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ENVIRONMENTAL CONTAMINATION BY HEAVY METALS AND RADIOACTIVE ELEMENTS IN WADI NASAB AND ITS SURROUNDINGS, SOUTHWESTERN SINAI, EGYPT

[187]

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ABSTRACT

Wadi (Valley) Nasab (WN) and its surroundings, is an area of mining located at the south-western Sinai, Egypt for some heavy metals and radioactive elements from Lower Carboniferous. This activity represent a source of environmental contamination. This contamination is the main target of this study. Ore material, solid wastes and soil samples were collected from 10 stations. Whereas, plants samples were collected from two types of herb weeds (i.e., *Zygophyllum siplex* and *Haloxylon salicornicum*) from WN and its surroundings southwestern of Sinai.

The original ore material is chemically consisted of around 50% SiO_2, 10.6% Al_2O_3, 10.2 Fe_2O_3, 13% (CaO + MgO) and around 10% loss on ignition. This figure means high carbonate and total iron contents. The mineralogic constituents of solid waste are consisted of Quartz, Kaolinite, Jarosite and Gypsum. The milling waste contains 60.2% SiO_2, 6.8% Al_2O_3, 9.4% Fe_2O_3, 4.7% (CaO + MgO) and 14.8% loss on ignition. The picture of some trace elements is as follow; Cu is 2900 ppm in ore materials and 359 ppm in milling waste. In the same order, Zn is 1865 and 92 ppm, Th is 14 and 26 ppm, U is 346 and 184 ppm, Ra^{226} is 135 and 179 Bq/ kg.

This picture of ore material and milling waste is reflected on the soil profile after the flash flood. The degree of contamination is pronounced in the upstream water of W. Nasab and decrease in the direction of downstream. Thorium (Th) ranged from 5 to 13 ppm at the upstream and from 4 to 5 ppm in the downstream. On the other hand, U ranged from 5 to 9 ppm in the upstream and from 2 to 7 in downstream. The contamination effect is also clear in two types of Herb weeds of WN. Uranium in roots of (*Zygophyllum siplex*) plant, ranged from 4 to 6 ppm while in roots of (*Haloxylon salicornicum*) plant was not detected. Also, it was not detected in the vegetable parts of the two herb plants and Th was not detected in roots and the vegetable parts of the two herb plants.

Keywords: Solid waste; Heavy metals; Radioactive elements; *Zygophyllum siplex*; *Haloxylon salicornicum*

INTRODUCTION

Heavy metal pollution in soil profiles can pose dangers and hazards to human beings. Extreme concentrations of some heavy metals in biological systems, are very dangerous to human health, and may even cause death **(Ayeni et al 2010)**. Generally, the presence of radionuclide elements (e.g., K, U, Th and Ra), as well as heavy metals (e.g., Cd, Pb and Ni), has negative effects on the environmental system, especially in agricultural soil **(Abdel-Haleem et al 2001, Pulhani et al 2003, Ayeni et al 2010** and **EPA, 2006).**

Human beings and animals may consume radionuclides accumulated in parts of some plants in the form of food. By time, accumulatinn of these radionuclides within different organs of the human body may cause serious health problems if they exceed the maximum allowable dose (**ICRP, 1990** and **UNSCEAR, 1982** and **IAEA, 1996).** Moreover, 80% of the radiation dose contributes to normal radioactivity and only 20% of cosmic rays and various other nuclear processes such as radionuclides (U, Th, Ra, K, etc.).

Soil contaminated by heavy metals may cause adverse effects on plant growth. Excessive accumulation of heavy metals beyond toxic limits of plant growth in soil. The accumulation of heavy metals can lead to many distortions on plant growth such as changes in germination, leaf chlorosis or plant death **(Kokkola et al 2000, EPA, 2006, Bonnet et al 2000** and **Ayeni et al 2010).**

Rumble et al (1986), observed in their study in South Dakota, USA, that when they measured the concentrations of U and Ra elements in plants growing in the soil around the uranium mill at the study sites. These plants had higher levels of U and Ra elements compared to the control site. They also noted that the number of radionuclides absorbed by plants from the soil depends mainly on the form of radionuclides, the moisture content, and the chemical and mineral composition of the soil. **Rosholt et al (1966)** and **Sully et al (1987)** recommended the necessity to study the processes that transport radioactive elements and their biological activity and to study the distribution of natural radioisotopes in soil profiles. Uranium and Th are natural sources of radiation in soil minerals common in nature and absorbed on to soil components (organic matter, clays, carbonates, Fe and Mn oxides).

