 to 108 mg/kg with a mean value of 60.98 mg/kg. Three soil samples

have exceeded the permissible limit of Cr in the soil as recommended by WHO (2007) and Pendias and Pendias (2000). The highest value of Cr content in plant species of Siwaqa area is 26.6 mg/kg and recorded in *Bellevalia* sp. The lowest value of Cr content is 1.19 mg/kg and is recorded in *Allium rothii*. The mean value is 11.65 mg/kg. Only *Allium rothii* has Cr concentration below the toxic limit as recommended by WHO (2007). Fig. 4 shows the Cr concentrations in plant species of Siwaqa area. The TF for Cr from soil to plant of Siwaqa area ranges from 0.02 to 0.6 with a mean value of 0.21. The highest TF value is found in *Bellevalia* sp. and the lowest TF is found in *Allium rothii*.

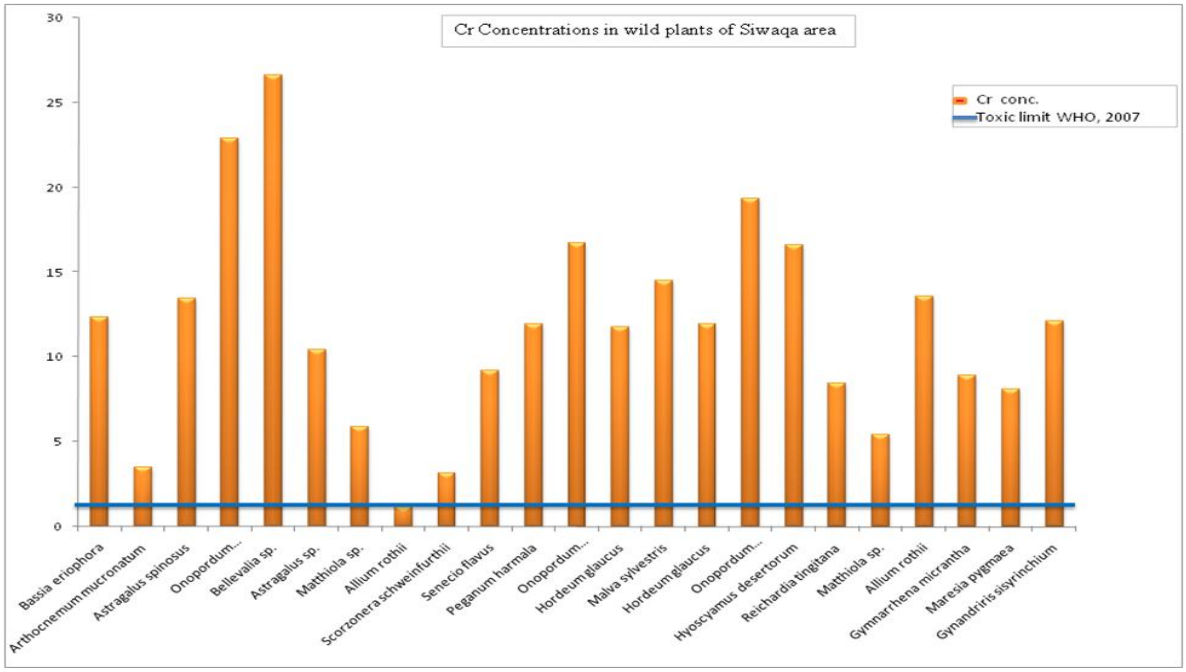

Fig. 4: Cr concentrations in wild plant sp. in Siwaqa area.