The danger of radionuclides for both animals, humans and plants may be very high if exposed to a maximum dose exceeding the recommended dose (**ICRP, 1990**, **UNSCEAR, 1982**, **Ayeni et al 2010** and **EPA, 2006)**. Health problems affecting humans, possibly through direct or indirect exposure to radioactive contamination sources through different pathways, such as ingestion of contaminated water or inhalation of contact with soil contaminated or the food series **(Wuana,** and **Okieimen, 2011).** The more recent recommendation of **IAEA** (**1996)** and **El Galy et al (2008)** suggested a permissible dose rate of 5 mSv/ y for members of the public and 20 mSv/ y from all natural and artificial radiation sources in the environment for work-related members**.**

Therefore, the study aims to assess the effect of environmental pollution of heavy elements and radioactive elements with ore materials and Solid waste in the mining area of wadi Nasab. Moreover, studying their effects on soil contamination and naturally growing herb plants.

MATERIALS AND METHODS

The purpose of this study is firstly to assess to what degree environmental contamination for some heavy metal and radioactive elements from lower Carboniferous sedimentary rocks (ore materials) and solid wastes. Secondly, assess to what degree contamination of heavy metals and radioactive materials on soil and some plants in Wadi Nasab and its surroundings.

Sampling

Ore material and solid waste samples were collected from W. Nasab and its surroundings. Soil samples were collected form 10 stations, while plant samples were collected and represented by two types herb weeds (*i.e., Zygophyllum siplex* and *Haloxylon salicornicum*) from the upstream and downstream of W. Nasab and its surroundings (as shown in **Fig. 1)**.

Analyses

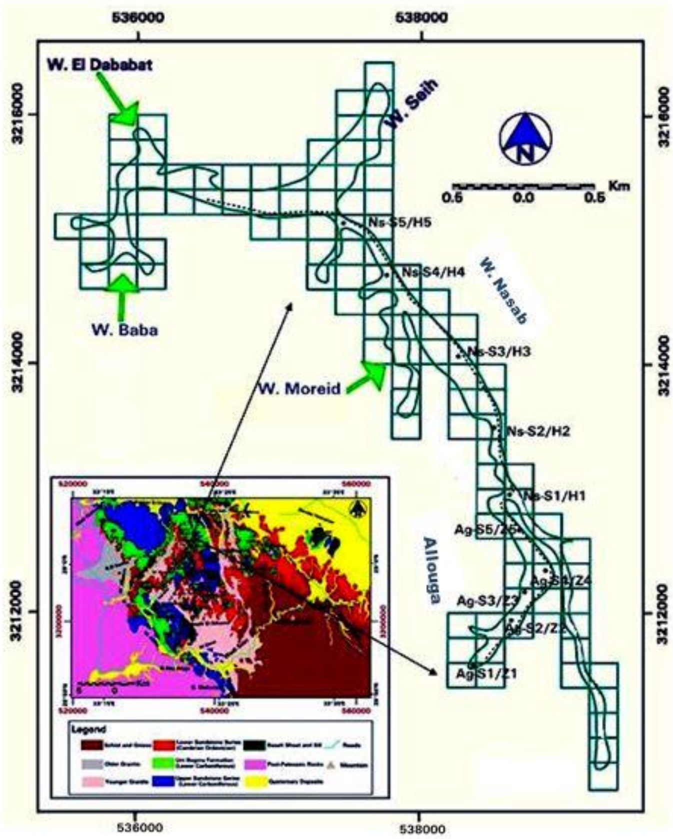
To identify the chemical composition of ore material, solid wastes and soil, a representative sample was crashed, ground to a 200 mesh grain size and the rapid analysis was applied as described by **Shapiro (1975**)**.**

Mineral composition of the solid waste sample was determined by X-ray diffraction. Bulk grinding to < 2 microns, quartering and sieved sample was subjected to Philips diffract meter, the X-ray tube was a Cu target model PW 22/23. The tube was operated at 40 kV and 20 mA. The obtained diffractograms are properly interpreted using standard diffraction mineral patterns XRD as well as the model mineralogical composition was calculated sample according to **Griffin (1967)**.

Total heavy metals, namely Zn, Mn, Pb, Ni, and Cu, were measured using a Unicam atomic absorption spectrophotometer model-969 (AAS) according to the method described by **Welz** and **Sperling (1999),** at flame type of 213.9, 279.5, 217, 232, and 324.8 nm respectively.

Total REEs and Th, were determined by UV-VIS spectrophotometer (Shimadzu UV-160) using arsenazo (III) **(Marczenko, 1976)** at 660 nm. Finally, for U an oxidimetric titration was used against ammonium metavanadate in the presence of diphenylamine sulphonate indicator was used. Prior to titration proper reduction was performed using ammonium ferrous sulphate as described by **Mahmoud (2003)**.

Electrical conductivity (EC) was determined in the extract of soil paste using electrical conductivity bridge according to the method of **Richards (1954)**.



Ag - Allouga Ns - Nasab Z - *Zygophyllum siplex* H - *Haloxylon salicornicum*

Fig. 1. Geologic map of east Abu Zenima aera, after (Omer, 2016) and locations samples map from W. Nasab and its surroundings in southwestern, Sinai

Chloride was determined volumetrically using silver nitrate method. Soluble sulfate was precipitated quantitatively with BCl_2 then excess B^{2+} was titrated with Na_2 EDTA as mentioned by **Jackson (1958)**.

RESULTS AND DISCUSSION

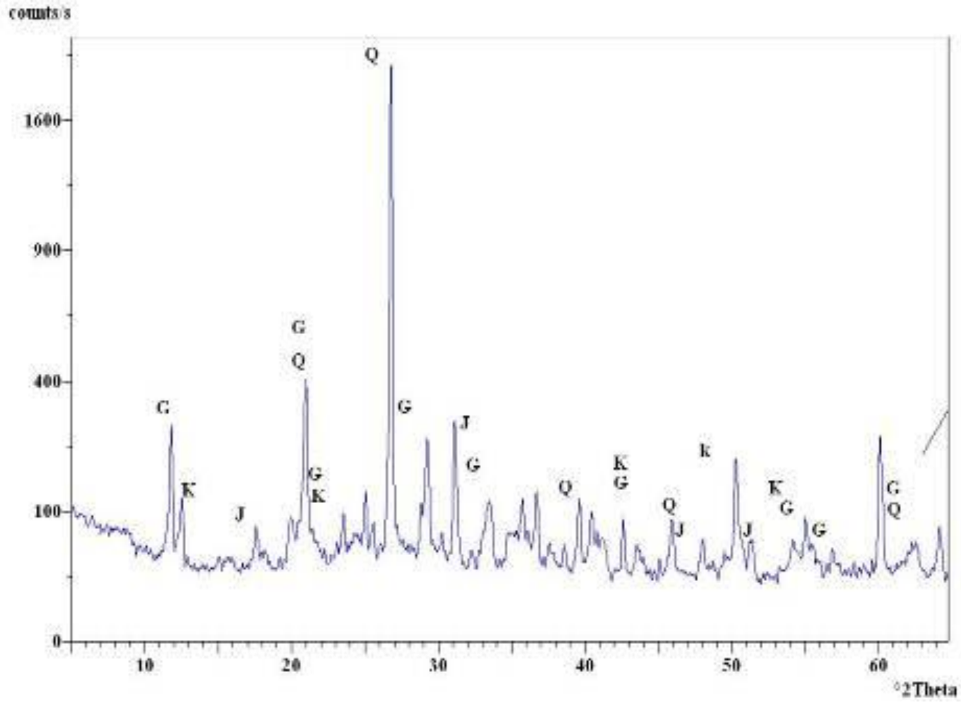
Specification of the solid waste and ore material

a- Mineralogy of solid waste

The minerals constituent of the collected mixed waste sample from processed radioactive materials after processing with acidified water and then analyzed by X-ray diffraction (XRD) and Emission Scanning Electron Microscopy (ESEM) techniques. The mineral constituents of the solid waste under study is shown in (**Fig. 2).** This figure shows XRD patterns of Quartz SiO_2, Kaolinite $Al_4(Si_4O_{10})(OH)_8$, Jarosite KFe_3 $(SO_4)_2(OH)_6$ and Gypsum $CaSO_4.2H_2O$.