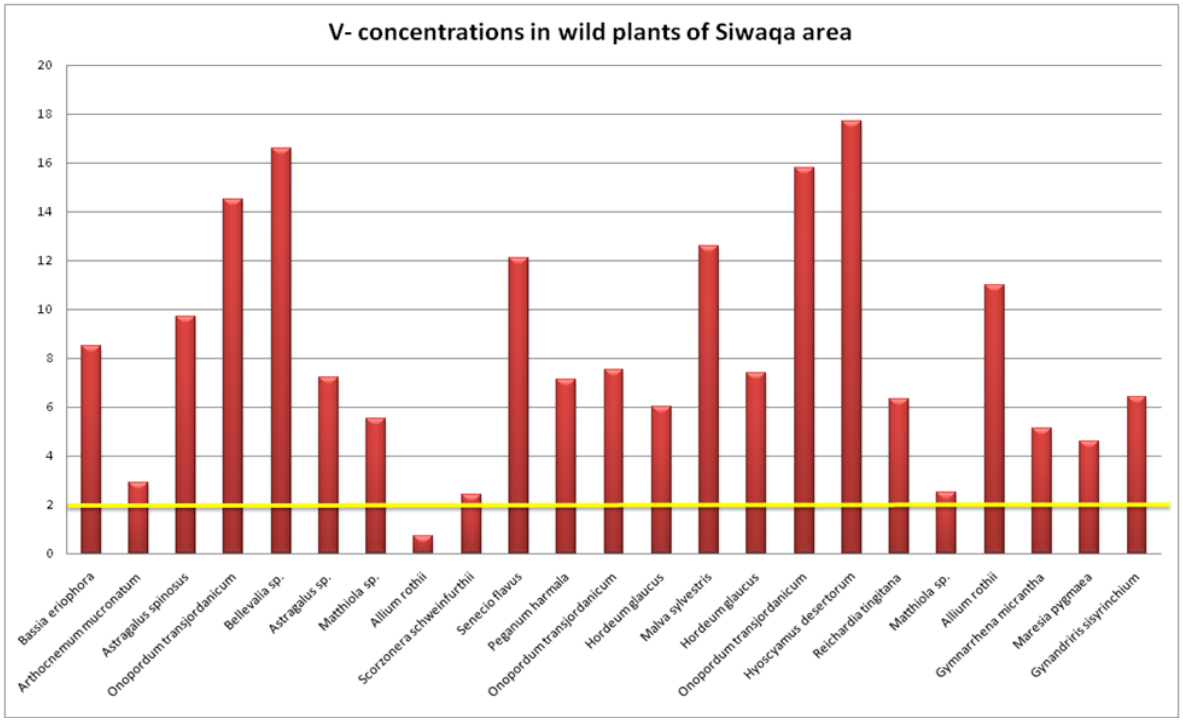

Fig. 5: V concentrations in wild plant species of Siwaqa area.

Levels of V in soils are closely related to the parent rock types. Its worldwide soil average is estimated at 129 mg/kg, within the range of 69–320 mg/kg (Pendias and Pendias, 2000). WHO (2007) recommended that 2 mg/kg V concentration is toxic to plants. The concentrations of V in soils of Siwaqa area range between 52 to 142 mg/kg with a mean value of 79.31 mg/kg. The highest value of V concentration in plant species of Siwaqa area is 17.7 mg/kg and is recorded in *Hyoscyamus desertorum* and the lowest V concentration value is 0. 7 mg/kg and is recorded in *Allium rothii*. The mean value is 8.27 mg/kg. Only one species of Siwaqa area has a concentration value below the toxic limit as recommended by WHO (2007). Fig. 5 shows the V concentrations in plant species of Siwaqa area. The TF for V from soil to plant of Siwaqa area ranges from 0.01 to 0.26 with a mean value of 0.11. The highest TF is found in *Astragalus sp.* The lowest TF is found in *Allium rothii*.

Exposure to uranium can result in both chemical and radiological toxicity. The main chemical effect associated with exposure to uranium and its compounds is kidney toxicity. This toxicity can be caused by breathing air containing uranium dusts or by eating substances containing uranium, which then enters the bloodstream. Once in the bloodstream, the uranium compounds are filtered by the kidneys, where they can cause damage to the kidney cells. Very high uranium intakes can cause acute kidney failure and death (ATSDR, 2005; Sheppard et al., 1985). The toxic limit for U concentrations in soils and plants are 1 and 0.3 respectively recommended by WHO (2007). The concentrations of U in soils of Siwaqa area range between 1.83 to 56.8 mg/kg with a mean value of 12.64 mg/kg. All soil samples have exceeded the toxic limit of U concentration in soils as recommended by WHO (2007). The highest value of U content in plant species of Siwaqa area is 7.38 mg/kg and is recorded in *Onopordum transjordanicum.* The lowest value of U content is 0.05 mg/kg and is recorded in *Allium rothii.* The mean value is 1.497 mg/kg. Fig. 6 shows that the most collected plant species of the Siwaqa area have U concentrations above the toxic limit as recommended by WHO (2007). The TF for U from soil to plant of Siwaqa area ranges from 0.024 to 0.25 with a mean value of 0.123. The highest TF is found in *Onopordum transjordanicum* and the lowest TF value is found in *Allium rothii*.