b- Chemical characteristics of the original ore materials and solid waste

Data in **Table (1)** show the chemical composition of the original ore material from lower Carboniferous sedimentary rocks and solid waste. The main component of major elements in ore material were SiO_2 and Al_2O_3 (50% and 10.6%), whereas the lowest major elements were MnO (0.38%), TiO_2 (0.51%) and P_2O_5 (0.6%). Besides, Fe_2O_3 (10.2%), (CaO + MgO) (13.9%). Alkalies as Na_2O was 1.4% and K_2O was 1.3%, chloride presented 0.76% and 9.8% of loss on ignition. This figure means high carbonate and total iron contents. The main component of major elements in the milling solid waste were SiO_2 (60.2%) and FeO_3 (9.4%).



2θ = Angle of diffraction Q = Quartz K = Kaolinite G = Gypsum J = Jarosite

Fig. 2. XRD pattern of solid waste from the studied area

Table 1. Chemical composition of original ore material and solid wastes from processed radioactive materials of studied Alouga area.

Major components	Concentration (%)		Heavy and radioactive element	Concentration (ppm)	
	Ore material	Solid waste		Ore material	Solid waste
SiO_2	49.80	60.2	V	350	89
TiO_2	0.51	0.20	Cu	2900	359
Al_2O_3	10.60	6.82	Cd	22	7
Fe_2O_3	10.20	9.42	Zn	1865	92.4
MnO	0.38	0.23	Mn	3746	3144
CaO	7.46	1.48	Pb	400	305
MgO	5.52	3.2	Ni	320	186
Na_2O	1.40	0.32	CO	250	126
K_2O	1.32	0.56	REEs	810	561
P_2O_5	0.60	0.32	U	346	184
Cl^-	0.76	0.13	Th	14	26
L.O.I.	9.8	14.8	K	18	11
Total	**98.35**	**97.58**	**Ra**	**•135**	**•179**

• = Bq/ kg

Whereas it was (4.7%) for (MgO + CaO). In addition, alkalies were Na_2O (0.3%), K_2O (0.5%), chloride presented (0.76%) and (14.8%) for loss on ignition (**Table 1**). This figure indicates high total iron content in the ore material.

Data in the same table show the main trace elements of the original ore material and solid waste. The main trace elements in ore material were Mn (3746 ppm). Cu (2900 ppm) and Zn (1865 ppm). The main trace elements in the milling solid waste were Mn (3144 ppm) and (REEs 561 ppm). Thorium recorded 14 ppm in ore material and 26 ppm in the milling waste respectively, while U record 346 ppm in the ore material and 184 ppm in the milling waste, and Ra^{226} has 135 Bq/ kg in the ore material and 179 Bq/ kg in the milling waste. This table showed increase of solubility with acidified water used in leaching ore materials for V, Cu, Zn and U, while Mn, Pb and Ni showed decrease in solubility.

Chemical characteristics of soil

The ore material and milling waste are subjected every year to the flash flood at days 11 to 16 of December (**Fig. 3)**.

The chemical composition of the studied soil profiles is shown in **(Table 2)**. The collected samples represent the upstream soil profile named Ag.S1, Ag.S2, Ag.S3, Ag.S4 and Ag.S5. The collected downstream soil samples are Ns.S1, Ns.S2, Ns.S3, Ns.S4 and Ns.S5 which collected from W. Nasab area. The values of SiO_2 concentration in the upstream soil profiles are ranged between 58.3 and 62.5% while, Al_2O_3 between 9.89 and 12.23%, Fe_2O_3 ranged between 6.65% and 11.5%. In the downstream the SiO_2 ranged between 57.8% and 66.5%. Aluminum oxide (Al_2O_3) ranged between 11.3% and 17.23%, Fe_2O_3 ranged between 5.25% and 9.58%. The loss on ignition ranged between 7.92% and 9.26% in downstream soil sediments and between 8.84% and 11.52% in the upstream.

Trace elements distribution **(Table 3)** in the upstream soil profile showed that Cu ranged between 2048 and 3637 ppm, while Zn ranged between 173 and 442 and REEs ranged from 8 and 26 ppm, while U ranged between 3 and 9 ppm, also, Th ranged between 4 and 13 ppm. The picture of some trace elements in the downstream soil profile is different as the Cu ranged between 1464 and 2168 ppm, while Zn ranged between 130 and 374 ppm, REEs ranged between 4 and 14 ppm, while U ranged between 2 and 5 ppm, and Th ranged between 4 and 7ppm. From the previous data, it is noticed that the distribution of major and trace elements decreased from upstream to downstream when can be due to the annual flash floods.

It is evident from the results that the concentrations of Cu, Cd, Zn, Mn, Pb and Ni are above phytotoxic limits in all soil samples taken from the soil profeils in W. Nasab and its surroundings. This may lead to abnormalities in the germination process of some plants or death of plants as was explained by **Kukkola et al (2000), Bonnet et al (2000)** and **Ayeni et al (2010).** Also, the results of measurement of radioactive elements in different soil samples of study area indicate U and Th at levels higher than those mentioned by International Standard Dose Rate Unit and plant life which is 1 sv/ kg **(Knoll 2000).**

Contamination of the Herb weeds plants by heavy metals and radioactive elements

The two studied types of plants which recorded in couse of W. Nasab from Upstream (*Zygophyllum siplex*) species to the downstream (*Haloxylon salicornicum*) species are shown in **(Fig. 4).**

Uranium in roots of *Zygophyllum siplex* ranged from 4 to 6 ppm and there is no contamination found in the vegetable growth, while in *Haloxylon salicornicum* species there is contamination in both roots and vegetable parts. In contrast, Th was not detected in roots and the vegetable parts of the two herb plants. However, this accumulation can be effective on the long run.

No contamination by Cu was found in both roots and vegetable parts in the two plants as shown in **(Tables 4** and **5).** On the other hand, there is variable concentration of different trace elements in both types of plants. On the other hand, Cd, Zn, Mn, Pb and Ni elements were found in soil profiles beyond those causing harm to the two herbs **(Abdel-Haleem et al 2001, Pulhani et al 2003, Ayeni et al 2010** and **EPA, 2006).** This means that these two herbs can accumulate heavy elements, and hence can be used for soil phytoremediation.

Start of upstream

End of Downstream

Fig. 3. Start of flash flood at Alouga and the look at downstream of W. Nasab.

Table 2. Chemical analyses of major components in soil profiles of W. Nasab and Alouga area

Component major	Concentration in different soil profiles (%)									
	Upstream					Downstream				
	Ag.S1	Ag.S2	Ag.S3	Ag.S4	Ag.S5	Ns.S1	Ns.S2	Ns.S3	Ns.S4	Ns.S5
SiO_2	58.33	61.95	62.5	62.3	58.66	57.8	59.65	66.5	62.60	62.67
TiO_2	0.10	0.11	0.09	0.07	0.07	0.10	0.15	0.09	0.09	0.12
Al_2O_3	12.23	10.15	9.89	11.45	10.6	17.23	12.80	11.30	12.10	13.25
Fe_2O_3	9.44	8.65	11.2	6.65	11.5	9.50	7.50	5.25	9.58	8.65
MnO	0.28	0.35	0.19	0.12	0.84	0.13	0.09	0.56	0.08	0.24
CaO	1.95	2.55	3.33	3.45	2.95	1.50	3.10	1.90	2.30	2.35
MgO	2.86	2.0	1.95	2.33	2.10	2.30	1.80	1.10	1.90	2.50
Na_2O	0.02	0.04	0.10	0.20	0.10	0.12	0.25	0.19	0.09	0.18
K_2O	0.32	0.12	0.14	0.35	0.09	0.11	0.34	0.22	0.14	0.16
P_2O_5	0.30	0.01	0.08	0.06	0.07	0.09	0.10	0.04	0.08	0.03
Cl^-	0.36	0.23	0.33	0.37	0.30	0.32	0.28	0.28	0.22	0.34
L.O.I.	11.52	10.51	8..84	9.46	9.21	8.92	9.56	8.76	8.43	7.92
Total	**97.71**	**96.67**	**98.28**	**96.81**	**96.44**	**98.12**	**95.62**	**96.19**	**97.61**	**98.41**