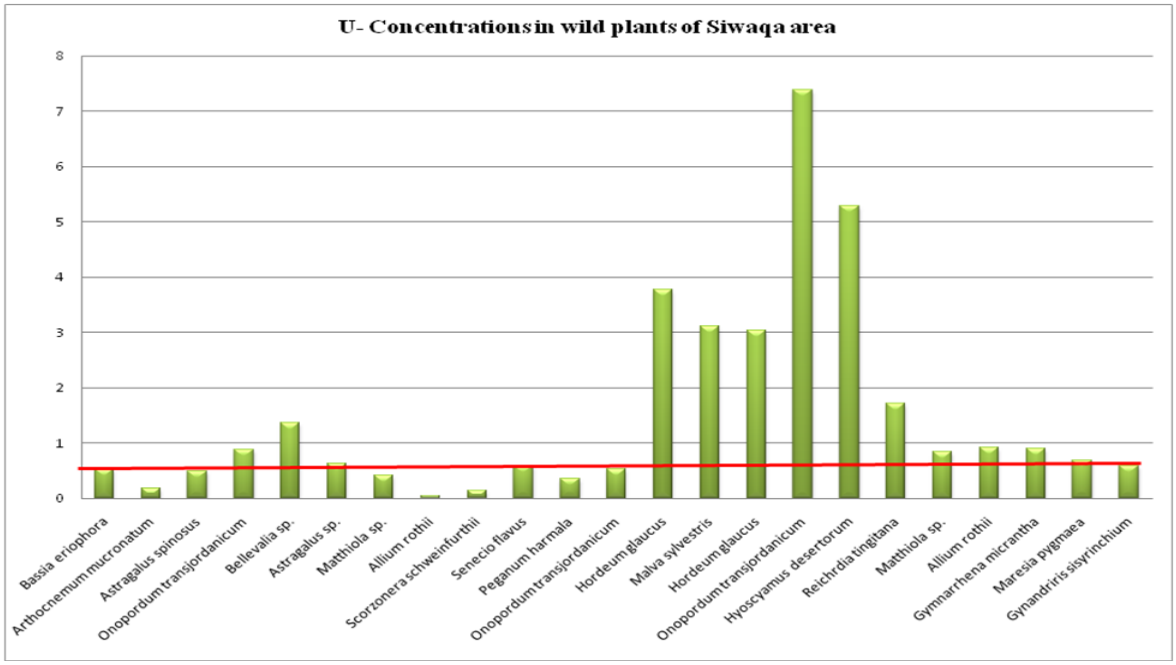

Fig. 6: U concentrations in wild plant species of Siwaqa area.

Conclusion

The present study has been conducted to assess wild plant species in Siwaqa area/ Central Jordan for the absorption of different hazardous trace elements. The environmental geochemical survey has revealed variations of trace elements concentrations in soils and plant species. The sources of trace elements are related to the leaching processes from the parent rocks of the area. The concentration of these elements was depending on

climatic geologic events in Central Jordan. Differences between trace elements TFs values for various plant species are related to the different characteristics and behavior of the wild plants. In general, the comparative uptake of trace elements is affected by numerous physical, chemical and biological conditions of the soil. The uptake of elements by plants depends on the plant species and the concentration of elements in the soil. The values are above the permissible limits according to world health organization (WHO). The information on the concentration level and transfer of Cr, V and U from soil to plant will provide important information during the environment risk assessment that is expected to be carried out by JUMCO in the near future. Further studies and investigations are needed to assess and evaluate the ecotoxicity of heavy metals on plant species by the different RSE in the different soils of Central Jordan.

Conflict of interest statement

Authors declare that they have no conflict of interest.

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