Table 3. Chemical analyses of heavy metal and radioactive elements from soil of W. Nasseib and Alluga area

Heavy and radioactive elements	Concentration in different soil profiles (ppm)									
	Upstream					Downstream				
	*Ag.S1	Ag.S2	Ag.S3	Ag.S4	Ag.S5	**Ns.S1	Ns.S2	Ns.S3	Ns.S4	Ns.S5
Cu	3637	2220	2048	2484	2484	1776	1706	2208	1464	2168
Cd	8	5	6	5	12	9	9	4	6	8
Zn	442	186	173	280	192	148	265	374	142	130
Mn	4702	2688	1213	1343	1804	949	422	1153	435	625
Co	71	154	178	68	70	54	50	62	41	52
Pb	189	287	239	258	224	202	164	194	148	156
Ni	196	257	230	216	179	177	144	130	184	140
REEs	26	16	12	16	8	14	4	10	10	8
U	9	9	7	7	3	5	5	5	2	2
Th	13	13	5	4	4	7	5	4	4	5

* Alluga area. ** W. Nasab area.

Zygophyllum siplex

Haloxylon salicornicum

Fig. 4. Herb weeds of W. Nasab and its surrounding areas.

Table 4. Chemical analyses of heavy metals and radioactive elements in *Zygophyllum simplex* species

Heavy and radioactive elements	Concentration in *Zygophyllum simplex* species (ppm)									
	Roots					Vegetable parts				
	Z1/R	*Z2*/R	*Z3*/R	*Z4*/R	*Z5*/R	*Z1*/V	*Z2*/V	*Z3*/V	*Z4*/V	*Z5*/V
Cu	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d
Cd	2	2	2	5	5	n.d	0.5	n.d	n.d	n.d
Zn	163	170	148	132	128	74	81	18	44	49
Mn	198	124	154	180	159	85	52	65	50	115
Co	76	77	66	52	96	41	52	47	n.d	n.d
Pb	220	179	181	196	186	119	n.d	108	170	54
Ni	52	42	38	30	101	10	n.d	n.d	n.d	21
U	6	5	4	4	n.d	n.d	n.d	n.d	n.d	n.d
Th	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d

(n.d) = not detected.

Table 5. Chemical analyses of heavy metals and radioactive elements in *Haloxylon salicornicum* species

Heavy and radioactive element	Concentration in *Haloxylon salicornicum* species (ppm)									
	Roots					Vegetable parts				
	H1/R	H2/R	H3/R	H4/R	H5/R	H1/V	H2/V	H3/V	H4/V	H5/V
Cu	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d
Cd	4	3	4	5	5	n.d	n.d	n.d	n.d	n.d
Zn	170	121	215	97	145	90	24	64	43	42
Mn	215	192	302	217	366	92	145	119	112	145
Co	54	27	29	47	71	39	n.d	n.d	n.d	57
Pb	168	116	89	100	93	34	60	86	47	45
Ni	111	181	105	149	70	59	n.d	n.d	47	n.d
U	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d
Th	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d

(n.d) = not detected.

CONCLUSION

From the previous complementary work for ore material, milling waste, soil and plants, it can be conceded that the activities in this location led to pollution in the soil by some heavy metals and radioactive elements. One of the positive results from this study is the action of *Zygophyllum simplex* plant in accumulating some of heavy metals and radioactive elements in its roots. In contrast*, Haloxylon salicornicum* did not accumulate, any of them. Hence, it is recommended to achieve more experiments on the ability of other types of plants in phytoremediation after chemical retreatment for milling solid waste.

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الموجـــــــز

وادي نصب من أحد المناطق المحيطة بمنطقة تعدين المعادن الثقيلة والمشعة من الصخور الرسوبية منخفضة الرتبة ويمثل هذا النشاط مصدراً للتلوث البيئي ودراسة تأثير هذا التلوث هوالهدف الرئيسي من هذه الدراسة. تم تجميع المادة الخام والمخلفات الصلبة وعينات التربة من 10 مواقع مختلفة وجُمعت عينات لنوعين من النباتات العشبية هما *Zygophyllum siplex* و *Haloxylon salicornicum* من وادي نصب ومحيطه في جنوب غرب سيناء. تحتوى المادة الخام على عدد من المكونات الكميائية هى أكاسيد السليكا (50%)، الألومنيوم (10.6%)، والحديد (10.2%)، والكالسيوم والمغنسيوم (13%) وقد وصل المفقود بالحرق إلى حوالي 10%. وتحتوى المخلفات الصلبة الناتجة من معالجة الصخور الرسوبية (المادة الخام) على معادن السليكا، الكاءولين، الجاروسيت والكالسيت وتحتوي أيضاً على أكاسيد السليكا (60.2%) والألومنيوم (6.8%)، الحديد (9.4%)، والكالسيوم والمغنسيوم يمثلان معاً نسبه 4.7% وقد وصل المفقود بالحرق إلى 14.8%. وتحتوى المادة الخام على العناصرالشحيحة بالتركيزات التالية النحاس 2900 جزء فى المليون، 359 جزء فى المليون فى المخلفات الصلبة ويمثل الزنك 1865 جزء في المليون في المادة الخام، 92 جزء فى المليون في المخلفات الصلبة. ويمثل الثوريوم 14 جزء فى المليون في المادة الخام، 26 جزء فى المليون في المخلفات الصلبة بينما يمثل اليورانيوم 346 جزء في المليون في المادة الخام ويمثل 184 جزء فى المليون في المخلفات الصلبة والرديوم يمثل 135 بيكرل/ كيلوجرام في المادة الخام، 179 بيكرل/كيلوجرام في المخلفات الصلبة. توضح تركيزات هذه العناصر درجة التلوث بوادي نصب بما يحتويه من تربة ونباتات طبيعية بعد الجريان السطحى للسيول المفاجئة على المادة الخام والمخلفات الصلبة من المنبع بمنطقة العلوجة وخلال مرورها بوادي نصب والذي يمثل المصب لهذه السيول وسرعة تدفق التيارالعلوي لمياه السيول عالية وتنخفض تدريجياً في إتجاه المصب. وتحتوي التربة عند بداية تدفق السيول العلوي على الثوريوم ويتراوح تركيزه ما بين 5 و13 جزء في المليون بينما يتراوح تركيزه ما بين 4 و5 جزء في المليون خلال مجرى السيول المنخفض بالوادي. واليورانيوم عند بداية تدفق السيول العلوية يتراوح تركيزه ما بين 5 و9 جزء في المليون، بينما يتراوح تركيزه ما بين 2 و7 جزء في المليون خلال مجرى السيول المنخفض بالوادي. بالنسبه لنوعي النباتات العشبية بوادي نصب وجد أن اليورنيوم يتراوح تركيزه بجذور نبات *Zygophyllum siplex* ما بين 4 و6 جزء في المليون، بينما في جذور نبات

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Haloxylon salicornicum لم يتم إكتشافه. وأظهرت نتائج الدراسة عدم وجود تلوث في المجموع الخضري باليورنيوم لكلا النوعين من النباتات العشبية وأيضاً عدم وجود الثوريوم في كل من الجذور والمجموع الخضري لكلا النوعين من النباتات العشبية النامية طبيعياً بمنطقة الدراسة.

الكلمات الدالة: المخلفات الصلبة، العناصر الثقيلة، العناصر المشعة , *Zygophyllum siplex* *Haloxylon salicornicum*.